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Kola Project - International Report,  
Catchment Study 1994

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<p>Summary:</p> <p>During the period 1992-1996, the Geological Survey of Norway (NGU) and Finland (GTK) and the Central Kola Expedition (CKE) are carrying out a joint project on eco-geochemical mapping and monitoring within an area extending from longitudes 24° to 35.5° E with the Barents Sea as the northern project boundary and southward to the Arctic Circle in Finland and to the boundary between the Murmansk and Karelia regions in Russia.</p> <p>As a part of this project, a detailed study of eight catchment areas (three in Finland (Kirakka, Naruska, Pallas), one in Norway (Skjellbekken) and four in Russia (Zapoljarnij, Monchegorsk, Kirovsk, Kurka)) was carried out during 1994. The following sample media were taken for this catchment study: snowpack, rainwater, stream water, groundwater, organic stream sediment, moss, topsoil, podzol profiles, quaternary deposits and bedrocks. The report summarises, compares and interprets all analytical results obtained from the catchment study.</p>				
Keywords:	Geochemistry		Podzol	
Snow	Moss		Streamwater	
Topsoil	Groundwater		Heavy metals	

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## 1. INTRODUCTION

The project “Joint ecogeochemical mapping and monitoring in the scale of 1:1 million in the West Murmansk region and the contiguous areas of Finland and Norway” or, short, «Kola Ecogeochemistry» was initiated in 1991 as a co-operation between Central Kola Expedition (CKE), Russia, and the Geological Surveys of Finland (GTK) and Norway (NGU).

It was decided to propose a major geochemical mapping project (see World Wide Web site <http://www.ngu.no/Kola>) in a 188,000 km<sup>2</sup> area stretching from 24° to 35.5° E and from the Arctic Circle to the Barents Sea (Fig. 1.1).

Some of the world’s largest point sources of sulphur dioxide (SO<sub>2</sub>) emission are located within the study area (Gunn et al., 1995): the nickel smelter at Nikel, the ore roasting plant at Zapoljarnij and the nickel smelter at Monchegorsk (Fig. 1.1) together accounting for about 300,000 t of SO<sub>2</sub>, 1900 t of Ni, 1100 t of Cu and 94 t of V<sub>2</sub>O<sub>5</sub> emissions yearly (for a detailed table see Reimann et al., chapter 3.3, this report). Further emission sources include several coal fired central heating plants, large iron ore mines and mills, large scale apatite mining and processing, an aluminium smelter, a nuclear power plant and the town and harbour of Murmansk, being a major Russian naval base.

Field work started in 1992 in the form of a pilot project, mapping a 12,000 km<sup>2</sup> area in the border zone of the three countries, containing the iron ore mine and mill at Kirkenes, the nickel smelter at Nikel and the ore roasting plant at Zapoljarnij. The main purpose of the pilot project was to compare and harmonise field sampling methods, analytical facilities of all three organisations and data processing. Media sampled include snowpack, stream water, stream sediment, overbank sediment, terrestrial moss, O-horizon soil, C-horizon soil and topsoil (0-5cm). All media were analysed in at least two different laboratories using different techniques. Different extractions were tested as well as the use of different grain size fractions for the soil samples. Results of this pilot project are documented in Chekushin et al. (1993) and in a number of separate publications (Äyräs et al. in prep., Niskavaara et al., 1996, Reimann et al, 1995, 1996, in prep. and Väisänen et al., in prep.).

As second part of this project and specific subject of this report, eight catchments (hereafter abbreviated as C 1 - C 8) widely distributed over the survey area (Fig. 1.1) were investigated in detail in 1994. Media sampled were snow (meltwater and filter residue), rainwater, stream water, organic stream sediments, terrestrial moss, topsoil (0-5 cm), complete podzol profiles, Quaternary deposits, bedrock and, in some catchments, groundwater. In some of the Russian catchments, where mosses were missing due to severe environmental degradation, some other plants (lichens, crowberries) were tested for their use as biomonitors. The main aims of the catchment study can be summarised as follows:

- (1) to evaluate the levels of atmospheric heavy metal deposition at different distances from source,
- (2) to compare element contents and variability in the different media and between the studied catchments to ensure the representativity of results for the large scale regional mapping project and
- (3) to identify the peculiarities of element distribution in different media at different levels of pollution and with different geological settings and thus varying background levels.

Catchment selection was based on distance to pollution sources and the ocean (element input via sea spray) and to represent the different possible settings (geology, climatic zones, soil type, vegetation, Quaternary deposits, altitude) in the survey area. Four of the catchments are in Russia (C1, Zapoljarnij, C2, Monchegorsk, C3, Kirovsk and C4, Kurka), one is in Norway (C5, Skjellbekken) and three are in Finland (C6, Kirakka, C7, Naruska and C8, Pallas). The size of the catchments varies between 12 and 35 km<sup>2</sup>. Table 1.1 summarises the most important features of the different catchments. An introductory description of the geology and Quaternary deposits within the catchments, including maps, is given in Chapter 2 of this report. Sample locations within the catchments are documented in Figs. 1.2 to 1.9.

This report documents and summarises the results from the catchment study 1994. It is organised according to the different media sampled and each chapter is written in such a way that it can be read independently without any necessity or obligation to read the complete report or even other chapters. This approach results in a number of repetitions in terms of introductory remarks or descriptions of analytical methods. The conclusions (chapter 10) give a short summary of the most important results. In addition to the references accompanying each chapter a general reference chapter (chapter 11) tries to summarise information about all relevant published literature from the survey area of which we are aware. A diskette accompanying this report contains all the analytical data from the catchment study in Excel-format.

Readers interested in even more details from single catchments are referred to the three national reports from the catchment study (Äyräs, 1995, Reimann (ed.), 1995 and Chekushin et al, 1995).

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## TABLES:

Table. 1.1. Summary of some characteristics of the different catchments.

## FIGURES

Figure. 1.1: Location of the study area for regional mapping 1995 (outer frame) and of the pilot project area (inner frame). The eight studied catchments are shown as no. 1-8: 1: Zapoljarnij, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska and 8: Pallas. K:Kirkenes, N: Nikel, M: Monchegorsk, Mu: Murmansk and Z: Zapoljarnij.

Figure 1.2.: Outline of and sample locations in catchment 1, Zapoljarnij.

Figure. 1.3.: Outline of and sample locations in catchment 2: Monchegorsk.

Figure 1.4.: Outline of and sample locations in catchment 3: Kirovsk.

Figure. 1.5: Outline of and sample locations in catchment 4: Kurka.

Figure. 1.6: Outline of and sample locations in catchment 5: Skjellbekken.

Figure. 1.7.: Outline of and sample locations in catchment 6: Kirakka.

Figure 1.8.: Outline of and sample locations in catchment 7: Naruska.

Figure. 1.9.: Outline of and sample locations in catchment 8: Pallas.

No.	Name	Coordinates of catchment outlet	Size (km <sup>2</sup> )	Elevation (m a.s.l.)	Annual precip. (mm)*	Vegetation	Bedrock	Surface cover, peculiarities
RUSSIA								
C1	Zapoljarniy	69°27'01"N 31°03'49"E	19.02	25-373	454	birch forest tundra	gneiss	till, fluvioglacial, outcrop
C2	Monchegorsk	67°50'30"N 32°54'48"E	22.38	128-507	391	technogenic desert, birch shrubs	dacite & andesite & tuffs, gabbro/norite	till, prone to erosion
C3	Kirovsk	67°32'50"N 33°48'55"E	20.01	240-1075	502	spruce forest, mountain tundra birch forest	nephelinite	till, diluvial/eluvial
C4	Kurka	67°41'25"N 32°50'14"E	20.49	152-466	502	north taiga spruce forest, birch; incipient deterioration	amphibolite, gneiss	till, fluvio-glacial
NORWAY								
C5	Skjellbekken	69°21'25"N 29°27'25"E	34.56	80-297	422	north taiga pine forest, birch	andesite, basalt & tuffs, 'black shale'	till, esker
FINLAND								
C6	Kirakka	69°35'12"N 28°51'46"E	11.86	110-200	386	north taiga pine forest	granite	outcrop, till, moraine ridge
C7	Naruska	67°21'44"N 29°22'05"E	20.16	263-490	513	north taiga spruce forest	gneiss	till, peat, outcrop
C8	Pallas	68°09'14"N 23°52'50"E	24.42	303-500	405	north taiga spruce forest	quartzite	till, peat

\* from the closest meteorological station (data from 1994)

Table. 1.1. Summary of some characteristics of the different catchments.

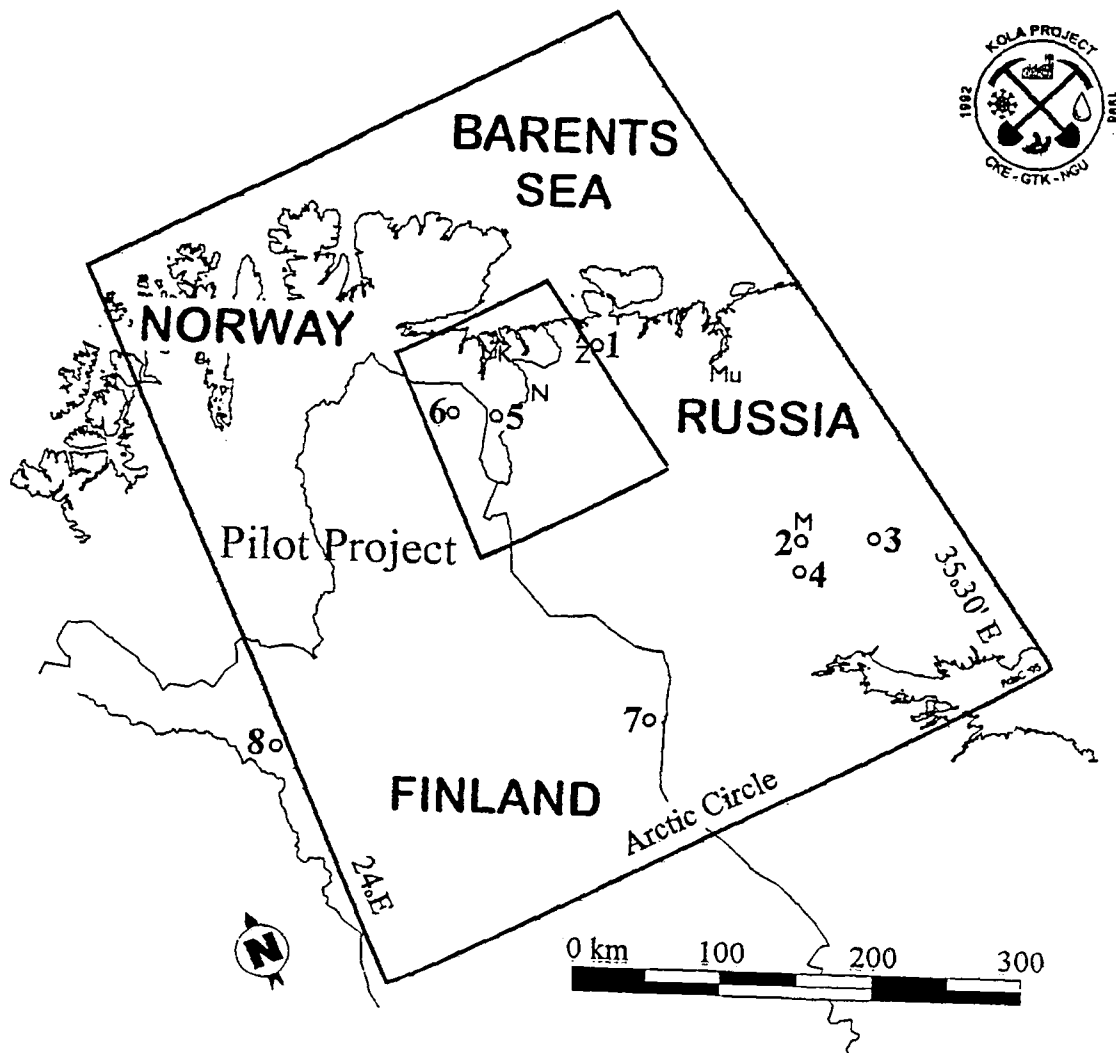
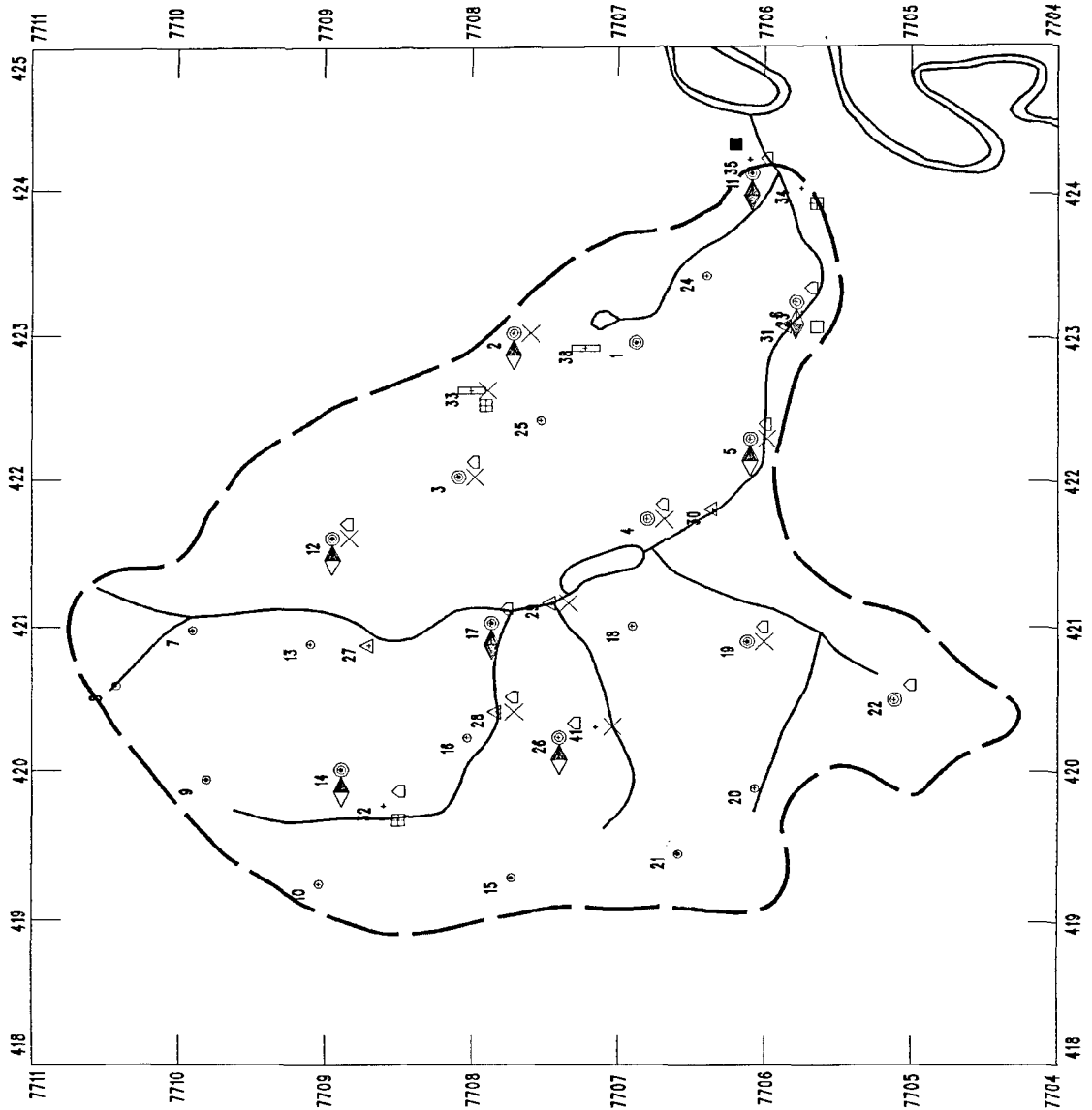


Figure. 1.1: Location of the study area for regional mapping 1995 (outer frame) and of the pilot project area (inner frame). The eight studied catchments are shown as no. 1-8: 1: Zapoljarnij, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska and 8: Pallas. K:Kirkenes, N: Nikel, M: Monchegorsk, Mu: Murmansk and Z: Zapoljarnij.

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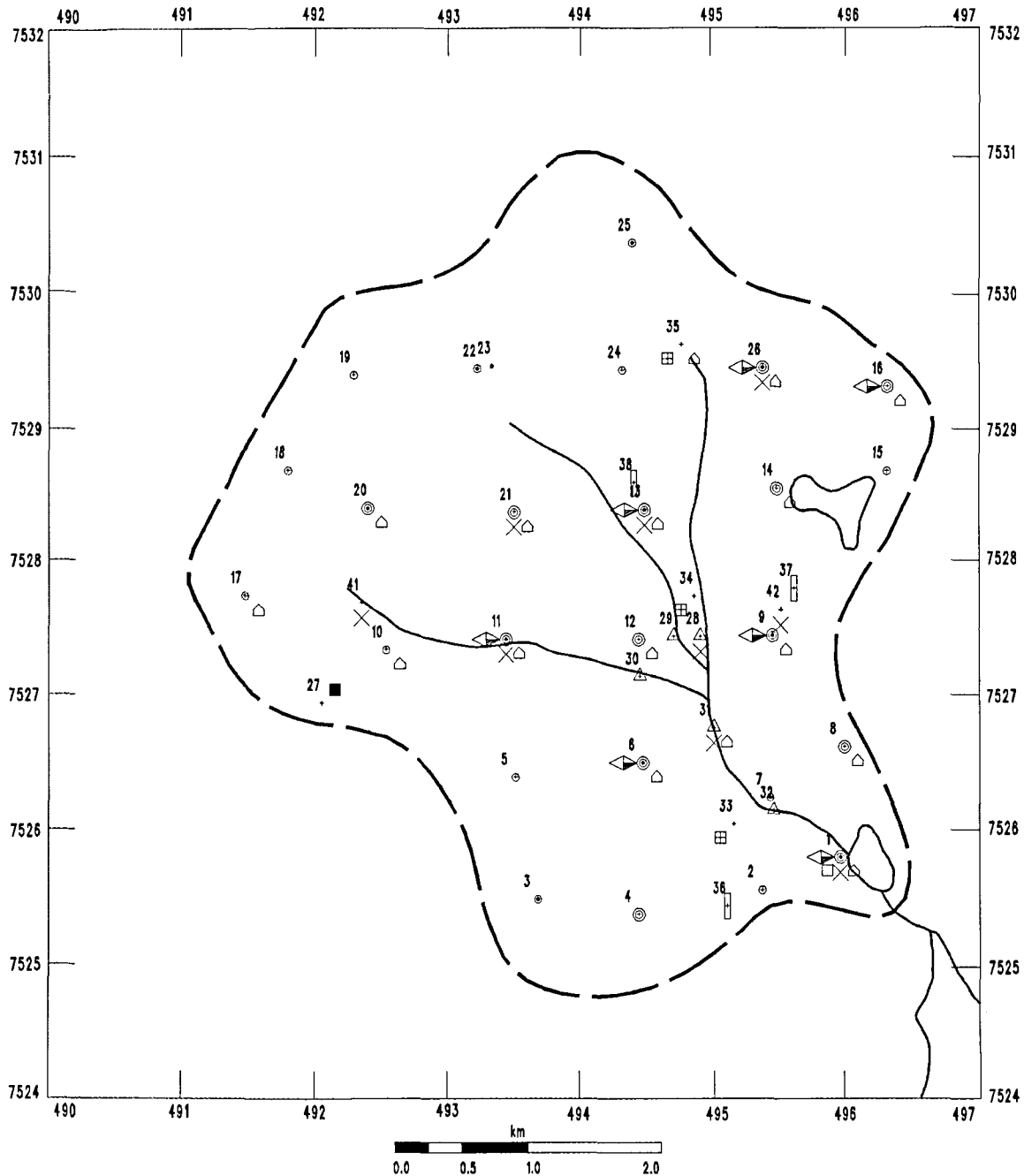


- Point of sampling
- Snow (sn)
- Topsoil(tp), 1st-2nd sampling
- Topsoil(tp), 3d-4th sampling
- Stream water (wa)
- ▣ Rain water (rw)
- Ground water (gw)
- △ Stream sediment (ss)
- ◇ Moss (vg, 1st digit)
- ◇ Lichen (vg, 2nd digit)
- ◇ Crowberry (vg, 3d digit)
- ◇ Soil profile (sp)
- Quaternary deposits (qd)
- × Bedrock (br)

No	br	gw	qd	rw	sn	ss	tp	vg	wa	sp
1							4	0	2	
2							4	0	2	
3							4	0	2	
4							4	0	2	
5							4	0	2	
6							4	2	0	
7							2			
8							2			
9							2			
10							2			
11							4	2	2	
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Fig 1.2.



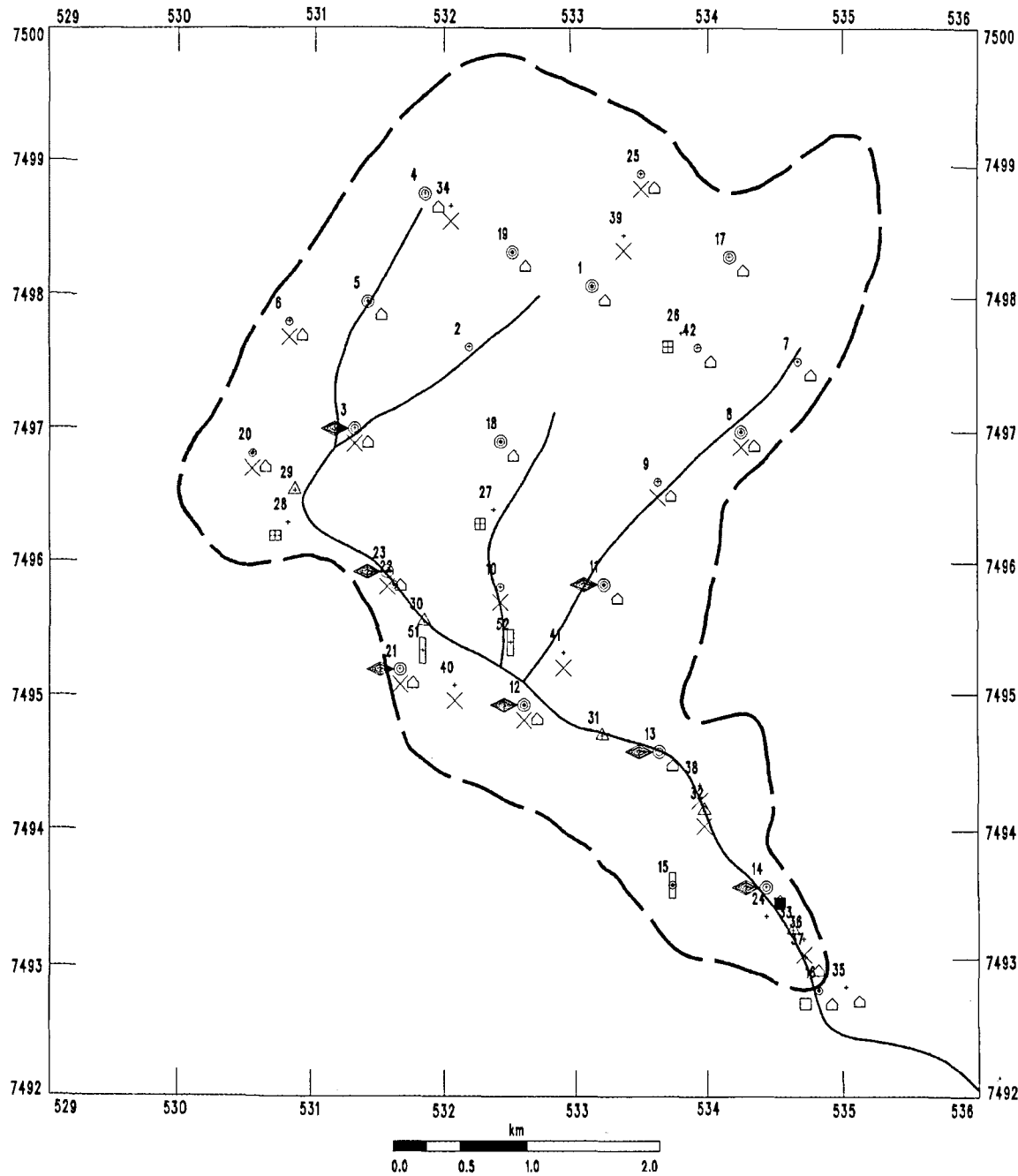


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- ▭ Soil profile (sp)
- △ Quaternary deposits (qd)
- × Bedrock (br)

No	br	gw	qd	rw	sn	ss	tp	vg	wa	sp		
1	1	-	1	-	2	-	4	002	32	-		
2	-	-	-	-	1	-	2	-	-	-		
3	-	-	-	-	-	-	2	-	-	-		
4	-	-	-	-	-	-	4	-	-	-		
5	-	-	-	-	-	-	2	-	-	-		
6	-	-	1	-	1	-	4	002	-	-		
7	-	-	-	-	-	-	2	-	-	-		
8	-	-	1	-	-	-	4	-	-	-		
9	-	-	1	-	1	-	4	002	-	-		
10	-	-	1	-	-	-	2	-	-	-		
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41	1	-	-	-	-	-	-	-	-	-		
42	1	-	-	-	-	-	-	-	-	-		
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Fig 1.3.

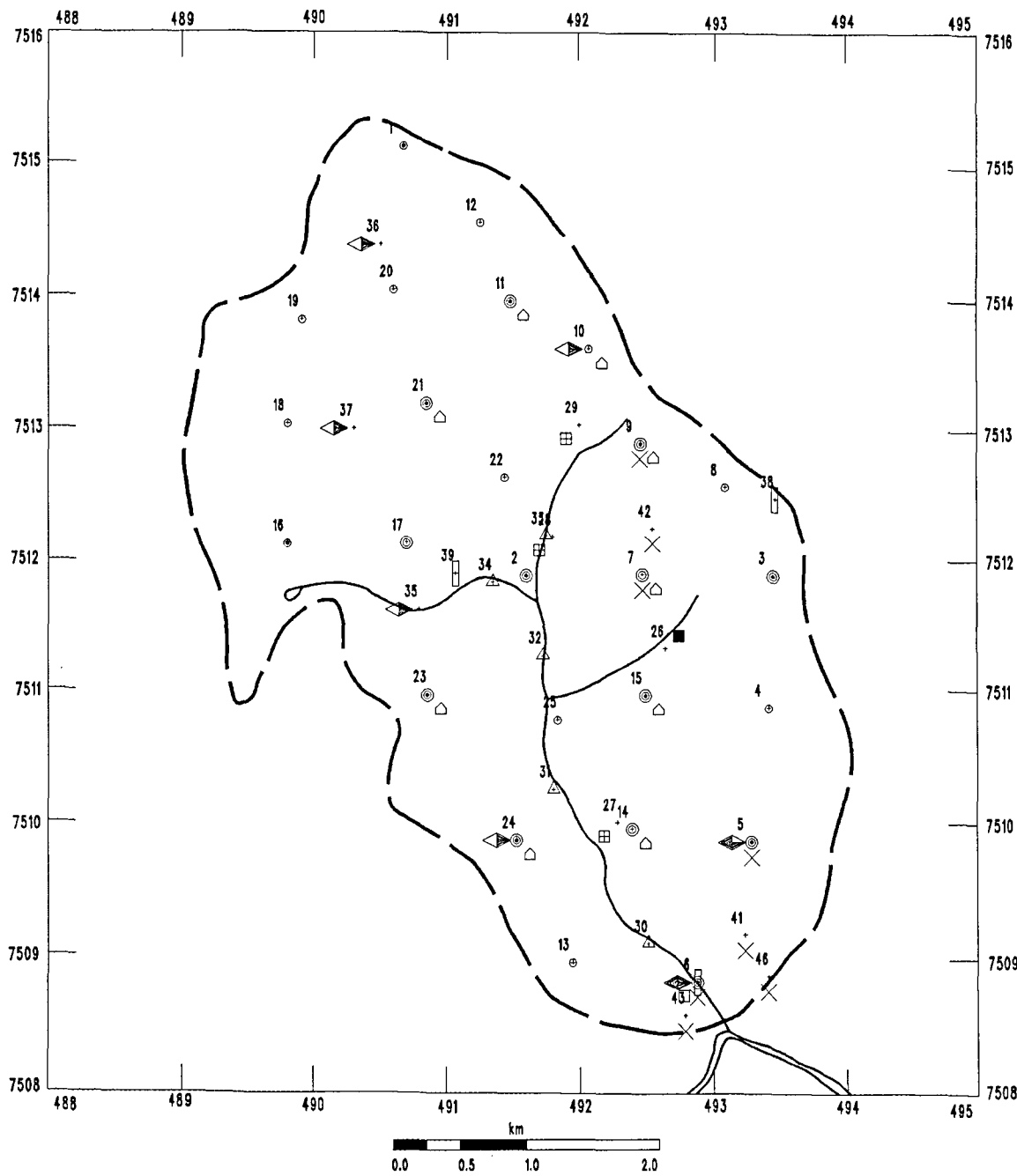


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- ◇ Crowberry (vg, 3d digit)
- ▭ Soil profile (sp)
- △ Quaternary deposits (qd)
- × Bedrock (br)

No	br	gw	qd	rw	sn	ss	tp	vg	wa	sp
1	-	-	1	-	1	-	4	-	-	-
2	-	-	-	-	-	-	2	-	-	-
3	1	-	1	-	-	-	4	222	-	-
4	-	-	1	-	-	-	4	-	-	-
5	-	-	-	-	1	-	2	-	-	-
6	1	-	1	-	-	-	4	-	-	-
7	-	-	-	-	-	-	2	-	-	-
8	1	-	1	-	1	-	4	-	-	-
9	1	-	1	-	-	-	2	-	-	-
10	1	-	-	-	-	-	2	-	-	-
11	-	-	1	-	1	-	4	222	-	-
12	1	-	-	-	1	-	4	222	-	-
13	-	-	1	-	-	-	4	222	-	-
14	-	-	1	-	-	-	4	222	-	-
15	-	-	-	-	1	-	2	-	-	8
16	-	-	1	-	1	-	3	-	7	-
17	-	-	1	-	-	-	2	-	-	-
18	-	-	1	-	2	-	4	-	-	-
19	-	-	1	-	1	-	3	-	-	-
20	1	-	-	-	1	-	2	-	-	-
21	1	-	1	-	-	-	4	222	-	-
22	-	-	-	-	1	-	2	-	-	-
23	1	-	1	-	1	-	4	222	-	-
24	-	8	-	-	-	-	-	-	-	-
25	1	-	1	-	-	-	1	-	-	-
26	-	-	-	5	-	-	-	-	-	-
27	-	-	-	5	-	-	-	-	-	-
28	-	-	-	-	-	-	-	-	-	-
29	-	-	-	-	-	3	-	-	-	-
30	-	-	-	-	-	3	-	-	-	-
31	-	-	-	-	-	3	-	-	-	-
32	1	-	-	-	-	3	-	-	-	-
33	-	-	-	-	-	3	-	-	-	-
34	1	-	-	-	-	-	-	-	-	-
35	-	1	-	-	-	-	-	-	-	-
36	1	-	-	-	-	-	-	-	-	-
37	-	-	1	-	-	-	-	-	-	-
38	1	-	-	-	-	-	-	-	-	-
39	1	-	-	-	-	-	-	-	-	-
40	1	-	-	-	-	-	-	-	-	-
41	1	-	-	-	-	-	-	-	-	-
42	-	-	1	-	-	-	1	-	-	-
51	-	-	-	-	-	-	-	-	-	7
52	-	-	-	-	-	-	-	-	-	7
	17	8	23	15	13	15	74	42	7	22

Fig 1.4.

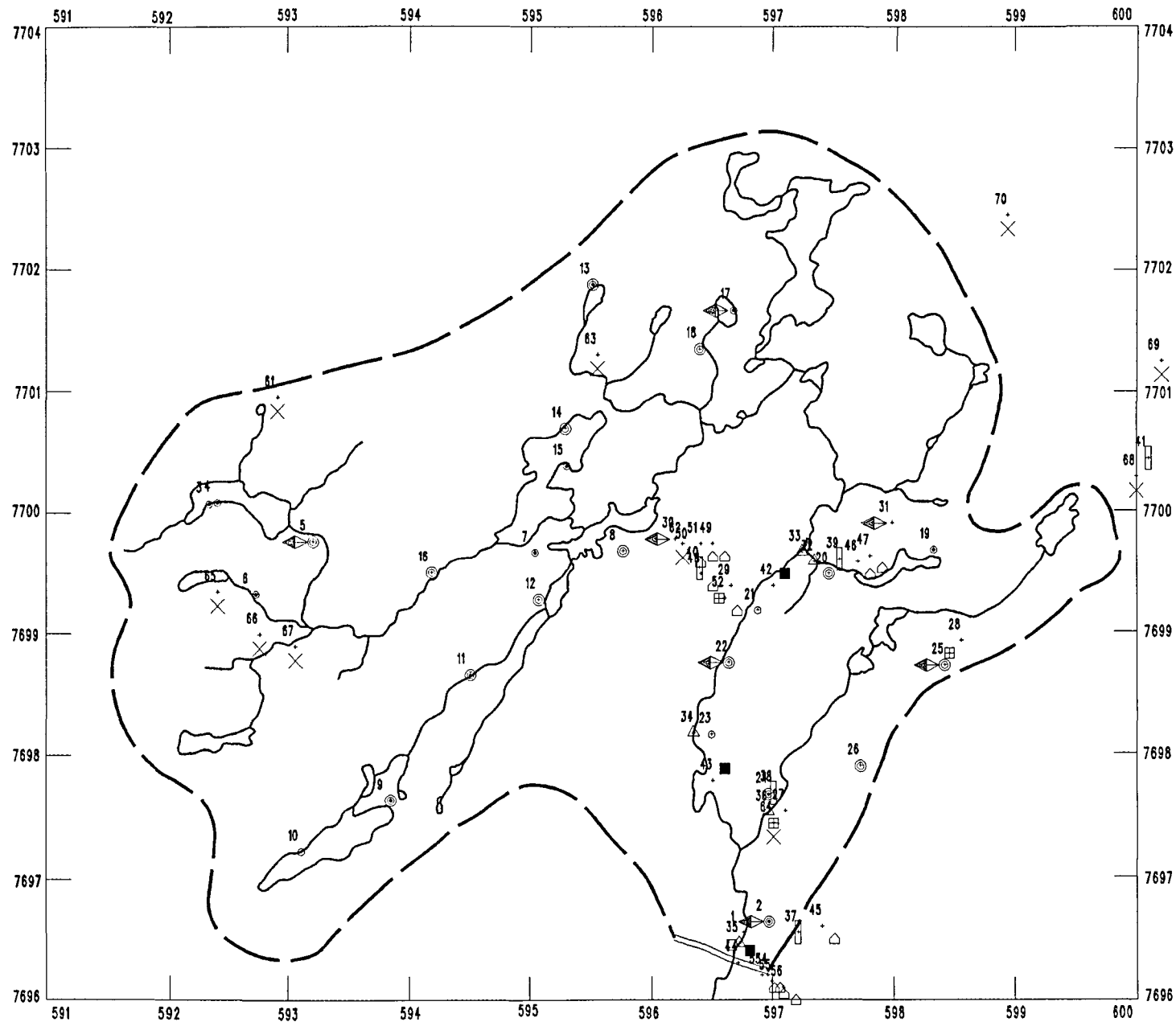


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- Point of sampling
- Snow (sn)
- Topsoil (tp), 1st-2nd sampling
- Topsoil (tp), 3d-4th sampling
- Stream water (wa)
- ▣ Rain water (rw)
- Ground water (gw)
- △ Stream sediment (ss)
- ◊ Moss (vg, 1st digit)
- ◊ Lichen (vg, 2nd digit)
- ◊ Crowberry (vg, 3d digit)
- ▭ Soil profile (sp)
- △ Quaternary deposits (qd)
- × Bedrock (br)

No	br	gw	qd	rw	sn	ss	tp	vg	wa	ap
1	-	-	-	-	1	-	2	-	-	-
2	-	-	-	-	1	-	4	-	-	-
3	-	-	-	-	1	-	4	-	-	-
4	-	-	-	-	1	-	2	-	-	-
5	1	-	-	-	1	-	4	202	-	-
6	2	-	-	-	2	-	4	222	8	7
7	1	-	1	-	-	-	4	-	-	-
8	-	-	-	-	-	-	2	-	-	-
9	1	-	1	-	1	-	4	-	-	-
10	-	-	1	-	-	-	2	022	-	-
11	-	-	1	-	1	-	4	-	-	-
12	-	-	1	-	-	-	2	-	-	-
13	-	-	1	-	-	-	2	-	-	-
14	-	-	1	-	-	-	4	-	-	-
15	-	-	1	-	1	-	4	-	-	-
16	-	-	-	-	1	-	2	-	-	-
17	-	-	-	-	-	-	4	-	-	-
18	-	-	-	-	-	-	2	-	-	-
19	-	-	-	-	-	-	2	-	-	-
20	-	-	-	-	-	-	2	-	-	-
21	-	-	1	-	1	-	4	-	-	-
22	-	-	1	-	-	-	2	-	-	-
23	-	-	1	-	1	-	4	-	-	-
24	-	-	1	-	1	-	4	022	-	-
25	-	-	-	-	-	-	2	-	-	-
26	-	7	-	-	-	-	-	-	-	-
27	-	-	-	5	-	-	-	-	-	-
28	-	-	-	5	-	-	-	-	-	-
29	-	-	-	5	-	-	-	-	-	-
30	-	-	-	-	-	3	-	-	-	-
31	-	-	-	-	-	3	-	-	-	-
32	-	-	-	-	-	3	-	-	-	-
33	-	-	-	-	-	3	-	-	-	-
34	-	-	-	-	-	3	-	-	-	-
35	-	-	-	-	-	-	-	022	-	-
36	-	-	-	-	-	-	-	022	-	-
37	-	-	-	-	-	-	-	022	-	-
38	-	-	-	-	-	-	-	-	7	-
39	-	-	-	-	-	-	-	-	8	-
41	1	-	-	-	-	-	-	-	-	-
42	1	-	-	-	-	-	-	-	-	-
43	1	-	-	-	-	-	-	-	-	-
46	1	-	-	-	-	-	-	-	-	-
	8	7	9	16	13	15	80	30	8	22

Fig 1.5.

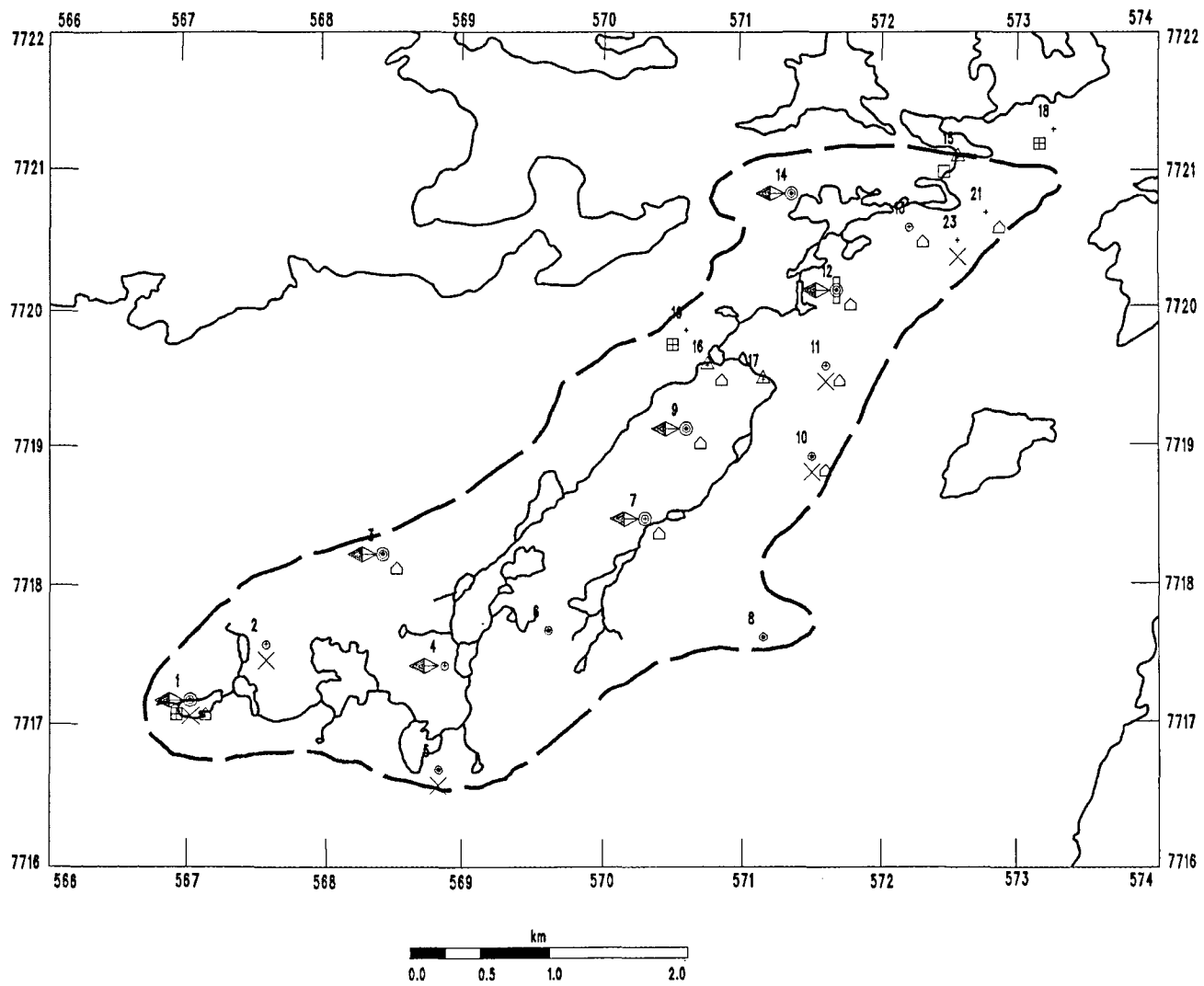


- Point of sampling
- Snow (sn)
- Topsoil (tp), 1st-2nd sampling
- Topsoil (tp), 3d-4th sampling
- Stream water (wa)
- ▣ Rain water (rw)
- Ground water (gw)
- △ Stream sediment (ss)
- ◊ Moss (vg, 1st digit)
- ◊ Lichen (vg, 2nd digit)
- ◊ Crowberry (vg, 3d digit)
- ▭ Soil profile (sp)
- ◊ Quaternary deposits (qd)
- × Bedrock (br)

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No	br	gw	qd	rw	sn	ss	tp	vg	wa	sp
1	-	-	-	-	-	-	-	-	-	37
2	-	-	-	-	-	-	4	200	-	-
3	-	-	-	-	-	-	2	-	-	-
4	-	-	-	-	-	-	2	-	-	-
5	-	-	-	-	-	-	4	200	-	-
6	-	-	-	-	-	-	2	-	-	-
7	-	-	-	-	-	-	2	-	-	-
8	-	-	-	-	-	-	3	-	-	-
9	-	-	-	-	-	-	4	-	-	-
10	-	-	-	-	-	-	2	-	-	-
11	-	-	-	-	-	-	4	-	-	-
12	-	-	-	-	-	-	3	-	-	-
13	-	-	-	-	-	-	4	-	-	-
14	-	-	-	-	-	-	4	-	-	-
15	-	-	-	-	-	-	2	-	-	-
16	-	-	-	-	-	-	4	-	-	-
17	-	-	-	-	-	-	2	200	-	-
18	-	-	-	-	-	-	4	-	-	-
19	-	-	-	-	-	-	1	-	-	-
20	-	-	-	-	-	-	3	-	-	-
21	-	-	-	-	-	-	2	-	-	-
22	-	-	-	-	-	-	4	200	-	-
23	-	-	-	-	-	-	2	-	-	-
24	-	-	-	-	-	-	3	-	-	-
25	-	-	-	-	-	-	4	200	-	-
26	-	-	-	-	-	-	3	-	-	-
27	-	-	-	-	-	-	5	-	-	-
28	-	-	-	-	-	-	5	-	-	-
29	-	-	-	-	-	-	5	-	-	-
30	-	-	-	-	-	-	-	200	-	-
31	-	-	-	-	-	-	-	200	-	-
32	-	-	-	-	-	-	3	-	-	-
33	-	-	-	-	-	-	3	-	-	-
34	-	-	-	-	-	-	3	-	-	-
35	-	-	-	-	-	-	3	-	-	-
36	-	-	-	-	-	-	3	-	-	-
37	-	-	-	-	-	-	3	-	-	11
38	-	-	-	-	-	-	3	-	-	8
39	-	-	-	-	-	-	3	-	-	7
40	-	-	-	-	-	-	3	-	-	9
41	-	-	-	-	-	-	3	-	-	7
42	-	57	-	-	-	-	-	-	-	7
43	-	49	-	-	-	-	-	-	-	7
44	-	51	-	-	-	-	-	-	-	7
45	-	3	-	-	-	-	-	-	-	7
46	-	3	-	-	-	-	-	-	-	7
47	-	3	-	-	-	-	-	-	-	7
48	-	8	-	-	-	-	-	-	-	7
49	-	3	-	-	-	-	-	-	-	7
50	-	4	-	-	-	-	-	-	-	7
51	-	4	-	-	-	-	-	-	-	7
52	-	3	-	-	-	-	-	-	-	7
53	-	2	-	-	-	-	-	-	-	7
54	-	1	-	-	-	-	-	-	-	7
55	-	2	-	-	-	-	-	-	-	7
56	-	1	-	-	-	-	-	-	-	7
57	1	-	-	-	-	-	-	-	-	7
58	1	-	-	-	-	-	-	-	-	7
59	1	-	-	-	-	-	-	-	-	7
60	1	-	-	-	-	-	-	-	-	7
61	1	-	-	-	-	-	-	-	-	7
62	3	-	-	-	-	-	-	-	-	7
63	1	-	-	-	-	-	-	-	-	7
64	2	-	-	-	-	-	-	-	-	7
65	1	-	-	-	-	-	-	-	-	7
66	1	-	-	-	-	-	-	-	-	7
67	1	-	-	-	-	-	-	-	-	7
68	1	-	-	-	-	-	-	-	-	7
69	1	-	-	-	-	-	-	-	-	7
70	5	-	-	-	-	-	-	-	-	7
	17	157	35	15	12	15	75	14	37	42

Fig 1.6.



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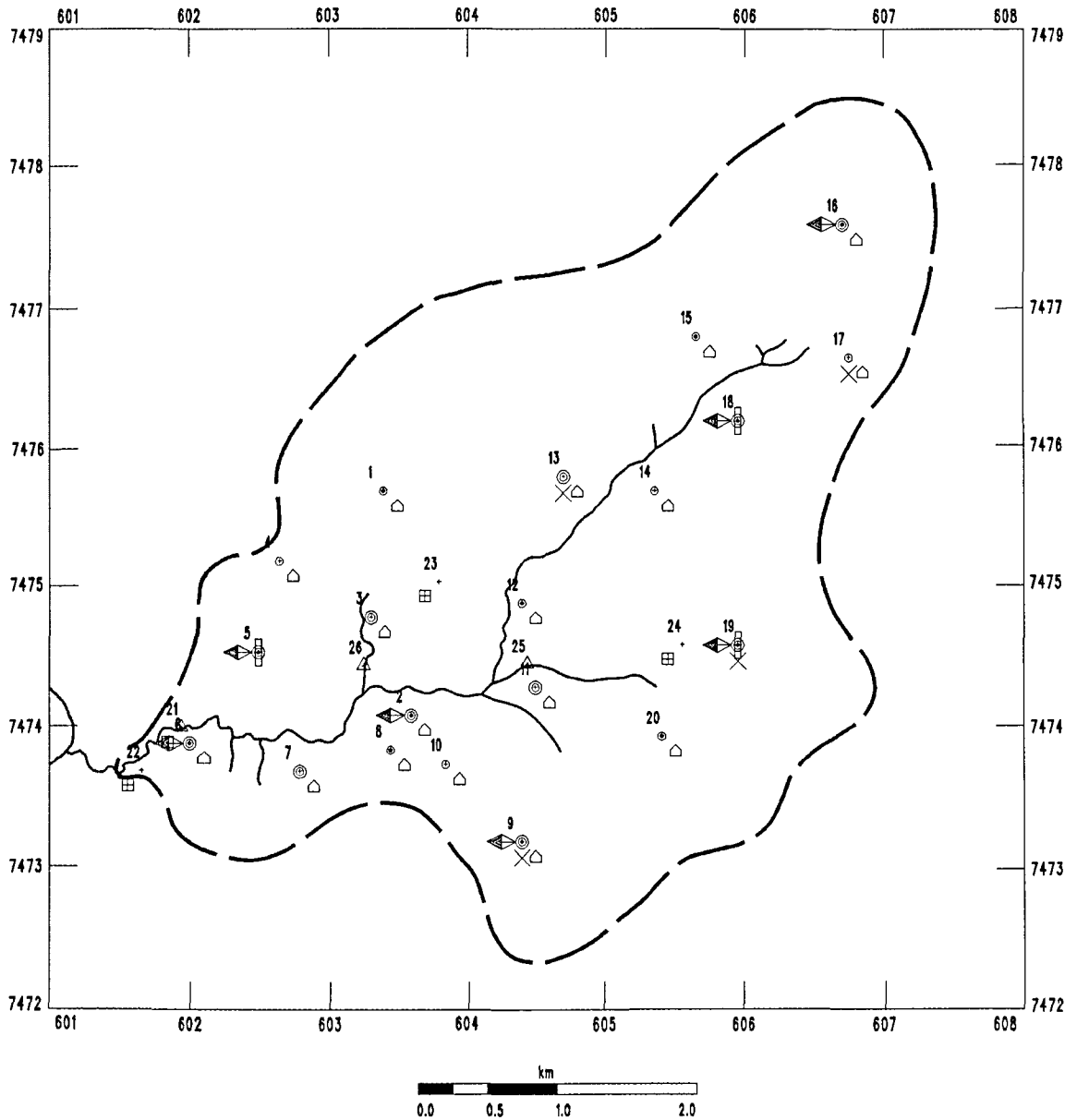
- Point of sampling
- Snow (sn)
- Topsoil (tp), 1st-2nd sampling
- Topsoil (tp), 3d-4th sampling
- Stream water (wa)
- ▣ Rain water (rw)
- Ground water (gw)

- △ Stream sediment (ss)
- ◊ Moss (vg, 1st digit)
- ◊ Lichen (vg, 2nd digit)
- ◊ Strawberry (vg, 3d digit)
- ▭ Soil profile (sp)
- △ Quaternary deposits (qd)
- × Bedrock (br)

No	br	gw	qd	rw	sn	ss	tp	vg	wa	sp
1	1	-	1	3	1	-	4	200	-	-
2	1	-	-	-	-	-	2	-	-	-
3	-	-	1	-	1	-	4	200	-	-
4	-	-	-	-	-	-	2	200	-	-
5	1	-	-	-	1	-	2	-	-	-
6	-	-	-	-	1	-	2	-	-	-
7	-	-	1	-	-	-	4	200	-	-
8	-	-	-	-	1	-	2	-	-	-
9	-	-	1	-	1	-	4	200	-	-
10	1	-	1	-	1	-	2	-	-	-
11	1	-	1	-	-	-	2	-	-	-
12	-	-	1	-	1	-	3	200	-	8
13	-	-	1	-	-	-	2	-	-	-
14	-	-	-	-	1	-	4	200	-	-
15	-	-	-	-	-	3	-	-	5	-
16	-	-	1	-	-	3	-	-	-	-
17	-	-	-	3	-	3	-	-	-	-
18	-	-	-	3	-	-	-	-	-	-
19	-	-	-	3	-	-	-	-	-	-
21	-	-	1	-	-	-	-	-	-	-
23	1	-	-	-	-	-	-	-	-	-
6 0 10 9 9 9 18 14 5 8										

Fig 1.7.

ECOGEOCHEMISTRY KOLA  
 joint CKE, GTK, NGU project  
 Catchment Study



- Point of sampling
- Snow (sn)
- Topsoil (tp), 1st-2nd sampling
- Topsoil (tp), 3d-4th sampling
- Stream water (wa)
- ▣ Rain water (rw)
- Ground water (gw)
- △ Stream sediments (ss)
- ◊ Moss (vg, 1st digit)
- ◊ Lichen (vg, 2nd digit)
- ◊ Crowberry (vg, 3d digit)
- Soil profile (sp)
- △ Quaternary deposits (qd)
- × Bedrock (br)

No	br	gw	qd	rw	sn	ss	tp	vg	wa	sp
1	-	-	1	-	1	-	2	-	-	-
2	-	-	1	-	1	-	4	200	-	-
3	-	-	1	-	-	-	4	-	-	-
4	-	-	1	-	-	-	2	-	-	-
5	-	-	-	-	1	-	4	200	-	9
6	-	-	1	-	1	-	4	200	-	-
7	-	-	1	-	-	-	4	-	-	-
8	-	-	1	-	1	-	2	-	-	-
9	1	-	1	-	1	-	4	200	-	-
10	-	-	1	-	-	-	2	-	-	-
11	-	-	1	-	-	-	4	-	-	-
12	-	-	1	-	1	-	2	-	-	-
13	1	-	1	-	-	-	4	-	-	-
14	-	-	1	-	-	-	2	-	-	-
15	-	-	1	-	1	-	2	-	-	-
16	-	-	1	-	1	-	4	200	-	-
17	1	-	1	-	-	-	2	-	-	-
18	-	-	-	-	1	-	4	200	-	11
19	1	-	1	-	1	-	4	200	-	9
20	-	-	1	-	1	-	2	-	-	-
21	-	-	-	-	-	3	-	-	8	-
22	-	-	-	-	5	-	-	-	-	-
23	-	-	-	-	4	-	-	-	-	-
24	-	-	-	-	5	-	-	-	-	-
25	-	-	-	-	-	3	-	-	-	-
26	-	-	-	-	-	3	-	-	-	-
4 0 17 14 12 9 62 14 8 29										

Fig 1.8.

ECOGEOCHEMISTRY KOLA  
 joint CKE, GTK, NGU project  
 Catchment Study



- Point of sampling
- Snow (sn)
- Topsoil (tp), 1st-2nd sampling
- Topsoil (tp), 3d-4th sampling
- Stream water (wa)
- ▣ Rain water (rw)
- Ground water (gw)
- △ Stream sediment (ss)
- ◀ Moss (vg, 1st digit)
- ◀ Lichen (vg, 2nd digit)
- ◀ Strawberry (vg, 3d digit)
- ▭ Soil profile (sp)
- △ Quaternary deposits (qd)
- × Bedrock (br)

No	br	gw	qd	rw	sn	ss	tp	vg	wa	sp
1	-	-	1	-	-	-	2	-	-	-
2	-	-	1	-	-	-	3	200	-	-
3	1	-	1	-	1	-	4	-	-	-
4	-	-	1	-	1	-	3	200	-	-
5	1	-	1	-	1	-	4	-	-	-
6	-	-	-	-	-	-	3	-	-	-
7	-	-	2	-	1	-	2	200	-	-
8	1	-	1	-	-	-	2	200	-	-
9	-	-	1	-	-	-	2	-	-	-
10	-	-	1	-	1	-	4	-	-	-
11	-	-	1	-	1	-	4	-	-	-
12	1	-	1	-	-	-	2	-	-	-
13	-	-	1	-	-	-	4	200	-	8
14	-	-	1	-	-	-	2	-	-	-
15	-	-	-	-	1	-	4	-	-	10
16	1	-	-	-	-	-	3	-	-	-
17	-	-	-	-	1	-	2	-	-	-
18	-	-	-	-	-	-	4	200	-	10
19	-	-	1	-	1	-	2	200	-	-
20	-	-	1	-	1	-	4	-	-	-
21	-	-	-	4	-	3	-	-	30	-
22	-	-	-	4	-	-	-	-	-	-
23	-	-	-	4	-	3	-	-	-	-
24	-	-	-	-	3	-	-	-	-	-
25	-	-	-	4	-	-	-	-	-	-
	5	0	15	12	11	9	60	14	30	28

Fig 1.9.

## 2. BEDROCK AND QUATERNARY DEPOSITS

### 2.1 Composition of bedrock and Quaternary deposits in eight catchments subjected to detailed ecogeochemical investigation in the Barents Region

*V.A. Pavlov, M.-L. Räsänen, V. Melezhik and P. de Caritat*

#### INTRODUCTION

As part of the catchment studies phase of the Kola Ecogeochemistry project (1994), bedrock and Quaternary deposits samples were collected and analysed. The chosen catchments are located in northwestern Russia (C1 to C4), northeastern Norway (C5) and northern Finland (C6 to C8), as shown on Figure 2.1.1. This chapter gives a brief description of the geology (bedrock and overburden) of the eight catchments, outlines the sampling methods, and presents the analytical results obtained for bedrock and Quaternary deposits.

#### LITHOLOGY AND OVERBURDEN

The nature and distribution of the lithology (rock types) and Quaternary deposits (surface cover) in the eight catchments is briefly described hereafter and illustrated in the maps of Figures 2.1.2 and 2.1.3.

##### *Zapoljarny (C1)*

C1 is located 10 km northeast of the Zapoljarny Ni-Cu ore roasting plant. The predominant bedrock types are amphibole-biotite gneiss and granite-gneiss (81% of the area), and microcline, plagioclase-microcline granite (17%); two small dikes of pyroxenite (not shown on map) are also present here (Panichev 1995).

The Quaternary deposits consist mainly of basal till (60% of the area) and meltout till (26%) (Panichev 1995). Eluvial sediments (7%) with blocks, break stones and nonrounded fragments of bedrocks are present in the northern part of the catchment.

##### *Monchegorsk (C2)*

C2 is located 5 km south of the "Severonickel" smelter in Monchegorsk. The main rock types present are gabbro, gabbro-norite and norite (40%), andesitic lava and tuffs (32%), and pyroxenite (16%) (Zaizevsky 1974).

The Quaternary overburden is mainly basal till (76%), eluvial (9%), glaciofluvial (9%) and glaciolacustrine (4%) deposits (Zaizevsky 1974).

##### *Kirovsk (C3)*

C3 is situated on the southern slopes of the Khibiny mountains at a distance of 3 km southeast from Kirovsk town. The main types of bedrock are trachytoid (47%), massive hibinite (31%), gabbro-norite (9%), basalt and andesite (6%), and black schist and mafic tuff (2%) (Snyatkova 1983).

Basal till (57%) is the dominant type of overburden, followed by deluvial-eluvial (41%) deposits (Snyatkova 1983).



#### *Kurka (C4)*

C4 is located east of Imandra Lake, 25 km southwest of the Monchegorsk smelting industry. The predominant types of bedrock in the catchment are amphibolite (32%), two-mica gneiss (28%), gabbro-norite (22%) and gabbro-amphibolite (17%) (Zaizevsky 1974).

Basal till is the most widespread type of overburden (54%), followed by peat (30%), glaciofluvial (6%) and lacustrine (5%) deposits (Zaizevsky 1974).

#### *Skjellbekken (C5)*

C5 is located 31 km southwest of the Nickel smelter. The main types of bedrock are andesite and andesitic volcanoclastic schist (40%), tholeiitic basalt and tuff (30%), black schist (20%), and ultramafic and mafic intrusion (5%); carbonate rocks associated with quartzite, occur sporadically (Melezhik et al. 1995).

The Quaternary overburden is represented mainly by a thick cover of basal till (59%), glaciofluvial deposits (17%), meltout till (12%) and rock terrain (5%) (Olsen 1995).

#### *Kirakka (C6)*

C6 is located 57 km west-northwest of the Nickel smelter. The bedrock types are predominantly potassium granite (Meriläinen 1976).

Rock terrain and block fields cover 60% of the catchment area, followed by meltout till (34%) (Niemelä et al. 1993).

#### *Naruska (C7)*

C7 is located 65 km northwest of the Kovdor apatite-iron mine and about 100 km northeast of the Kemijärvi chemical pulp mill. Bedrock is predominantly tonalitic gneiss, with mica gneiss and amphibolite inclusions (H. Juopperi, pers. comm. 1995).

The overburden is mainly basal till (63%), peat-covered river valleys (23%), and rock terrain and block fields (14%) (Niemelä et al. 1993).

#### *Pallas (C8)*

C8 is remote from all major human activity centres in the study area. Bedrock consists mainly of quartzite (90%); amphibolite dikes (not shown on map) occur as well (Mikkola 1941).

The Quaternary overburden is mainly basal till (69%), with smaller occurrences of peat-covered river valleys (29%) and rock terrain (2%) (Niemelä et al. 1993).

#### *Grouping of catchments*

Lithologically, the studied catchments fall into one of the following three groups:

Group 1: those with a predominantly mafic to intermediate volcanic-sedimentary and plutonic rocks: C2, C4 and C5,

Group 2: those with relatively homogeneous, felsic rocks: C1, C6, C7 and C8, and

Group 3: one with alkaline rocks from the nepheline-syenite group: C3.

## SAMPLING

Bedrock samples were drilled out of outcrops of all representative rock types, and, in the Russian catchments, taken with a hammer. The most representative types of Quaternary deposits were recovered from dug pits, and, in the Finnish catchments, from percussion drilled holes. The till samples were taken from the C-horizon, usually at a depth of 0.8-1 m; in C5, three different till layers (A, B and C) were sampled as well. The number and type of samples of bedrock and Quaternary deposits collected in every catchment is given in Tables 2.1.1 and 2.1.2. More information on the sampling methods is given in Äyräs and Reimann (1995).

## SAMPLE PREPARATION AND ANALYSIS

### *Bedrock*

Sample preparation, consisting in the digestion of a 2 g subsample of crushed and milled bedrock for 2 hours at 90°C in 12 ml aqua regia (1:3 nitric acid/hydrochloric acid, v/v) followed by dilution to 60 ml, was performed at NGU. After mixing and centrifugation, the clear solution was analysed at GTK by ICP-AES for 26 elements, by graphite furnace AAS for Ag, As, Cd, Pb, Bi, Sb, Se and Te, and by cold vapour AAS for Hg (Niskavaara 1995). Determination of major mineral forming oxides was made by XRF at the NGU Laboratory.

### *Quaternary deposits*

The Quaternary deposits samples were prepared at GTK, where they were dried at 40°C, split, and one subsample was sieved to the fraction <0.064 mm (for the till samples, an additional subsample was sieved to the fraction <2 mm). A 300 mg subsample was digested for 2 hours at 90°C in 3 ml aqua regia followed by dilution to 15 ml. After mixing and centrifugation, the clear solution was analysed at GTK by ICP-AES for 25 elements (Niskavaara 1995). Determination of major mineral forming oxides was made by XRF at the NGU Laboratory.

Analytical methods are described more fully in Reimann (1995). The GTK and NGU Laboratories are accredited according to European Norm standard EN45001.

*Important remark:* the aqua regia extraction of bedrock and Quaternary deposits yields compositional results that **do not reflect total content**; aqua regia selectively leaches certain minerals, while leaving others mostly unaffected. The choice of this method, in the context of this ecogeochemical study, is to get results that are consistent and comparable with what has become a *de facto* standard in the environmental community. Total element contents are available for the major rock-forming oxides in the form of the XRF results.

## RESULTS

Results of the chemical analysis by ICP-AES and AAS are given for bedrock in Table 2.1.1, and for Quaternary deposits in Table 2.1.2. Results from the XRF analysis are given for bedrock in Table 2.1.3, and for Quaternary deposits in Table 2.1.4.

## DISCUSSION

### *Bedrock*

The bedrocks from the three groups of catchments previously identified are characterised by typical associations of major and minor elements, as shown by the composition of the aqua regia leachates. The *general* trends (Fig. 2.1.4) are described below.

Group 1: Relatively high abundances of Fe, Al, Mg, Ca, Mn, S, Sr, Ni, Co, Cr, Cu, Y, Sc, Se and Cd. Note that the black schists (rock type 5a) from C5 have extremely elevated contents of some of these elements, as well as of Mo, Ag, Sb and Te.

Group 2: Relatively low contents of most elements. Generally, pyroxenites (1a) of C1 have the highest content of many elements in this group, whereas the quartzites (8a) of C8 have the lowest.

Group 3: Relatively high concentrations of Al, Na, S, Sr, Y, As, Se, Cd and Sb, and low concentration of Li. The hibinites (3a and 3b) are in addition relatively rich in K, Ba, La, Th, Pb and Mo, and poor in Fe, Mg, V, Ni, Co, Cr, Cu, B and Sc.

### *Quaternary deposits*

The composition of the tills (Fig. 2.1.5) is relatively homogeneous. As expected, the fine fraction of the tills is enriched in nearly all elements relative to the coarse fraction. This is generally true especially (ratio of fine/coarse >2 in at least 6 of the 8 catchments) for S, Pb and As. In C1, nearly every element analysed for is more than twice more abundant in the fine than in the coarse fraction of till. In C1, C2 and C4, the composition of the Quaternary deposits is governed not only by geology, but also by technogenic deposition.

Quaternary deposits other than till can have chemical compositions that are significantly different from either of the two fractions from the till. For instance, in C3, Al and Sr contents in weathered rock and alluvial sediments are much higher than in till. These differences must be taken into account when interpreting the impact of Quaternary deposit composition on other media, e.g. stream water.

### *Bedrock vs. Quaternary deposits*

Figure 2.1.6 shows the composition of bedrock compared to Quaternary deposits in each catchment for the major oxides (XRF data). All bedrock and Quaternary deposits samples were pooled together here, regardless of type. Discrepancies in major oxide composition can be discerned, and may be useful in interpreting the genetic history of Quaternary deposits, for instance. Table 2.1.5 summarises the main geochemical features of bedrock and Quaternary deposits in the eight catchments.

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## TABLES:

Tab. 2.1.1 Mean composition (ICP-AES) of aqua regia leached bedrock samples (mg/kg)

Tab. 2.1.2 Mean composition (ICP-AES) of aqua regia leached Quaternary deposits (mg/kg)

Tab. 2.1.3 Mean total composition (XRF) of bedrock samples (%)

Tab. 2.1.4 Mean total composition (XRF) of Quaternary deposit samples (%)

Tab. 2.1.5 Overview of the bedrock and Quaternary deposits characteristics, and their geochemical peculiarities

## FIGURES

Figure 2.1.1. Location map of catchments in Russia (C1: Zapoljarnij, C2: Monchegorsk, C3: Kirovsk and C4: Kurka), Norway (C5: Skjellbekken) and Finland (C6: Kirakka, C7: Naruska and C8: Pallas). K: Kirovsk, Ki: Kirkenes (open-pit iron mine and mill), M: Monchegorsk (nickel-copper smelter, cobalt smelter, etc.), Mu: Murmansk, N: Nikel (nickel-copper smelter), Z: Zapoljarnij (nickel-copper ore roasting plant).

Figure 2.1.2. Lithological maps of C1-C8.

Figure 2.1.3. Maps of Quaternary deposits of C1-C8.

Figure 2.1.4. Composition (in mg/kg) of bedrocks in C1-C8 (aqua regia extractable fraction). Elements are ranked in order of decreasing abundance in C7 (rock type 7a) from left to right. The grey line represents the trend in C7, and allows easier comparison with other catchments.

Figure 2.1.5. Composition (in mg/kg) of fine and coarse fractions of till in C1-C8 (aqua regia extractable fraction). Elements are ranked in order of decreasing abundance in C7 (fine fraction) from left to right. The grey line represents the trend in C7, and allows easier comparison with other catchments.

Figure 2.1.6. XY-diagram of Quaternary deposits vs. bedrock composition (in %) in each catchment for major oxides (XRF data).

Tab. 2.1.1 Mean composition (ICP-AES) of aqua regia leached bedrock samples (mg/kg)

Rock type	C1			C2			C3					C4				C5					C6	C7	C8
	1a Plagio- cline micro- cline granite	1b Pyrox- ene nite	1c Amphi- bole biotite granite & gneiss	2a Gabbro	2b Gabbro- norite	2c Dacite, andesite	3a Trachy- toid libinitite	3b Massive hibinitite	3c Gabbro	3d Diabase, andesite	3e Black schist	4a Gabbro- norite	4b Ultra- mic fic rock	4c Two-mica gneiss	4d Am- phibo- lite	5a Black schist	5b Limestone	5c Ande- sitic volcano- clastic sedi- ment basalt	5d Thole- ite andes- itic	5e Andesite siltstone	6a Grani- te	7a Tona- litic gneiss	8a Quartz- ite
n=	2	2	8	5	1	3	5	6	3	2	1	2	2	5	8	7	2	2	3	2	6	4	5
Ag	0.04	0.18	0.04	0.04	0.12	0.02	0.025	0.025	0.05	0.065	0.08	0.02	0.013	0.02	0.02	0.44	0.033	0.065	0.03	0.035	0.04	0.045	0.1
Al	4290	16400	5110	20700	27600	17900	70700	63500	21100	33900	37000	20900	31300	17700	20500	18000	6450	26100	18200	26900	7150	9210	5870
As	0.2	0.23	0.075	0.05	0.3	0.1	0.8	1.25	1.1	19.9	6.9	0.13	0.05	0.05	0.05	0.2	1.45	2.7	0.05	0.15	0.05	0.1	0.1
B	1.5	2.25	1.5	1.5	5	3	1.5	1.5	1.5	4	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	2.75	3
Ba	19.2	59.9	20.8	45.3	488	124	216	236.5	19.8	23.7	13.5	40.7	33.3	178	241	82.4	23.7	59.2	6.1	232.5	59.7	95.4	30.2
Bi	0.013	0.02	0.005	0.01	0.02	0.02	0.01	0.008	0.01	0.005	0.02	0.008	0.005	0.01	0.015	0.14	0.023	0.1	0.01	0.175	0.01	0.02	0.02
Ca	1240	19700	1230	13900	6040	9200	2120	2080	10600	19900	25800	15400	2250	13300	3840	5900	238500	25800	15200	3760	3770	3630	870
Cd	0.03	0.08	0.03	0.06	0.02	0.05	0.02	0.025	0.06	0.07	0.09	0.025	0.013	0.05	0.02	0.15	0.1	0.12	0.05	0.025	0.025	0.01	0.005
Co	1.6	17.1	2.1	16.4	18.8	13.2	1	1.15	16.3	32	21.9	5.35	35.2	9.7	16.6	34.6	6.6	17.2	13.9	17	3.9	5.4	3.5
Cr	1.6	14.4	1.3	7.6	88.3	0.8	0.25	0.25	23.2	72.3	52.1	54.9	1450	10.6	97	71.5	20	33.9	19.5	71	6.3	4.15	8.8
Cu	5.1	246.5	5.15	117	42.5	45	0.25	0.25	141	152.5	135	22.8	24.5	39.7	47.3	192	13.3	15.9	50.5	26.8	2.93	3.35	0.25
Fe	6190	43600	7880	22100	33800	50200	2830	5400	36100	48800	25800	6170	32800	50700	39900	106000	18900	51500	28200	47300	17700	13300	19900
Hg	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.025	0.01	0.01	0.01	0.01	0.01	0.03	0.055	0.01	0.01	0.01	0.01	0.01	0.01
K	2050	2100	2300	1300	14500	12200	17600	13400	300	650	300	700	200	6600	10100	5000	450	7300	500	17300	3650	5500	3900
La	11.1	14.3	12	1.4	11.7	23.6	97.3	99.4	4.8	2.5	4.4	4.05	1.83	18	8.3	20.5	4.1	33.3	3.6	30.8	95.1	18.5	26.3
Li	8.7	8.15	12	4.4	24	7.4	4.3	4.5	6	12.6	10.3	3.15	10.7	8.3	8.51	15.5	8.8	19.4	13.8	34.8	19	21.2	12.8
Mg	1560	9120	2420	9960	21400	5620	150	280	4390	15100	9120	4920	52500	8410	14100	12000	27300	17400	17100	16000	3610	4950	2550
Mn	65	454.5	97.3	235	344	611	231	293	384	558	344	109.5	247.5	615	388.5	519	1310.5	352	382	388.5	195.5	147.5	91.4
Mo	0.5	0.5	0.5	0.5	0.5	0.5	1.3	2.55	0.5	0.5	0.5	0.5	0.5	0.5	0.5	5.3	2.6	0.5	0.5	0.5	0.5	0.5	0.5
Na	450	1720	420	2040	510	340	46200	43800	2660	2970	4170	2650	140	920	1130	390	13	130	1100	230	460	520	180
Ni	2.55	18.1	2.55	26.3	70.8	9.7	0.5	0.9	27.7	52.9	38.7	15.1	387.5	14.6	30.4	86	19.5	22.2	21.7	24.9	3.2	7	6.8
P	285	1246	182.5	216	726	1370	232	286	688	434.5	573	175	161	543	357	414	406	732	643	698	361.5	523	412
Pb	4.85	5.9	3.85	1.1	2.3	3.9	4.3	5.8	0.5	0.25	0.9	1.75	0.6	2.2	2.3	4.6	2.95	3.65	0.4	4.3	11.5	1.75	1.8
S	28	437.5	20	23	42	64	73	61.5	869	868.5	966	30.56	23	41	44.5	17700	1769	1348.5	90	184	32.5	14	10
Sb	0.005	0.0075	0.005	0.005	0.005	0.005	0.03	0.035	0.02	0.15	0.07	0.005	0.005	0.005	0.005	0.05	0.013	0.045	0.005	0.02	0.005	0.005	0.02
Sc	0.8	9.15	1.35	3.2	2	6.2	0.05	0.1	9.2	10.7	9.6	2.65	1.1	4.5	7.45	5.7	3	6.2	7.7	7.85	2.35	1.45	0.7
Se	0.005	0.34	0.005	0.05	0.03	0.03	0.02	0.02	0.31	0.185	0.31	0.015	0.025	0.06	0.045	4.4	0.123	0.11	0.03	0.063	0.02	0.02	0.005
Si	180	300	170	270	170	280	380	370	340	300	300	300	410	390	360	230	330	320	300	340	220	210	230
Sr	5.55	12.9	6.1	28.3	38.1	47.9	1410	1056	29.3	65.8	97.1	78	3.2	27.4	6.05	14.1	144.9	49.5	16.6	38.8	11	12.8	1.8
Te	0.005	0.0275	0.005	0.01	0.005	0.01	0.005	0.005	0.005	0.0175	0.03	0.005	0.005	0.005	0.005	0.22	0.028	0.005	0.005	0.018	0.005	0.005	0.005
Th	9	4.75	7	2.5	7	10	18	18.5	2.5	2.5	2.5	2.5	2.5	7	11	16	2.5	14	2.5	14.1	54.5	5	13
Ti	481	3370	615.6	1100	2750	2540	1820	1745	2010	2485	2440	510.5	321	3140	2170	1610	72.7	2065	2370	2865	1215	1335	1330
V	5.05	122	8.1	44.5	60.6	24.8	2.9	4.95	81.3	143.7	127	18.4	48.5	26.6	52.1	180	29.4	66.6	96	93.2	16.8	22.1	17.8
Y	2.55	15.7	2.55	2	3.6	15.6	22	23.3	9	9.35	9.8	1.9	0.6	10.4	4.85	11.9	8.2	21	10	10.7	6.85	1.7	6.6
Zn	31.4	51	30.4	36.3	74.3	81.2	78.7	89.6	44.4	44.3	51.8	16.2	40.3	58.6	68.7	65	23.7	72.2	35.4	96.4	52.5	48.4	15.8

Tab. 2.1.2 Mean composition (ICP-AES) of aqua regia leached Quaternary deposits (mg/kg)

Type	C1						C2				C3					C4			C5						C6				C7		C8	
	TL <.064	TL <2.0	WR	WG	MS	AM	TL <.064	TL <2.0	WG	LG	TL <.064	TL <2.0	WR	WG	ALV	TL <.064	TL <2.0	WG	TL <.064	TLA	TLB	TLC	TL <2.0	WR	TL <.064	TL <2.0	WR	WG	TL <.064	TL <2.0	TL <.064	TL <2.0
n=	6	1	1	1	1	4	13	4	2	1	10	1	10	1	1	5	3	2	14	4	8	7	6	2	4	1	4	2	17	7	15	6
Al	22900	5250	35900	21700	18100	18500	29700	18500	67400	20100	21100	9570	99400	17200	45500	21500	14000	26000	12900	15500	16800	17000	11500	22900	13050	5230	9450	9300	27700	20600	12400	8340
As	5.5	1	1.5	4	8	4	1.5	0.25	1.5	1.5	1.5	0.4	8	3	7.5	1.5	0.47	4.5	9	3.25	5.5	6	2.58	65.5	5.5	0.05	3	2.75	3	1.06	1.5	0.4
Ba	44	16.6	41.9	137	137	48.2	136	87.7	78.3	58.7	104	64.7	385.5	105	276.5	39.7	122.3	109.5	90.1	91.2	84	92.6	52.7	216.5	55.4	22.7	35.4	24	82.3	84.2	60.5	45.8
Ca	4200	2420	2600	4800	5700	4200	4800	5830	4950	4800	4850	3570	2350	4600	5150	4800	4090	6850	4600	4900	4800	6100	4270	4700	3500	3190	3500	2800	1900	1440	2900	2430
Co	13.3	4.7	9.6	18.4	13.8	14	20.3	14.7	20.5	10.2	9	5.9	6.9	9	6.25	13.8	14.7	15.6	42.9	13.1	20.6	24.7	11.1	54	25.4	3.8	12.9	12.9	64.1	8.81	32.2	5.53
Cr	50.2	12.9	53.4	73.1	65.4	44.3	73.1	58.5	71.7	38.1	35	21.1	11.7	27.5	16.9	49.2	64.3	47.2	28.7	32.7	33.5	34.6	23.2	18.6	18	6.3	15.4	6.7	16.3	40.8	14.7	22.1
Cu	63.8	24.8	32.6	47.4	29.5	80.8	75	46.6	82.6	30.3	38.1	27.7	22.2	53.5	31.3	70.9	67.5	71	57.5	60.1	130.5	97.9	38.7	214.5	18	12.7	15.4	6.7	16.3	11.1	14.7	6.73
Fe	28000	9600	31400	37100	31600	25700	31600	19800	27000	16200	17300	10400	15700	15300	14600	26700	23900	24400	25900	24600	38200	39700	19800	85300	18500	10300	12300	9600	21200	19500	19800	12900
K	950	100	500	2600	3500	950	3600	1950	750	1100	1600	1100	1600	1700	1750	2900	5030	2250	1850	2350	1960	2700	1920	5950	1200	1100	550	450	2300	2700	1900	1650
La	17	8.3	13	28	29.7	17.2	15.5	10.4	11.4	13.2	22.4	11.1	163.5	39	124.1	22.4	12.4	17.4	22.2	27.6	20.6	23.7	12.9	19.4	31.6	32.2	24.6	23.8	24.6	18.9	29.6	18.3
Li	10.4	5	14.2	12.8	13	8.8	10.5	7.88	8.7	6.2	7.6	4.9	11.1	6.5	6.6	12.9	12.4	9.8	7.4	8.8	8.3	9.3	7.67	9.9	14	8.3	6.8	8.9	24.6	22.2	13.1	11.1
Mg	7000	3000	5700	11000	9600	6300	9700	7680	6250	4400	4550	3070	3050	3900	2650	6000	8600	5550	6850	7060	10900	11200	6370	13800	4850	2690	2350	2200	6900	6660	5200	3740
Mn	236	123	173	339	252	269	280	255.8	392.5	141	269.5	151	738	270	543	223	297.7	270.5	256.5	271.5	658	285	245.7	1570	196	134	132	97.5	194	180	197	226
Na	450	170	400	500	500	400	600	790	950	700	600	500	950	600	2600	500	343	800	400	450	400	400	392	300	350	300	400	300	300	190	400	250
Ni	31.9	11.7	27.9	41	36.1	28	103	65.4	163	47.9	21.4	14.1	15.4	34	17.6	45.6	37.8	43.4	24	37	36.8	38.5	20.6	32.6	14.8	4.9	7.5	7.4	36.4	24.8	46.8	11.6
P	688	246	518	640	1140	869	686	424.8	615	798	764	452	809	812	1391	1100	613	1185	996	856	1020	955	655.3	1330	538	548	522	508	715	436	572	349.8
Pb	6	1.1	9	6	5	6	1.5	0.85	1.5	4	7	1	16	5	26.5	5	2	4	6.5	6	5	9	1.68	2.75	6.5	2.4	7.5	7.5	7	2	7	1.4
S	209	13	425	45	47	105	158	41.5	628	146	108.5	19	371	35	564.5	124	27	159	33	35.5	24.5	799	25.8	56.5	44	13	83.5	159	95	51.7	51	19.5
Sc	5.45	1.9	5.9	7.8	6.1	5.1	5.1	3.85	5	3.1	3.2	1.9	1.9	2.5	1.45	4.5	3.87	3.75	4.9	5.5	6.85	6.9	4.28	22.2	3.75	2.5	3.4	2.6	3.5	3.57	3.3	2.42
Si	800	110	1300	300	300	250	800	200	1500	900	650	180	1100	500	2050	600	220	1000	300	400	600	400	220	500	400	190	600	550	400	290	400	270
Sr	13.4	6.9	10.2	21.4	20.1	14.4	16	18.4	28.9	14	55	41.1	451.5	115	681.5	11.5	8.47	17.7	12.9	11.9	12.2	20.7	9.67	19.1	10.5	7.4	8.8	8	10.9	10.8	12.1	8.77
Ti	1940	972	1690	2570	2030	1780	1820	1245	1265	1070	1190	887	1695	956	1430	1670	1793	1267	1195	1495	1226	1490	1093	1875	1375	1020	1100	950.5	1380	1499	1490	977.8
V	62.4	25.1	55.5	84.4	73.8	58.5	57.7	45.8	61.5	36.8	35.9	24.7	25.4	29.4	26.9	49.9	48.6	51.6	55	52.5	82.6	93.3	46.1	201	37.1	18.9	25.5	19.5	44.2	41.2	39.5	30.5
Y	7.9	3.1	6.2	9.9	10.5	8.2	6.2	4.75	4.5	5.7	8	3.8	51.5	11.5	31.5	9	7.9	7.65	11.7	11.7	13	12.8	5.72	23	10.2	8.7	8.35	7.8	5.7	4.51	8.2	5.87
Zn	38.5	14.6	40	55.5	49.8	37.4	43	26.5	45.9	21.7	31	18	141	38.7	82.3	39.9	36.7	40.3	44.7	48.3	56.2	62	42	72.3	25.9	20.5	16.3	18.8	40.5	38.9	20.7	12.3

WR - weathered crystal rocks  
 TL <.064 - till, fraction <0.064  
 TLA - till, horizon A, fraction <0.064  
 TLB - till, horizon B, fraction <0.064  
 TLC - till, horizon C, fraction <0.064  
 TL<2.0 - till, fraction <2.0  
 WG - water-glacial sediments  
 LG - lake-glacial sediments  
 MS - marine sediments  
 AM - ablation moraine  
 ALV - alluvial sediments

Tab. 2.1.3 Mean total composition (XRF) of bedrock samples (%)

Catch	C1	C2	C3	C4	C5	C6	C7	C8
# samples	12	9	17	17	17	6	4	5
SiO2	69.34	57.30	53.68	57.64	55.56	70.41	69.71	75.67
Al2O3	14.29	14.76	18.17	13.26	10.84	14.45	15.85	11.22
Fe2O3	3.76	9.20	7.60	9.39	9.97	2.45	2.03	3.05
TiO2	0.55	0.69	1.36	0.73	0.97	0.30	0.27	0.34
MgO	1.09	4.72	2.19	6.54	3.27	0.52	1.14	1.14
CaO	2.61	7.06	3.66	5.69	7.46	1.48	2.43	1.71
Na2O	4.03	2.72	7.08	2.63	2.12	3.74	4.61	1.98
K2O	3.06	1.28	4.18	1.72	1.39	4.70	2.44	2.85
MnO	0.05	0.13	0.21	0.12	0.22	0.03	0.02	0.06
P2O5	0.11	0.16	0.13	0.13	0.25	0.08	0.17	0.11
LOI	0.54	0.96	0.90	1.32	6.79	0.59	0.75	0.86
Sum	99.43	98.99	99.17	99.17	98.83	98.74	99.40	99.02



Tab. 2.1.4 Mean total composition (XRF) of Quaternary deposit samples (%)

Catch	C1	C2	C3	C4	C5	C6	C7	C8
# samples	1	4	1	3	6	1	3	6
SiO <sub>2</sub>	66.36	60.21	68.78	62.50	63.93	72.55	68.38	71.61
Al <sub>2</sub> O <sub>3</sub>	13.53	14.67	13.75	13.60	14.58	13.22	14.28	12.05
Fe <sub>2</sub> O <sub>3</sub>	5.57	7.43	4.14	7.41	6.93	2.20	3.74	3.69
TiO <sub>2</sub>	0.72	0.68	0.46	0.75	0.73	0.32	0.50	0.46
MgO	2.40	4.86	2.02	3.62	2.73	0.78	1.89	1.64
CaO	4.10	5.51	4.02	4.79	4.28	1.78	2.70	2.51
Na <sub>2</sub> O	3.40	2.81	3.67	3.21	3.18	3.61	3.49	2.94
K <sub>2</sub> O	1.76	1.20	1.58	1.67	1.42	3.52	1.62	2.56
MnO	0.08	0.11	0.07	0.11	0.10	0.04	0.05	0.07
P <sub>2</sub> O <sub>5</sub>	0.14	0.12	0.11	0.16	0.16	0.12	0.10	0.09
LOI	1.47	2.19	1.26	1.79	1.64	0.81	2.73	1.59
Sum	99.54	99.78	99.86	99.61	99.68	98.94	99.48	99.19

Tab. 2.1.5 Overview of the bedrock and Quaternary deposits characteristics, and their geochemical peculiarities

Catchment (No.)	Bedrock	Quaternary deposits	Geochemical characteristics of rocks & tills
Zapoljamij (C1)	amphibole-biotite gneiss >> granites	basal till (60%) >> meltout till (26%) >> eluvial dep. (7%) >> rock terrains, water	Moderately high conc. of Mg, Ca, S, Cu, Cr and Ti in tills, low in rocks
Monchegorsk (C2)	andesitic effusives, tuffs > gabbros, gabbro-norite > pyroxenite	basal till (76%) >> glaciofluvial sed. (9%) > eluvial dep. (8%) >> glacio-lacustrine sed. (4%) > water, lacustrine sed., rock terrain	High conc. of Ni, Cr, S, Cu, Fe, Mg, Zn in rocks, moderately in tills
Kirovsk (C3)	Trachytoid>hibinite >> gabbro-norite > metabasalts & meta-andesites > black schists, mafic tuffs	basal till (57%) > deluvial - eluvial dep. (41.4%) >>rock terrain	High conc. of Al, Na, Sr, Y, K, Ba, As, Mo and S in rocks and lake sediments, less in tills
Kurka (C4)	amphibolite > two-mica gneiss > gabbro-norite > gabbro-amphibolite	basal till (54%) > peat (30%) >> glaciofluvial sed. (6%) > lacustrine sed. (5%)> rock terrain, water	Moderately high conc. of Fe, Ni, Cr, Ba, Zn, V, S, Cu in rocks, less in tills
Skjellbekken (C5)	andesites, andesitic volcanoclastic schists (40%) > tholeiitic basalts & tuffs (30%) > black schists (20%) > others	basal till (59.2%) >> glaciofluvial sed. (17%) > meltout till (12.4%) >> rock terrain, water	Moderately high conc. of Mg, Ca, S, Fe, Cu, Ni and V in black schists; high conc. S, As and Cu in tills
Kirakka (C6)	granite (100%)	rock terrain, block fields (60%) > meltout till (34%) > water	High conc. of Th, La, Ba, Pb, Zn and Mo in granite, less in tills
Naruska (C7)	tonalitic gneiss (mica gneiss, amphibolite interlayers)	basal till (63%) >> peat (22%) > rock terrain, block fields (14%) > water	Moderately high conc. of Mg, Co and S in tills, less in rocks
Pallas (C8)	quartzite >> amphibolites	basal till (69%) >> peat (29%) >> rock terrain (2%) > water	Moderately high conc. of Mg and Ni in tills, low in quartzites

*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
 Catchment locations

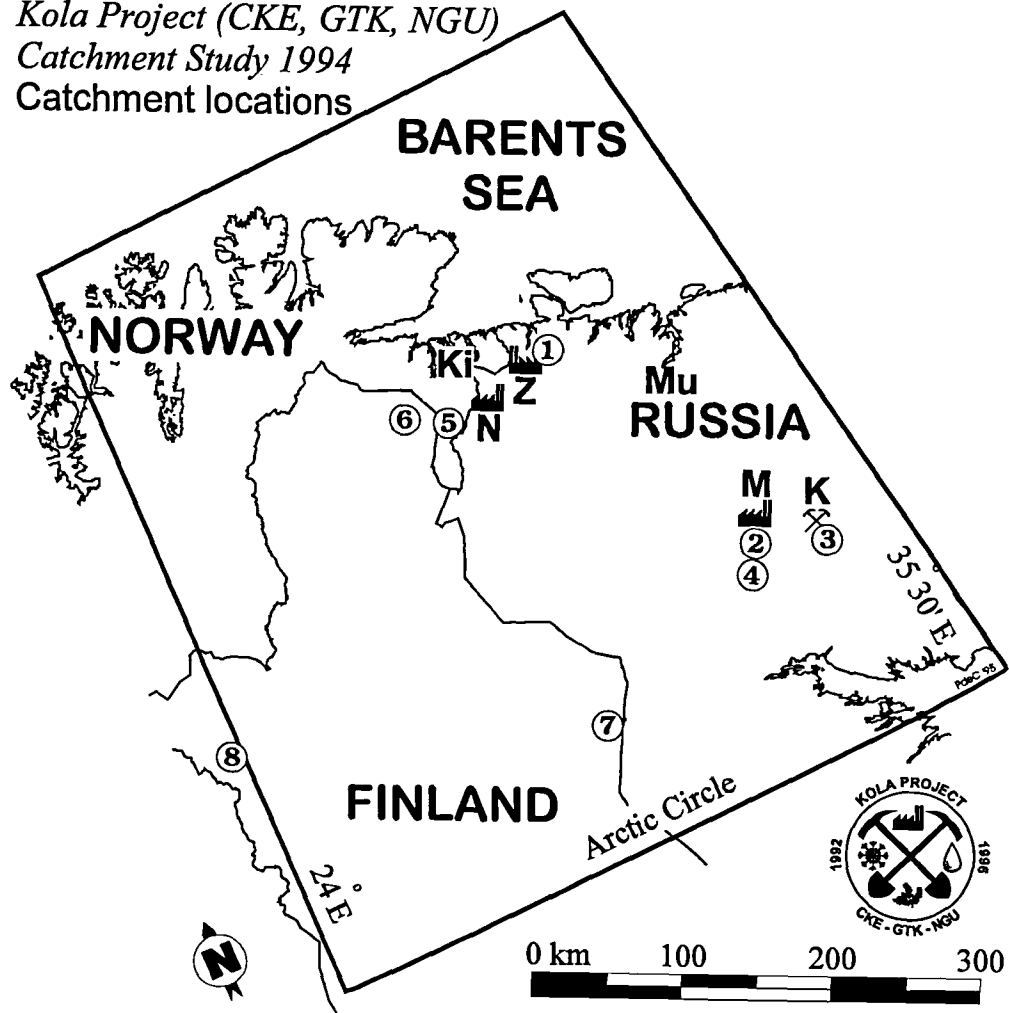
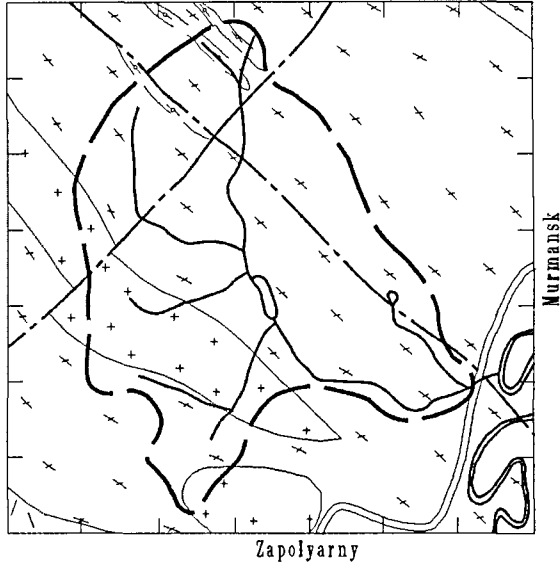
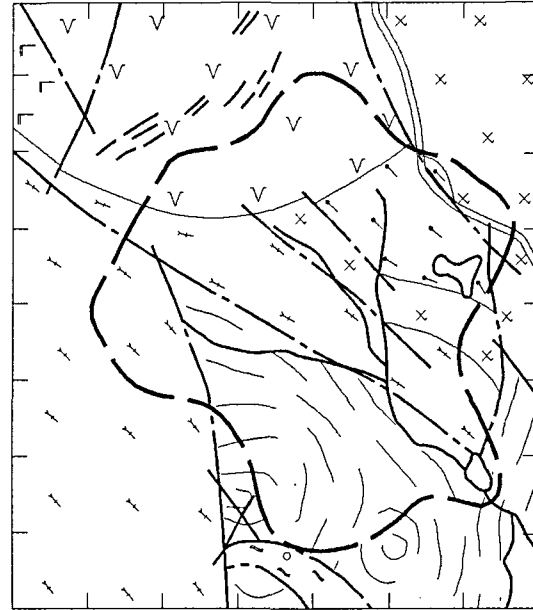


Figure 2.1.1. Location map of catchments in Russia (C1: Zapoljarnij, C2: Monchegorsk, C3: Kirovsk and C4: Kurka), Norway (C5: Skjellbekken) and Finland (C6: Kirakka, C7: Naruska and C8: Pallas). K: Kirovsk, Ki: Kirkenes (open-pit iron mine and mill), M: Monchegorsk (nickel-copper smelter, cobalt smelter, etc.), Mu: Murmansk, N: Nickel (nickel-copper smelter), Z: Zapoljarnij (nickel-copper ore roasting plant).

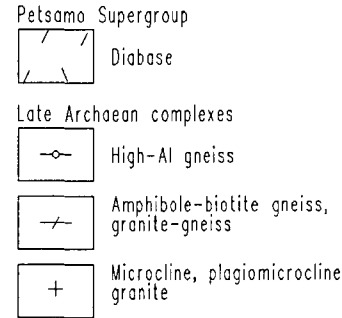
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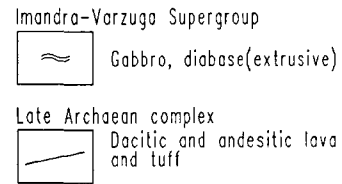
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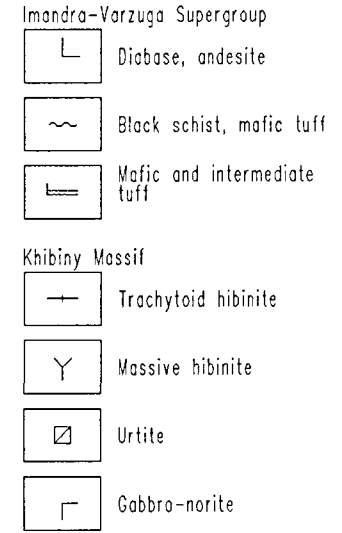
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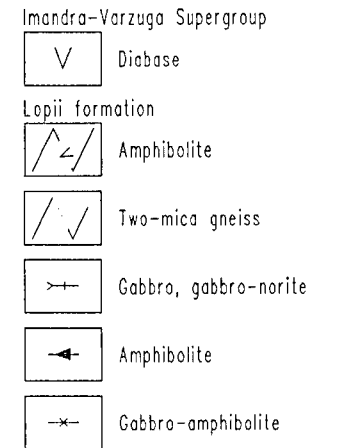
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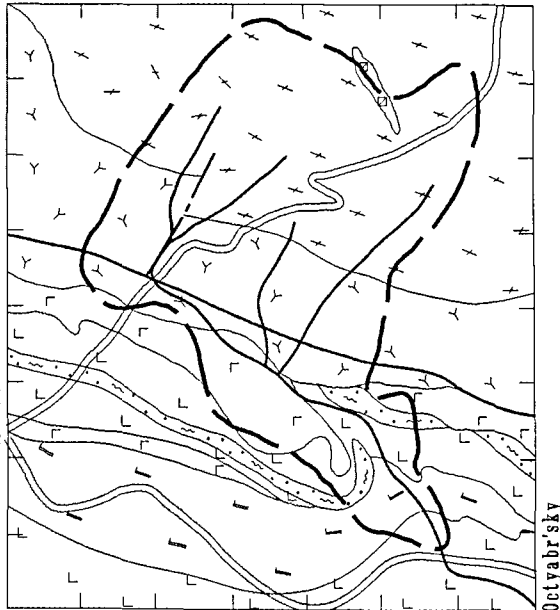
Catchment 3



Catchment 4



Catchment 3



Catchment 4

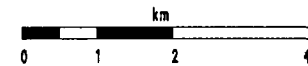
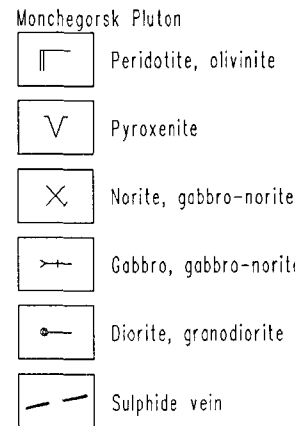
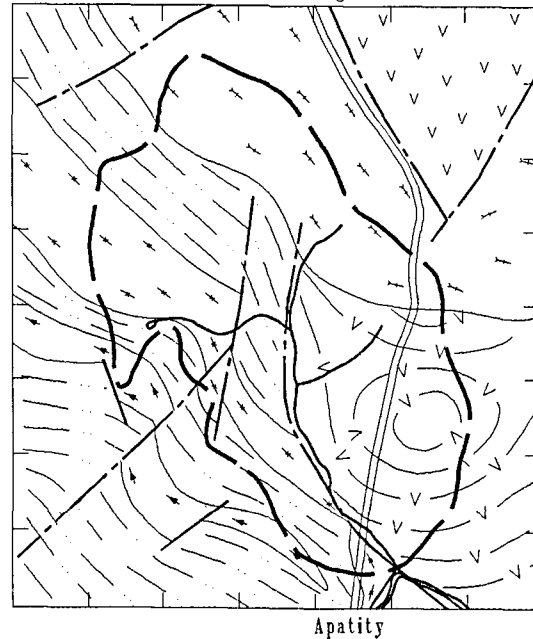
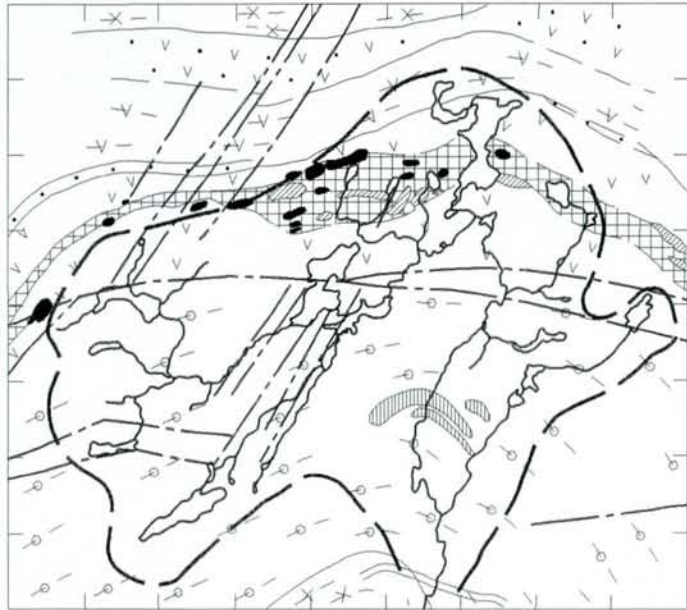


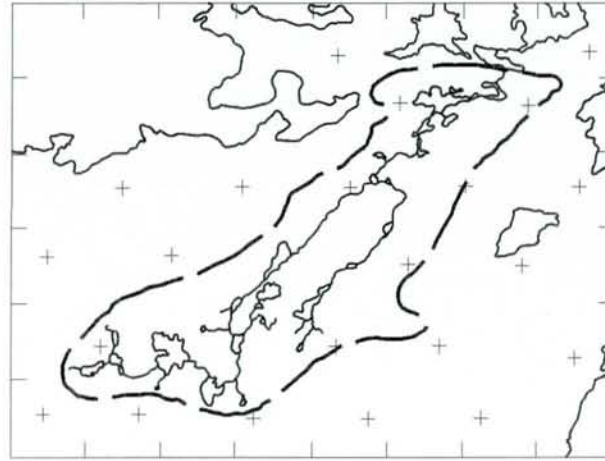
Figure 2.1.2 Lithological maps of C1-C8

Catchment 5

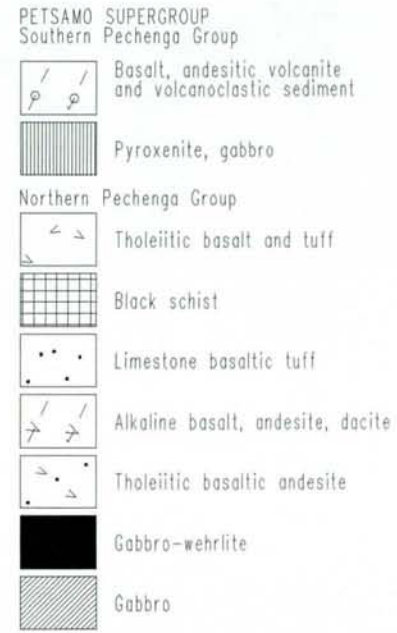


Nirod Kirkenes

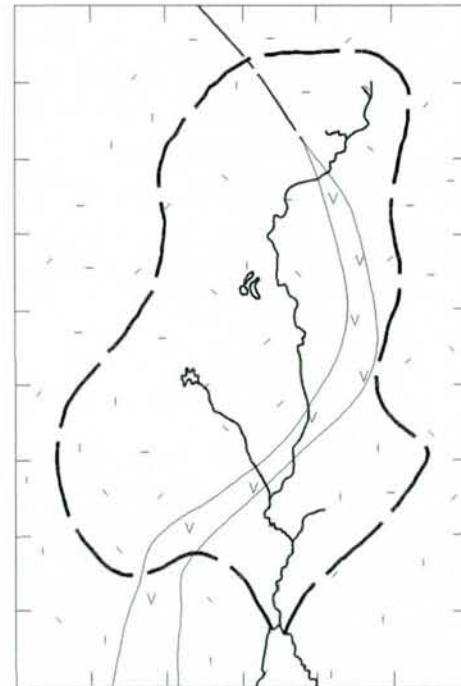
Catchment 6



Catchment 5



Catchment 8



Catchments 6,7,8

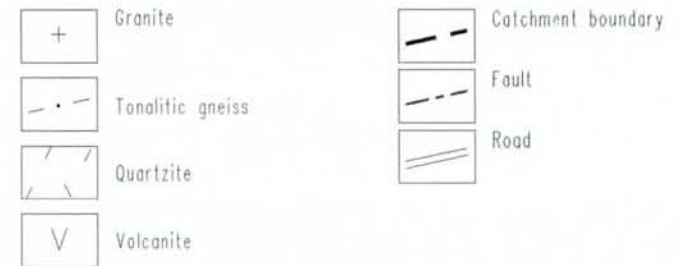
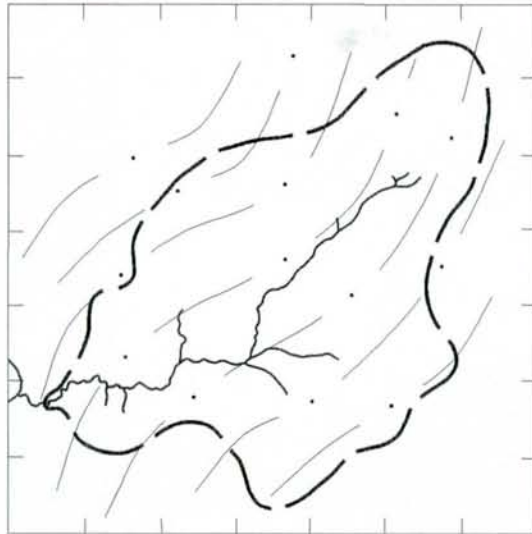
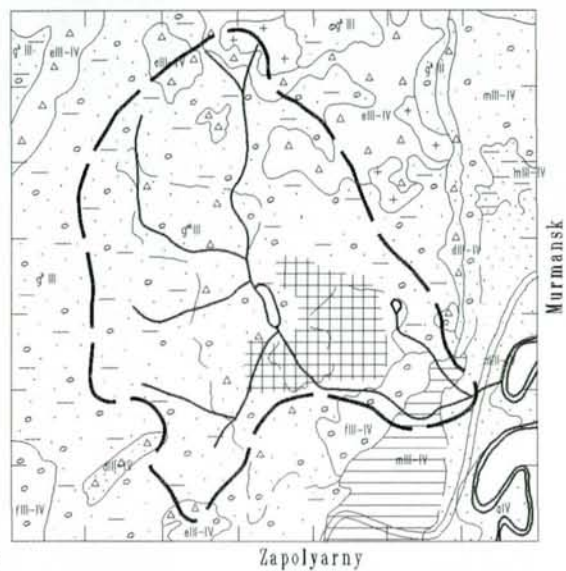


Figure 2.1.2 Lithological maps of C1-C8

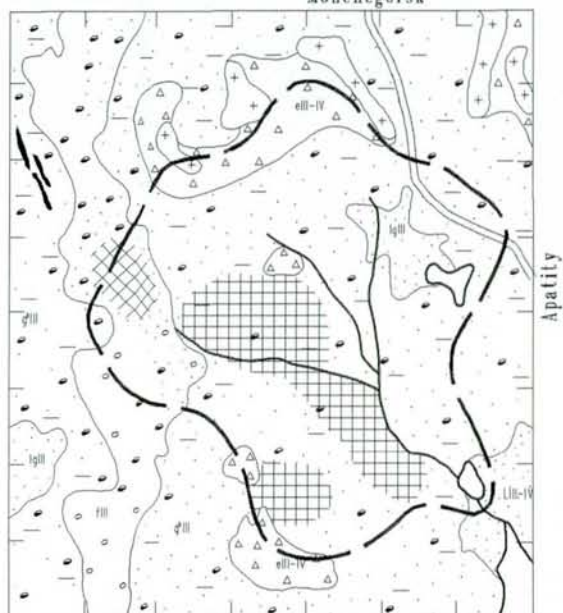
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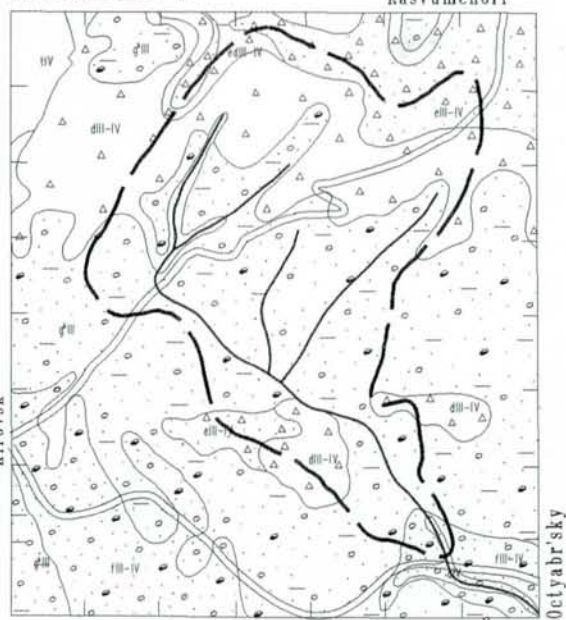
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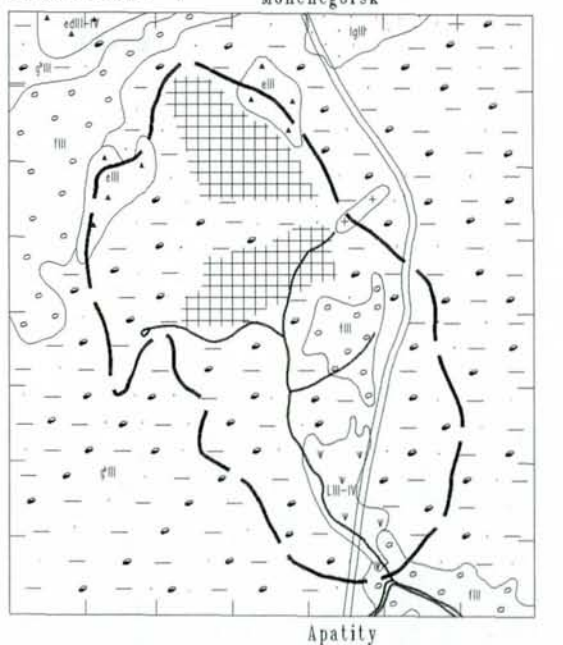
Catchment 2



Catchment 3



Catchment 4



Catchments 1,2,3,4

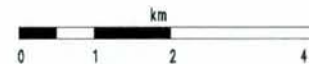
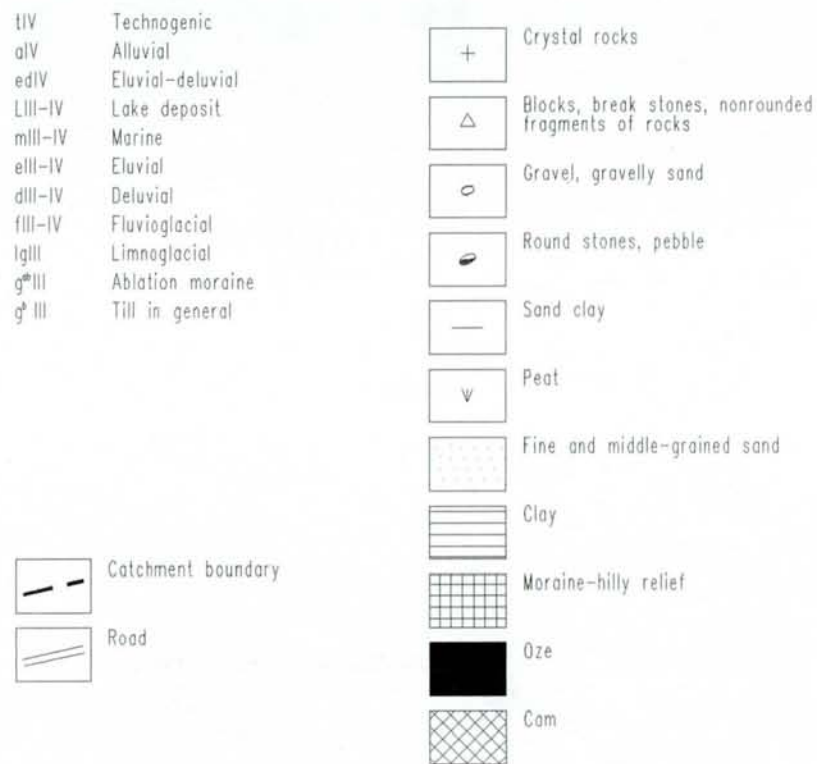
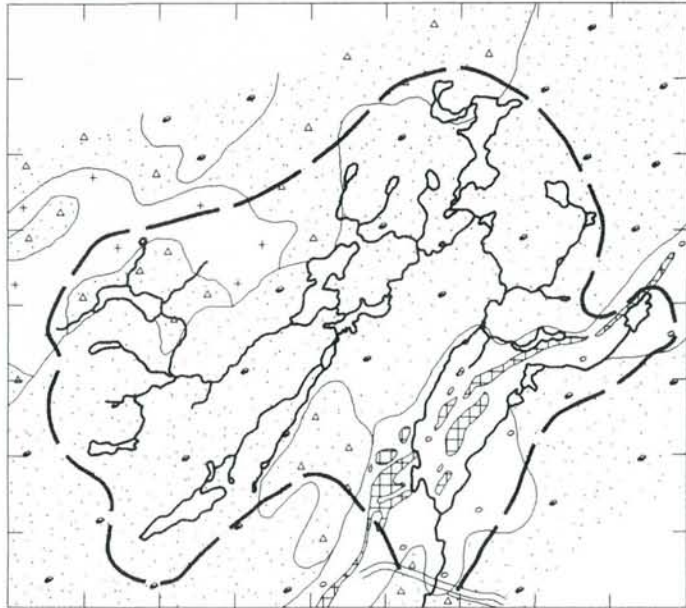


Figure 2.1.3 Maps of Quaternary deposits of C1 - C8

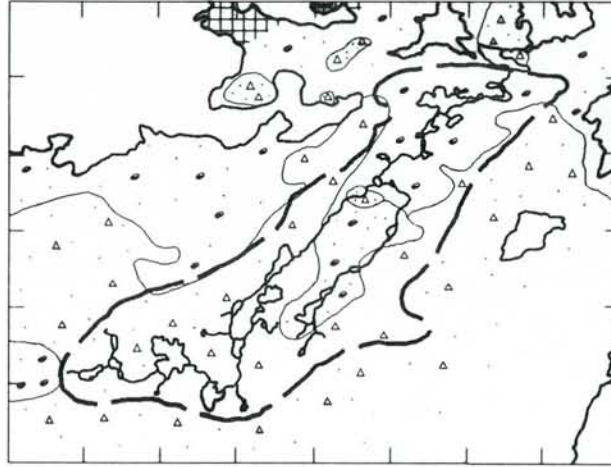


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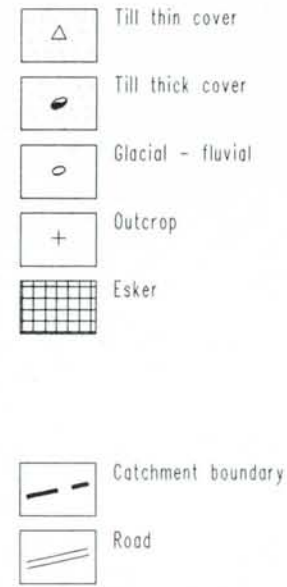


Nirod Kirkenes

Catchment 6



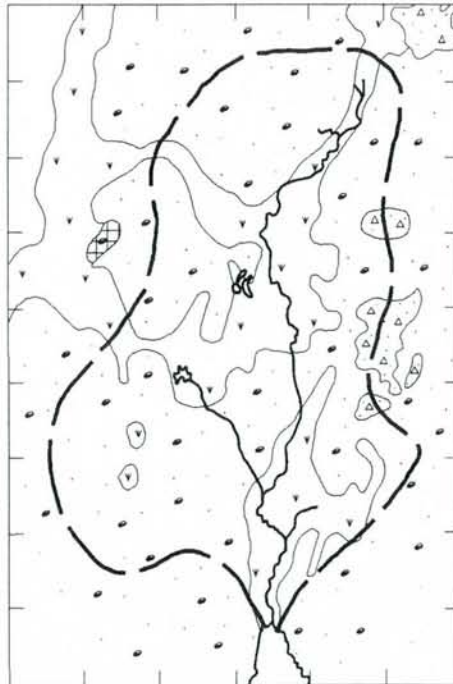
Catchment 5



Catchments 6,7,8



Catchment 8



Catchment 7



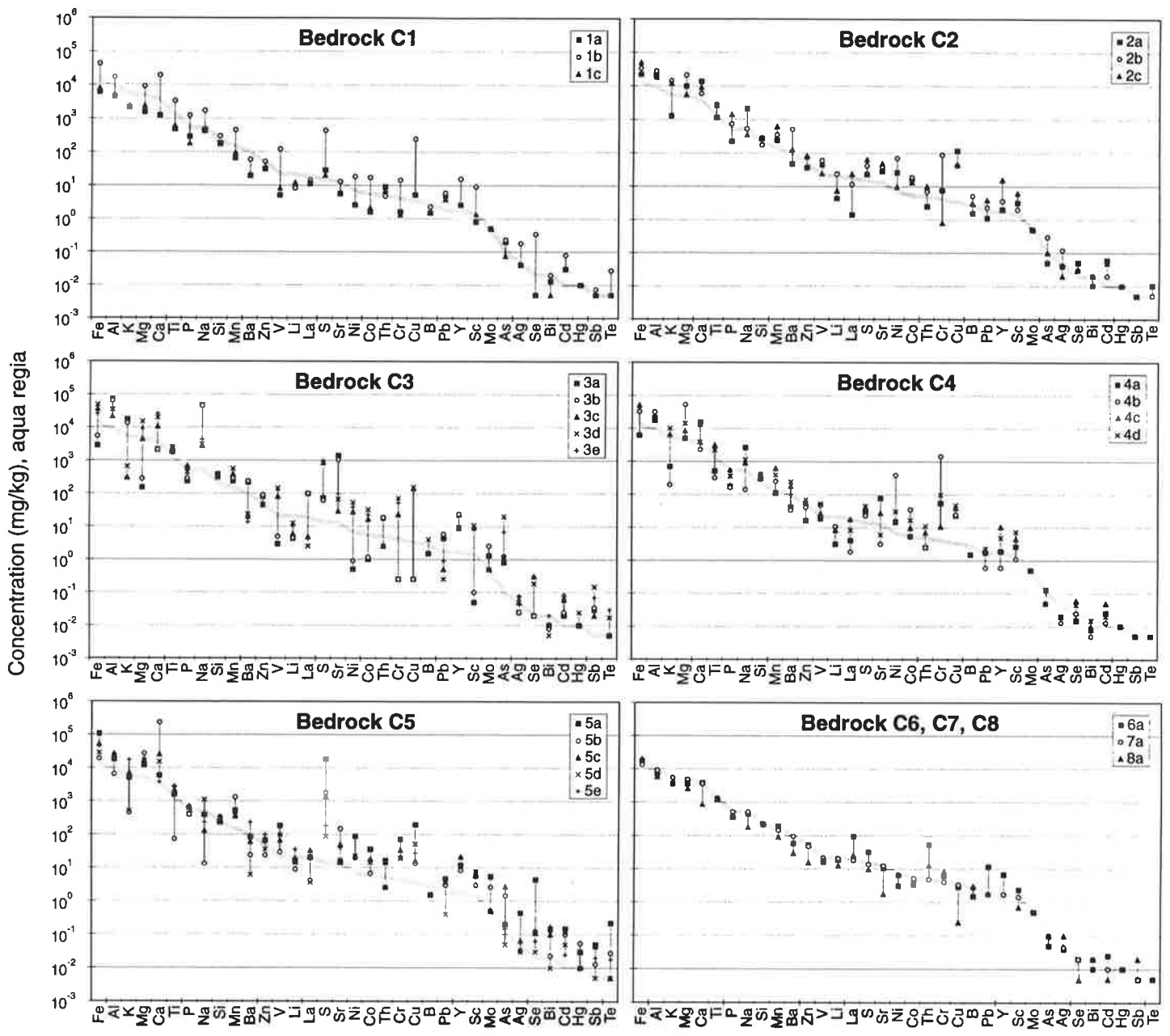


Figure 2.1.4. Composition (in mg/kg) of bedrocks in C1-C8 (aqua regia extractable fraction). Elements are ranked in order of decreasing abundance in C7 (rock type 7a) from left to right. The grey line represents the trend in C7, and allows easier comparison with other catchments.



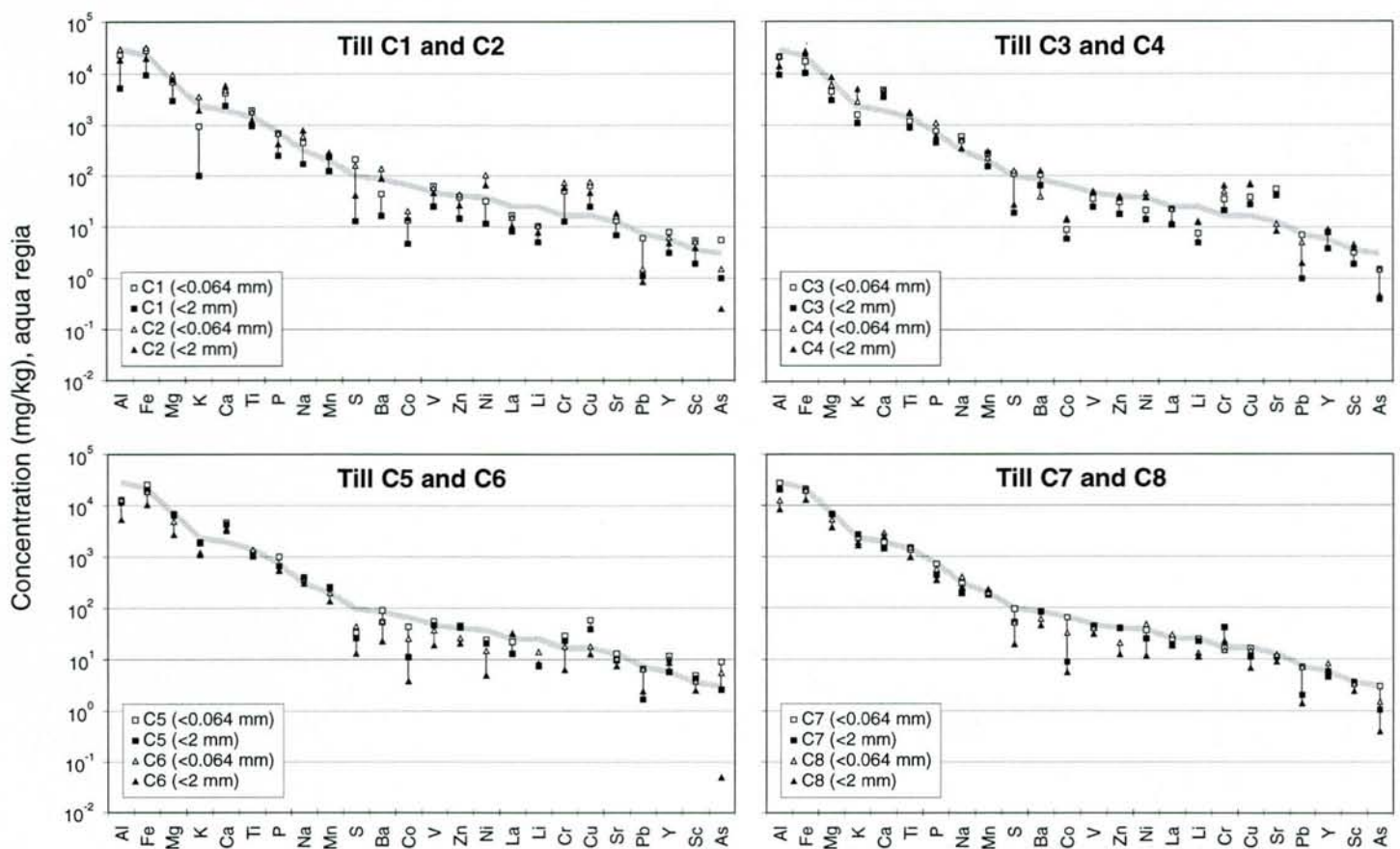


Figure 2.1.5. Composition (in mg/kg) of fine and coarse fractions of till in C1-C8 (aqua regia extractable fraction). Elements are ranked in order of decreasing abundance in C7 (fine fraction) from left to right. The grey line represents the trend in C7, and allows easier comparison with other catchments.

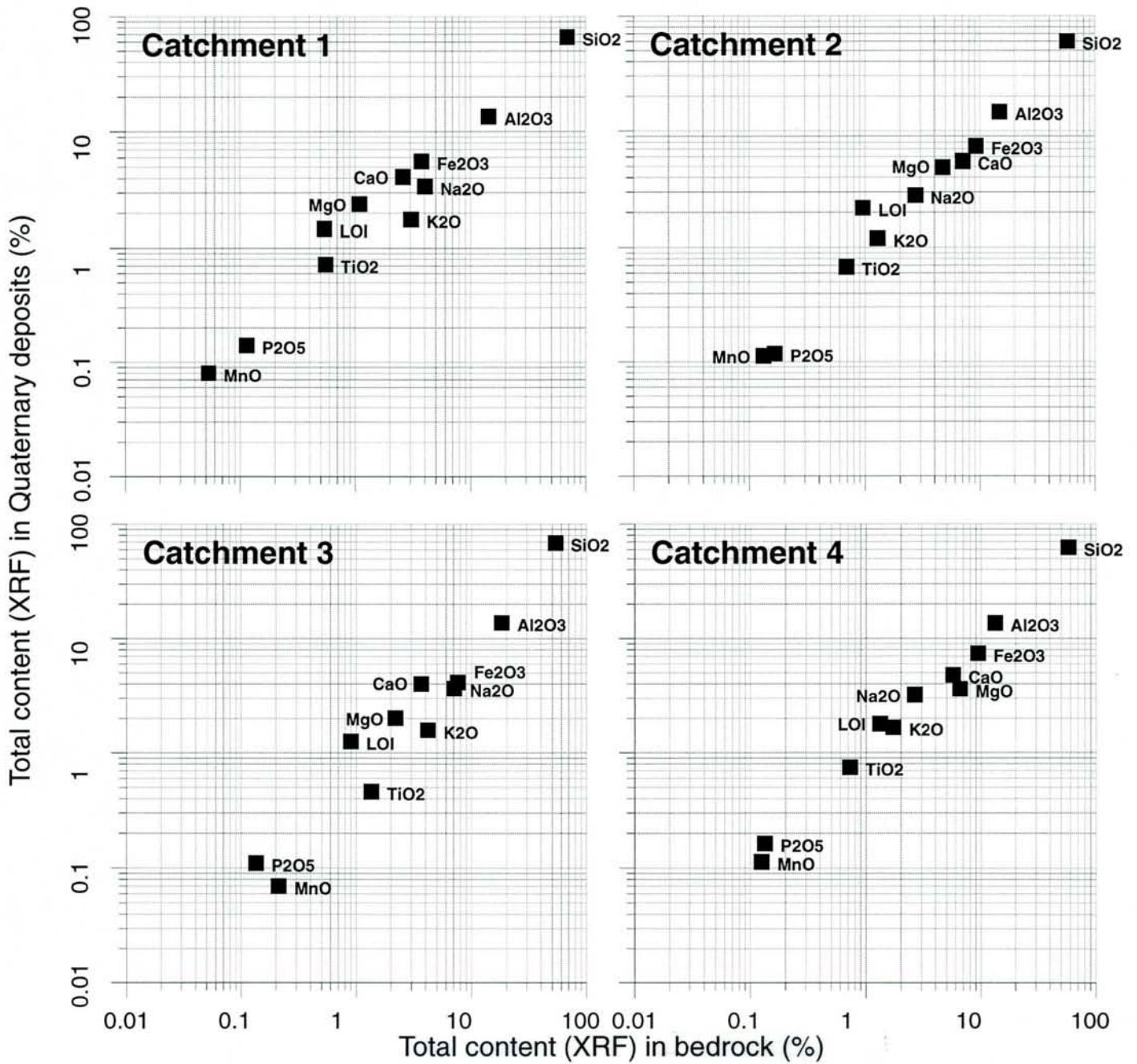


Figure 2.1.6. XY-diagram of Quaternary deposits vs. bedrock composition (in %) in each catchment for major oxides (XRF data).

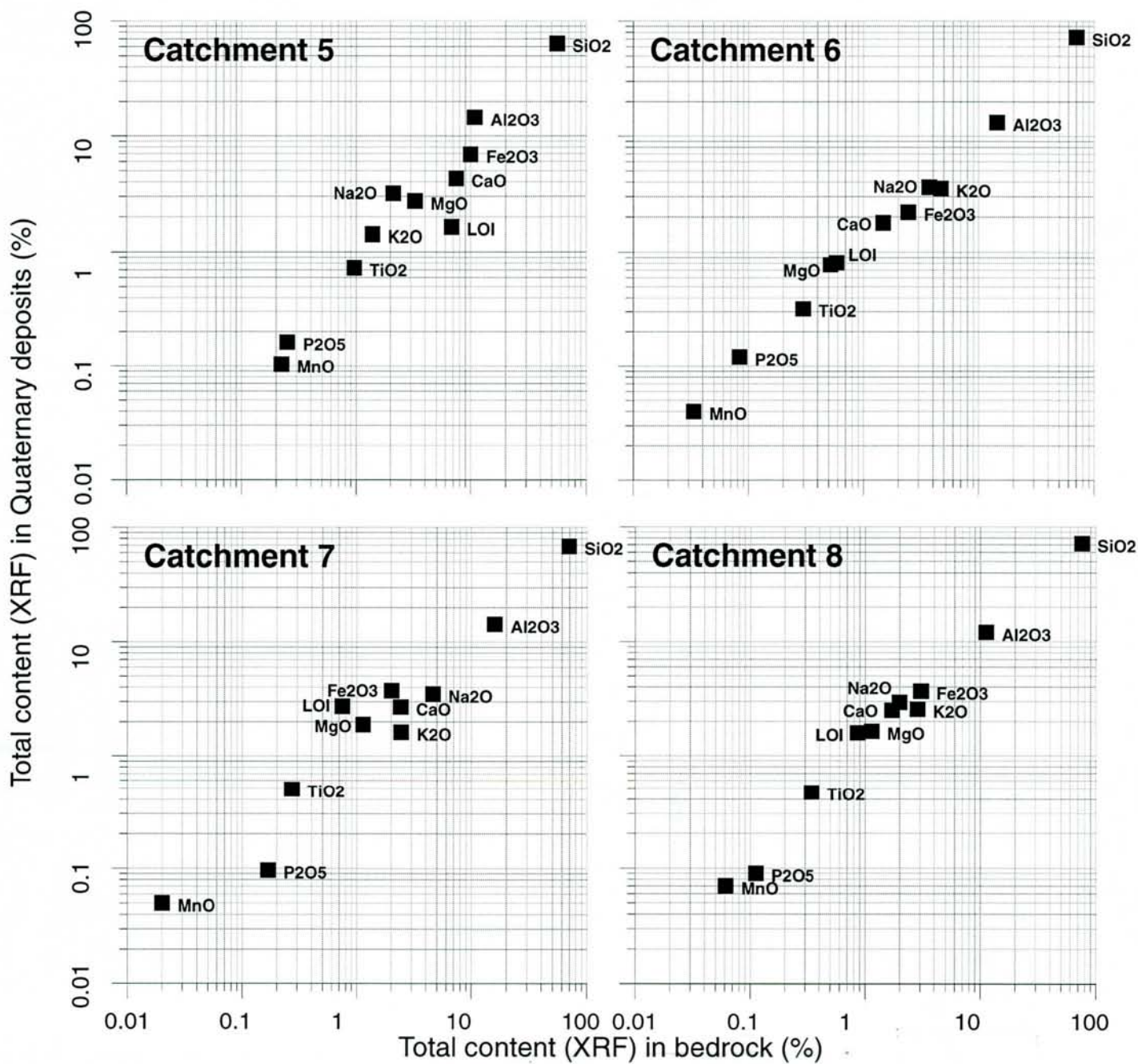


Figure 2.1.6. XY-diagram of Quaternary deposits vs. bedrock composition (in %) in each catchment for major oxides (XRF data).

### 3 PRECIPITATION

#### 3.1 Ecogeochemical Investigation, Kola Peninsula: sulphur and trace element content in snow

*Matti Äyräs, Patrice de Caritat, Viktor A. Chekushin, Heikki Niskavaara and Clemens Reimann*

##### ABSTRACT.

The Central Kola Expedition and the Geological Surveys of Norway and Finland have undertaken a multi-media investigation of eight catchments situated in Russia (Zapoljarniy, Monchegorsk, Kirovsk, Kurka), Norway (Skjellbekken) and Finland (Kirakka, Naruska, Pallas) to determine the environmental impact of local industrial pollution. Results of analysis of snow samples collected in March/April 1994 are presented for both filtered meltwater and filter residues. Many heavy metals (As, Cd, Co, Cr, Cu, Mo, Ni, Pb, V and Zn), as well as S, Al and Fe, are unusually enriched in both phases in the Russian catchments, primarily in Monchegorsk and/or Zapoljarniy. Typically, some elements are found mostly in the meltwater phase (e.g. Na, S, Sr, Zn), others in the filter residues (e.g. Cr, Fe, Mo, Ni). Cu and Ni are well correlated, both in meltwater and in filter residue, and the ratio of Cu to Ni can be useful in identifying pollution sources. Snow is a well-suited medium for fingerprinting environmental pollution sources. Estimates of deposition from snow meltwater only, will be seriously underestimated for many elements.

##### INTRODUCTION

The Central Kola Expedition (CKE), the Geological Survey of Norway (NGU) and Geological Survey of Finland (GTK) are participating in a co-operative project in the Russia -Norway-Finland tri-state area, north of the Arctic Circle. A pilot project was carried out in 1992-93 to harmonise field and laboratory methods and test different sampling media (Chekushin et al., 1993, Reimann et al., in press). In 1994, 10 different sampling media, including snow, were collected in eight catchments in Russia (Zapoljarniy, Monchegorsk, Kirovsk, Kurka), Norway (Skjellbekken) and Finland (Kirakka, Naruska, Pallas) as part of the project «Kola Ecogeochemistry» (Fig. 3.1.1). The main aims of this study are to evaluate element input and cycling in the catchments, and to determine the environmental impact of local industrial pollution.

The Kola Peninsula in NW Russia has been a region of intense industrial activity (mining, ore processing, smelting, etc.) for several decades. Resulting emissions of sulphur (S), aluminium (Al) and heavy metals have had a serious impact on local terrestrial and aquatic ecosystems (Sivertsen et al., 1992, Tuovinen et al., 1993, Moiseenko et al., 1995). Jaffe et al. (1995) reported deposition estimates for S and heavy metals based on analysis of snow samples collected on the eastern part of the Kola Peninsula. Here, we focus on the chemical composition of snow samples (both filtered meltwater and filter residues) collected in the western part of the Kola Peninsula, while deposition estimates are introduced in Reimann et al. (1995). Snow cover in early spring ideally is an accumulation of last winter's precipitation, containing not only solid snow with its solute components and co-deposited particles, but also blown-in dust of local natural and anthropogenic origin.



## SAMPLING AND ANALYSIS

In every catchment, snow samples were collected from a level, 20 x 20 m area having a uniform snow thickness, with a colourless plastic tube of 1 m length and 10 cm diameter (Äyräs & Reimann, 1995). Sampling took place from end of March to beginning of April 1994. Depending on catchment size, 9 to 13 samples were collected in each catchment. The snow cover at the end of winter 1994 was relatively thick, ranging from 40 to 120 cm, the smallest median value (54 cm) being from Kirakka, the biggest (107 cm) from Naruska.

The snow samples were kept frozen until arrival at the laboratory (GTK). Five samples per day were allowed to melt at room temperature, the meltwater dripping down onto, and being filtered through, 0.45 mm Millipore® mixed cellulose filters using a vacuum pump. The total volume of meltwater was recorded, and three aliquots of the filtrate were separated, one for pH and conductivity measurement, the other for ion chromatographic determination of bromine (Br), chlorine (Cl), nitrate (NO<sub>3</sub>) and sulphate (SO<sub>4</sub>), and the last for direct analysis by ICP-MS following acidification with ultrapure nitric acid. The filters were digested with 10 ml concentrated nitric acid in a microwave oven, the resulting solution was diluted to 50 ml with water and analysed with ICP-OES. Results for filter residues were recalculated to the volume of snow sample to be directly comparable with meltwater concentrations. The total number of analysis performed was 83.

## RESULTS AND DISCUSSION

### *Concentration levels: meltwater vs. filter residue*

The median pH of snow meltwater varies from 4.96 to 4.52, the highest pH value being in the Zapoljarniy catchment followed by Kirakka, Skjellbekken, Kirovsk, Monchegorsk, Naruska, Pallas and Kurka. Rather unexpected are the low pH values in Naruska and Pallas, where there are no significant local emission sources. The higher snow pH in the very polluted Zapoljarniy catchment likely is caused by basic rock dust released by open-pit mining activity and handling of ore stock piles.

Median S content in *meltwater* (Fig. 3.1.2) is highest in the Zapoljarniy (900 ppb) and Monchegorsk (825 ppb) catchments. Al concentration is highest at Kirovsk (140 ppb) and Monchegorsk (120 ppb). Highest median values for trace elements in snow meltwater are from Monchegorsk, unless stated otherwise: arsenic (As, up to a median value of 1.1 ppb), cadmium (Cd, 0.3 ppb), cobalt (Co, 33 ppb), chromium (Cr, 1.15 ppb), copper (Cu, 550 ppb), molybdenum (Mo, 0.23 ppb), nickel (Ni, 250 ppb), lead (Pb, 3.4 ppb), rubidium (Rb, 0.5 ppb, Kirovsk), strontium (Sr, 7 ppb, Kirovsk), vanadium (V, 16 ppb), and zinc (Zn, 12 ppb).

The *filter residues* (Fig. 3.1.3) have detectable S content at Zapoljarniy (1500 ppb), Monchegorsk (200 ppb), Kurka and Kirovsk. Al concentration is highest at Zapoljarniy and Monchegorsk (ca. 300 ppb). Highest median values for trace elements in snow filter residue are also from Monchegorsk, unless stated otherwise: As (5 ppb), Cd (ca. 0.1 ppb, Zapoljarniy and Monchegorsk), Co (45 ppb), Cr (11 ppb, Zapoljarniy), Cu (300 ppb, Zapoljarniy), Mo (1.5 ppb), Ni (ca. 600 ppb, Zapoljarniy and Monchegorsk), Pb (5 ppb), antimony (Sb, ca. 0.7 ppb, Zapoljarniy and Monchegorsk), scandium (Sc, 0.12 ppb, Zapoljarniy), Sr (10 ppb, Kirovsk), titanium (Ti, ca. 30 ppb, Monchegorsk and Kirovsk), V (18 ppb), and Zn (6 ppb, Zapoljarniy).

The partitioning between solid and liquid phase depends upon pollution source. In the Monchegorsk catchment, elements that are mostly in meltwater are sodium (Na), Cu, S, Cd and Zn, while those that are mostly in particulate form (filter residue) are Ni, Cr, As, Mo and iron (Fe). In Zapoljarniy, the partitioning differs, e.g. Cu and S are found chiefly in particulate form. The relative abundance

of elements in meltwater and filter residue reflects the type of polluting industry (nickel smelter in Monchegorsk vs. ore roasting plant in Zapoljarniy).

#### *Sulphur, copper and nickel in different catchments*

It is obvious that the catchments studied in Finland and Norway are at background levels, e.g. in terms of S, Cu and Ni content, compared to the heavily polluted areas in Russia (Figs. 3.1.2, 3.1.3). Relative to the Finnish catchments, the median values at Zapoljarniy and/or Monchegorsk are 4, 1000, and 800 times higher for S, Cu, and Ni (meltwater), respectively. The highest content of S in any catchment typically is in snow meltwater, except at Zapoljarniy, where the filter residue contains more S than the meltwater. In Finland and Norway, the concentration of particulate S is close to the analytical detection limit, with only a few locations in Kirakka and Skjellbekken showing detectable S contents.

In *snow meltwater*, Cu and Ni are well correlated (Fig. 3.1.4a). In Monchegorsk, Kirovsk, Kurka and Skjellbekken, Cu is more abundant than Ni, in accordance with the 1991 average emission figures quoted by Jaffe et al. (1995) for the Monchegorsk and Nikel smelters. In Zapoljarniy, the reverse is true, and this agrees with the emission values from the Zapoljarniy roasting plant (V. Pavlov, pers. comm.). In Kirakka, Naruska and Pallas, Cu and Ni concentrations are more similar.

In *snow filter residue* from all Russian catchments and in Skjellbekken and Kirakka, the Ni contents are clearly higher than respective Cu contents (Fig. 3.1.4b). The Pallas and Naruska catchments are at background level, with Ni content near or below analytical detection limit. The trends for Zapoljarniy, Skjellbekken and Kirakka are quite similar, clearly showing the «fingerprint» of the emission source, i.e. the plants of Zapoljarniy and Nikel. However, content of Cu and Ni is at least 100 times smaller in Norway and Finland than in Zapoljarniy and Monchegorsk on the Russian side.

## CONCLUSIONS

The Central Kola Expedition and the Geological Surveys of Norway and Finland have collected and analysed snow samples, both meltwater and filter residue, from eight catchments located in Russia (Zapoljarniy, Monchegorsk, Kirovsk, Kurka), Norway (Skjellbekken) and Finland (Kirakka, Naruska, Pallas). Many trace elements (As, Cd, Co, Cr, Cu, Mo, Ni, Pb, Pb, V and Zn), as well as S, Al and Fe, are strongly enriched in both meltwater and particulate phases in the Russian catchments, primarily in Monchegorsk and/or Zapoljarniy, compared with the other catchments. Typically, some elements are found mostly in the meltwater phase (e.g. Na, S, Sr, Zn), others in the filter residues (e.g. Cr, Fe, Mo, Ni). The details of this division, however, are specific of the type of industrial pollution source that dominates atmospheric chemistry in a particular area: for instance Cu and S are mostly in solution in Monchegorsk (smelting industry), but mostly particulate in Zapoljarniy (ore roasting industry). Cu and Ni are well correlated, both in meltwater and in filter residue, and the ratio of Cu to Ni is a useful indicator of pollution source. Snow is a well-suited medium for fingerprinting environmental pollution sources. For many elements (e.g., Fe), deposition estimates based on snow meltwater only will seriously underestimate deposition.

## ACKNOWLEDGEMENTS

We would like to thank the sampling teams from Finland, Norway and Russia, which consisted of Riitta Pohjola, Jouni Aarveaara, Tore Volden, Øystein Jæger and Vladimir Pavlov. The Norwegian participation in this project was financed by the Norwegian Ministry of the Environment with special project funds of the Norwegian Ministry of Foreign Affairs.

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## FIGURES

Figure 3.1.1. Location of study area for regional geochemical mapping (frame) and of the eight catchments discussed herein (1: Zapoljarniy, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, and 8: Pallas).

Figure 3.1.2. Boxplot diagrams for selected elements in snow meltwater (log units). C1-C8 refers to catchment numbers (see Fig. 1).

Figure 3.1.3. Boxplot diagrams for selected elements in snow filter residue (log units). C1-C8 refers to catchment numbers (see Fig. 1).

Figure 3.1.4. Scatter plot of Cu vs. Ni in snow meltwater (a) and snow filter residue (b). Note the Log scales. Open symbols: Russia; star: Norway; solid symbols: Finland.

*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
Catchment locations

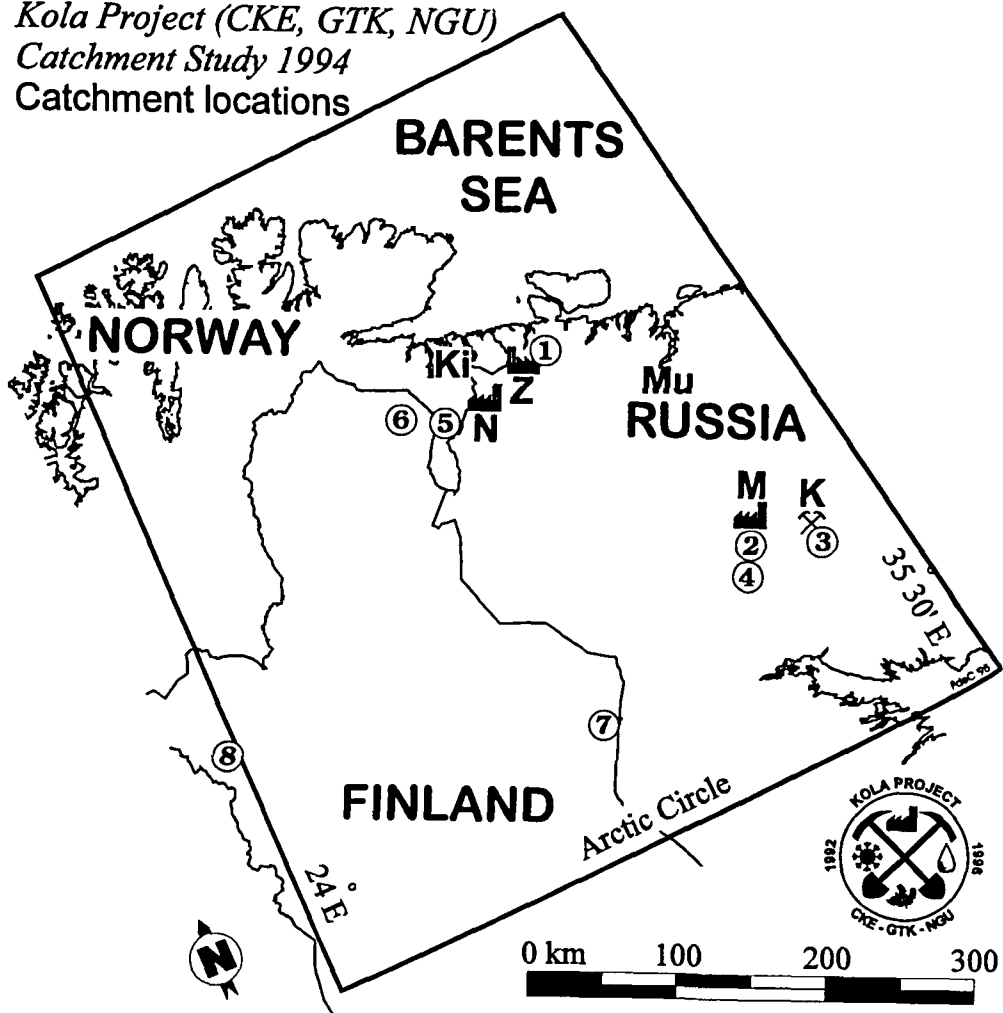


Figure 3.1.1. Location of study area for regional geochemical mapping (frame) and of the eight catchments discussed herein (1: Zapoljarniy, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, and 8: Pallas).



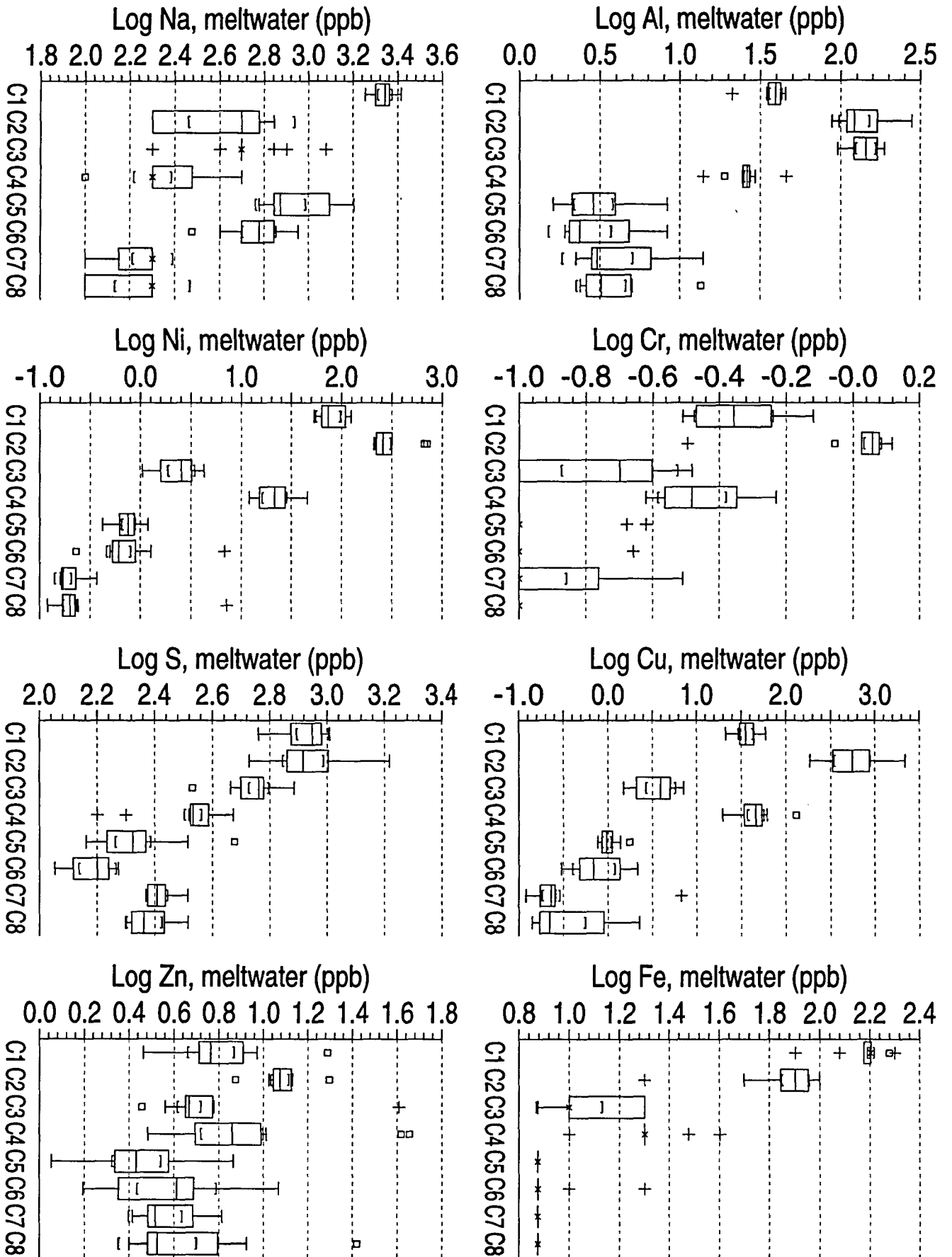


Figure 3.1.2. Boxplot diagrams for selected elements in snow meltwater (log units). C1-C8 refers to catchment numbers (see Fig. 1).

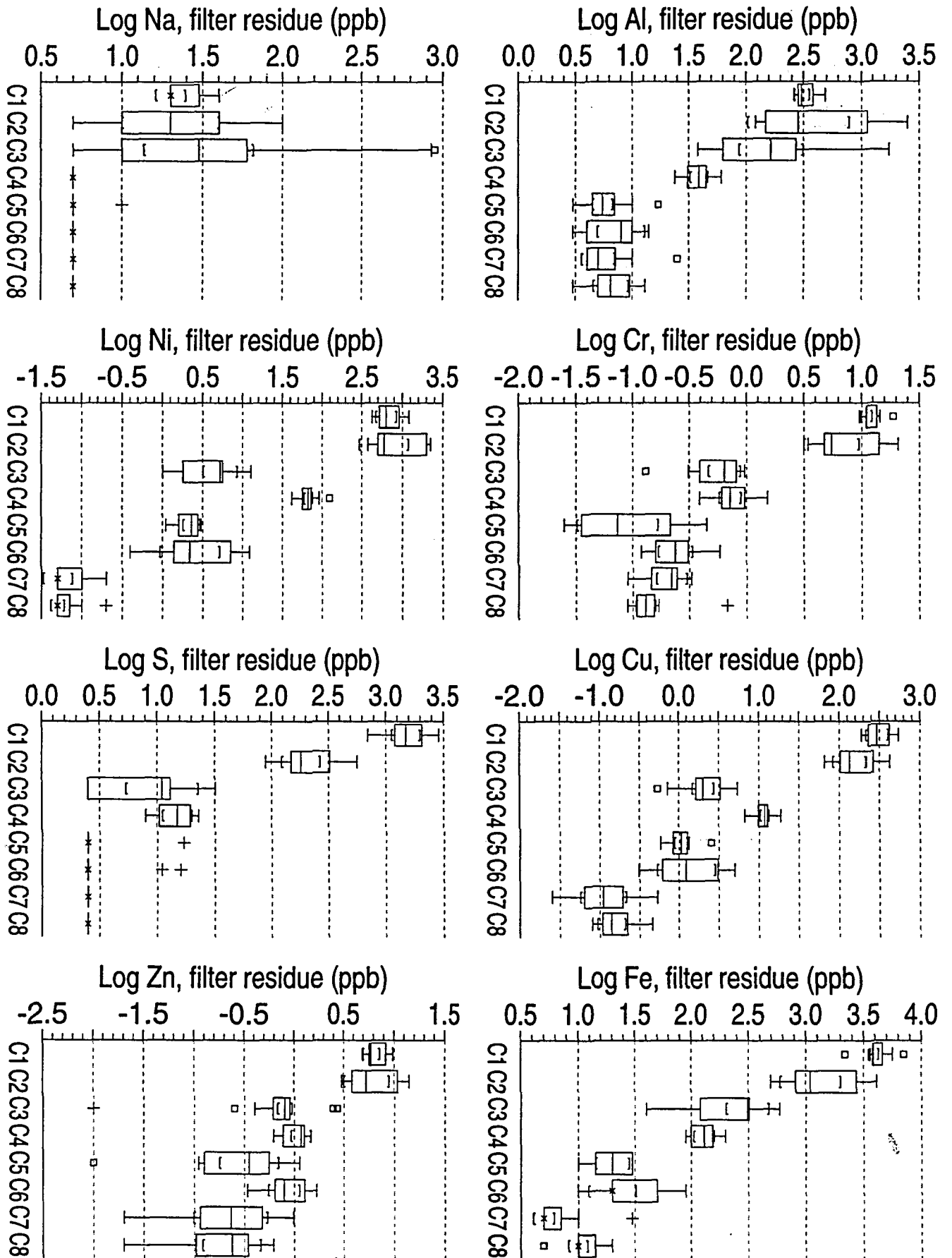


Figure 3.1.3. Boxplot diagrams for selected elements in snow filter residue (log units). C1-C8 refers to catchment numbers (see Fig. 1).

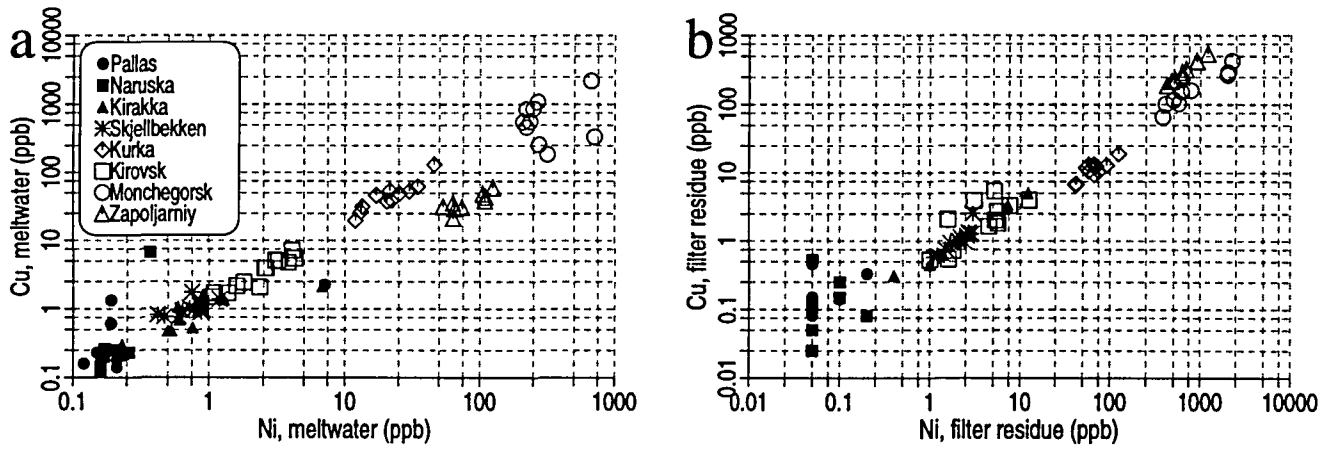


Figure 3.1.4. Scatter plot of Cu vs. Ni in snow meltwater (a) and snow filter residue (b). Note the Log scales. Open symbols: Russia; star: Norway; solid symbols: Finland.

### 3.2 Snow chemistry in eight catchment areas in the central part of the Barents Region: meltwater vs. filter residue

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#### ABSTRACT

Snowpack samples representing atmospheric deposition of the winter 1993-94 were taken in eight catchments at different distances to the industry on the Kola Peninsula, NW Russia, and in contiguous areas of Norway and Finland. About 3-5 kg of snow were collected from about 12 stations per catchment and delivered frozen to the laboratory. There, they were melted and filtered (<0.45 mm) on-line. Meltwater (MW) and filter residue (FR) fractions were analysed separately using ICP-MS and ICP-AES techniques.

The ratios of FR ("particular deposition") to MW ("wet deposition") concentrations show very characteristic patterns for the different industrial sources in the area. Nickel smelting at Monchegorsk results for example in predominantly water soluble deposition of Cu and S, while close to the ore roasting plant at Zapoljarniy these elements are mainly deposited in particulate form. For many elements more than 60% of the total deposition is particular in all catchments, demonstrating that analysing only the meltwater fraction of snow samples will seriously underestimate total deposition.

#### INTRODUCTION

Snow meltwater has been widely used to calculate and estimate anthropogenic deposition (Wright & Doveland, 1978, Vasilenko & Nazarov, 1985, Jeffries, 1989, Jaffe *et al.*, 1995, Lindroos *et al.*, 1995). The collection of snowpack samples is a very easy and cheap way to study element deposition. However, by analysing only the filtered meltwater, the total deposition is seriously underestimated (Reimann *et al.*, 1996). Snowpack samples are very well suited to fingerprinting large local emission sources, even at a very low sampling density (Reimann *et al.*, 1996). In Russia, intensive studies of the snowpack chemistry have been carried out in recent years (Makarova *et al.*, 1994, Jaffe *et al.*, 1995).

On the Kola Peninsula, there are several important sources of pollution, mainly Zapoljarniy, Nikel, Monchegorsk and Kirovsk (Fig. 3.2.1). In 1994, the emission of sulphur dioxide (SO<sub>2</sub>) from Zapoljarniy was 69 000 t, and about 98 000 t from Monchegorsk; the emissions of Ni were 160 t and 1600 t, respectively, and those of Cu 80 and 930 t (MRCENR, 1995, after Reimann *et al.*, *in prep.* a).

The Geological Surveys of Finland (GTK) and Norway (NGU) and the Central Kola Expedition (CKE) are presently undertaking a large environmental ecogeochemical mapping project in the western part of Kola Peninsula and in northern Finland and Norway (see World Wide Web site <http://www.ngu.no/Kola>). A pilot project, which took place in 1992-93 to harmonise field methods and to test various sampling media (including snow, see Reimann *et al.*, 1996), was followed by a catchment study in 1994-1995, and a regional geochemical mapping phase in 1995-1996. For the catchment study phase of interest here, eight catchments were selected in the project area (Fig. 3.2.1) for intensive study of element input and cycling

in the catchment areas. Media sampled include rain water (Reimann *et al.*, *in prep.* a), stream water (Caritat *et al.*, *in prep.* a,b), terrestrial moss (Äyräs *et al.*, *in prep.*), organic stream sediments (Pavlov *et al.*, *in prep.*), topsoil (Reimann *et al.*, *in prep.* b, Boyd *et al.*, *in prep.*) and soil profiles (Räisänen *et al.*, *in prep.*, Kashulina *et al.*, *in prep.*), in addition to snow (Äyräs *et al.*, 1995).

Four catchments are on the Kola Peninsula, representing very strongly to moderately polluted areas due to different causes: Zapoljarniy (C1) is near an open pit Cu-Ni ore mining and an ore concentration (roasting) plant, Monchegorsk (C2) is in the vicinity of a Cu-Ni ore smelter, Kirovsk (C3) is close to an open pit apatite mine and processing plant, and Kurka (C4) is located some 20 km S of Monchegorsk. Wind directions near the two main pollution point-sources (Monchegorsk and Nikel/Zapoljarniy) are dominantly to the N and S (Mäkinen, 1994). One catchment is in Norway, representing a slightly polluted area: Skjellbekken (C5). Three are in Finland, representing the background area: Kirakka (C6), Naruska (C7) and Pallas (C8). The sampling of the catchment study was carried out in 1994 and the analysis during winter 1994-95. Several different sampling media, including snowpack, were used in this part of project. The contents of sulphur and some trace metals in snowpack in these eight catchments have been discussed earlier (Äyräs *et al.*, 1995). In the present paper, we focus on the relationships between the element concentrations in snow meltwater (MW) and filter residue (FR) as a means to fingerprint different pollution sources.

## SAMPLING AND ANALYSIS

Snowpack sampling was carried out at the end of March and in the beginning of April 1994 in all countries. The samples are assumed to represent the whole winter fallout in the catchment areas. Seven to 13 samples were collected in each catchment with a colourless Plexiglas tube (Soveri, 1985), as detailed in the field manual (Äyräs & Reimann, 1995). The samples were kept frozen until arrival at the GTK laboratory. During storage, some of the samples partly melted and froze again, or leaked; these samples were discarded. The total number of samples analysed was 83.

The snow samples were melted and filtered (using Millipore® 0.45 mm membrane filters) on-line at the GTK laboratory. After measuring the pH and electrical conductivity, the filtered meltwater (MW) was divided into two aliquots. The first aliquot was acidified and analysed by ICP-AES (for Ca, Mg, Na, P, S, Si) and ICP-MS (for Ag, Al, As, B, Ba, Be, Bi, Cd, Co, Cr, Cu, Fe, K, Li, Mn, Mo, Ni, Pb, Rb, Sb, Se, Sr, Th, Tl, U, V, Zn). The second aliquot was analysed by potentiometry (for F) and by ion chromatography (for Br, Cl, NO<sub>3</sub>, SO<sub>4</sub>). After dissolution with nitric acid in a microwave oven (Niskavaara, 1995) the filter residue (FR) samples were analysed by ICP-AES (for Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, La, Li, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Sc, Si, Sr, Th, Ti, V, Y and Zn). The latter analytical results were recalculated to the total volume of snow meltwater to allow direct comparison with the meltwater results.

## RESULTS AND DISCUSSION

### *Element levels*

Median, mean and range values for the analysed parameters are given in Table 3.2.1 for the meltwater (MW) results, and in Table 3.2.2 for the filter residue (FR) results (median and mean values were calculated with results below detection limit set to half detection limit). The

variation of element concentrations generally is much greater between catchments than within catchment. The results show clearly that the fallout of the most harmful elements is high near the industrial areas of Kola Peninsula. The catchments in Finland represent background areas when examining the pollution from the Kola Peninsula.

#### *Copper/nickel ratios*

Figure 3.2.2a shows the variation in the Cu/Ni ratio in snow meltwater for the eight catchments studied; it is clear that C2 and C4 have higher median values for this ratio compared both with the other catchments in Russia, and those in Norway and Finland. C2 and C4 are the two watersheds most influenced by the anthropogenic emissions from the Monchegorsk smelter (Fig. 3.2.1). The great variability in the Cu/Ni ratio at C2 reflects that two types of ores are smelted in Monchegorsk: local ore with Cu/Ni of ca. 0.5, and Norilsk ore with Cu/Ni of 2 (see Reimann *et al.*, *in prep.* a). The significant point is that the emissions change with time, alternating from a low to a high Cu/Ni ratio; further transport downwind fractionates these elements and changes the ratio beyond the range of the original ore types. By the time the airborne contaminants reach C4, 25 km S of Monchegorsk, the median Cu/Ni ratio is still high, but the variability has decreased dramatically, as compared with C2 (5 km from Monchegorsk). C1, which is near another type of industry, namely ore concentrating, shows a much lower Cu/Ni in MW. Only local ore is concentrated here, and this corresponds very well with both the low Cu/Ni in snow MW (actually near 0.5, the value in the local ore), and the very tight box (very low variability, i.e. constancy in composition at the pollution source). The other catchments (C3, C5, C6, C7 and C8) have median Cu/Ni between 1 and 1.5, i.e. intermediate between the concentrating (0.5) and the smelting (>2) signatures. Therefore, Cu/Ni in MW is not a very sensitive indicator of pollution, since moderately polluted areas (C3 and C5, C6?) are indistinguishable from pristine ones (C7 and C8). Note the large natural variability in Cu/Ni snow MW in the most background watershed (C8).

To better discriminate between moderately polluted and unpolluted areas, we use the Cu/Ni ratio in the FR (Fig. 3.2.2b). Here, the smelter signature (C2 and C4) is around 0.2 (note the low variability even at C2), the concentration signature (C1) around 0.45, and progression toward more pristine areas give increasing Cu/Ni (FR) ratios, up to 2.2 at C8. So, with this ratio, the moderately contaminated to pristine areas are not constrained to values intermediate between the two main industrial processes, making this ratio a very useful one for identifying incipient pollution in remote areas (compare C6 and C7). Note, again, the large variability at C7 and C8, resulting presumably from the very low contents of Cu and Ni, and of many other trace elements typical of industrial emissions.

Having identified an element ratio diagnostic of industrial processes, Cu/Ni (MW), and one diagnostic for pristine areas, Cu/Ni (FR), we attempt to combine both to gain insight about both industrial fingerprinting and degree of 'pristineness' at once. This is done with the help of the two previous ratios divided by one another, namely  $\text{Cu/Ni (MW)} / \text{Cu/Ni (FR)}$ . This ratio of ratios may not be particularly intuitive at first, but Figure 3.2.2c shows that it is rather useful. Information about smelting and type of ore smelted is given by the high median values of this ratio (ca. 12-13, for both C2 and C4) and the variability or size of the boxes, respectively. Ore concentration (C1) has a very tight signature around 1. Intermediate pollution in this study area (C3, C5) yields values around 3-4. Progressively more pristine catchments have progressively lower ratio values, from 2 to 0.5 (C6 to C8).

#### *Filter residue and soluble elements*

The greatest advantage of the unusual method applied here to snow analysis is that we have obtained the composition of both the particulate (filter residue) and solute (meltwater) phases of the snowpack. This gives us the uncommon ability to compare the relative importance or concentration of elements in the two phases. One practical way to visualise this, as shown by Reimann *et al.* (1996), is to plot the ratio FR/(FR+MW), i.e. the relative importance of the FR to the total (FR+MW) composition. A low value of this ratio indicates predominance of the solute phase (dissolved elements); a high value means that the particulate phase (suspended solids >0.45 mm) is most important in the snowpack.

Figures 3.2.3a-h show, for a selection of elements, how the variation of this FR/(FR+MW) ratio looks for the eight catchments studied. In all catchments, the elements are ranked in the order of increasing importance of the filter residue in C2. At a glance, one can get an impression of how different or similar this ranking is in any catchment compared to C2 (by looking at how the median values deviate from the 'diagonal' for C2), and of how variable or constant the relative importance of solute and particulate phases are (by looking at the size of the boxes).

In C2, our henceforth arbitrary 'reference' catchment to study this ratio (Fig. 3.2.3b), several elements are typically more abundant in the melt water (see median values): Na, Cu (though the distribution is wide and skewed), S, Cd, Zn, Sr, Ca and K; others are characteristically dominant as solids: Fe, Mo, As, Cr, Ni, Al, Mn, Pb and Mg. For Ni, for instance, the median value of the ratio is ca. 0.7, meaning that 70% of the Ni is found as solid particles in the snowpack. For Fe this number exceeds 90%. This underscores the importance of taking filter composition into account when estimating deposition via snow analysis, not a standard practice up to now. Note that the boxes are quite large for C2, indicating a relatively great variability in the FR/(FR+MW) ratio, accounted for by the two types of ores smelted in nearby Monchegorsk (see above).

For C1 (Fig. 3.2.3a), under the influence of another industrial process (ore concentration) than the 'reference' catchment 2 (ore smelting), the solute/particulate distribution looks very different, firstly in the order of elements, secondly in the constancy of composition (box size). Elements typically present as solutes are Na, Ca, K and Sr, while those dominated by particles are V, Cr, Fe, As, Pb, Cu, Co, Ni, Al, Mo and Mg. Proximity to the ocean suggests that the more soluble elements are at least partly sourced from seaspray. All the main trace elements of the ore (Ni, Cu, Co) and several minor ones (V, Cr, As, Mo) as well as S are deposited primarily as solid particles, suggesting that a sizeable amount of sulphide particles are lost to the atmosphere during the concentrating process and redeposited in the nearby vicinity. The dominantly particular forms of Cu and Co (90% particular), S (65%) and V (98%) are significant, and potentially diagnostic, differences between airborne contamination emanating from ore concentration (C1) and smelting (C2, C4) processes. The generally tight boxes indicate constancy in the industrial source supply, while wider boxes can be diagnostic of the presence of another source (e.g., seaspray for S).

In C3 (Fig. 3.2.3c) the boxes are large indicating great variability in the FR/(FR+MW) ratio due to different sources, mining and smelting. Elements present as solutes are S, Na, Zn and K and those mainly in particulate form Sr, Ba, Co, Pb, Ni, Cr and especially Fe, of which almost all is in particulate form. Note that S is dominantly as solute, not accounted for by mining activity but by oil burning.

The ratios of FR/(FR+MW) in C4 (Fig. 3.2.3d) resemble loosely those in C2, suggesting a similar contamination source, namely the Monchegorsk smelting industry. Typically abundant in the meltwater in C4 are Na, S, Zn, Sr, Mg, Cu, Ca, K and V. Elements characteristically occurring as solids include Fe, Ni, Mo, Cr and Co. Based on the comparison of median values and confidence notches between C2 and C4, the most significant differences are for As, Mg, Mn, S, Sr, Zn, Cr and Mo, which are more soluble in C4 than in C2. However, the main sulphide metals Cu, Cd, Co and Ni are distributed similarly (95% confidence) between the solid and aqueous phases in the two catchments. This means that most elements have become relatively more abundant in the soluble phase with increasing distance from source, due to preferential deposition of solid particles close to the smelter; the relative importance of the deposition of solid particles tapers off away from the smelter. However, Cu, Cd, Co and Ni, despite dropping in abundance by one order of magnitude between C2 and C4 (see Tabs. 3.2.1 and 3.2.2), have a constant distribution between wet and dry deposition, *regardless of distance*. Cu from Monchegorsk remains principally (ca. 80%) soluble, Ni principally (ca. 70%) solid. This has important implications for *fingerprinting emission sources* at various distances.

For C5 (Fig. 3.2.3e) the elements Ni, Fe, Al and Co are dominantly in particulate form. The source of these elements is clearly the smelter of Nikel, distance being about 30 km. Elements typically more abundant in the meltwater are Na, S, Sr, Mg, K, Zn, Mn and V. Seaspray is likely to be an important source of e.g. Na, S and Mg. The high particular proportions of Cu (50%) and Co (65%) suggest some influence of the ore roasting (concentrating) plant in Zapoljarniy (near C1), though the signature is complicated by the emissions from the ore smelter in nearby Nikel.

For C6 (Fig. 3.2.3f) the elements Co, Ni, As, Fe, Cr, Al and Cu dominantly occur as solids and thus resemble quite closely C5, having moderate pollution from same sources (Zapoljarniy and Nikel). Elements typically more abundant in the meltwater are Na, S, Sr, Mg, Zn and K. Some of these elements (Na, S, Mg) originate from the Barents sea, and are blown onto land as seaspray during storms. This catchment represents transition zone between polluted areas and background.

The ratios FR/(FR+MW) for the background catchments C7 (Fig. 3.2.3g) and C8 (Fig. 3.2.3h) are very similar and differ totally from our reference catchment (C2). In both the elements Cd, Al, Cr and Mo occur as solids, for elements Ca and Fe the ratios are quite close to 0.5 showing that these elements are in equal amounts both in solute and solid phases. Dominantly as solutes are Na, S, Zn and V. Looking at Ni, for instance, which occurs mainly as a soluble element (20-25% particular) in both C7 and C8, the signatures from either contamination sources, i.e. ore smelting (70% particular) and ore roasting (90% particular) have disappeared in these background areas. Absolute concentrations of many elements are also the lowest of all catchments in C7 or C8 (see Tabs. 3.2.1 and 3.2.2). Thus we have reached as pristine background levels as there are known to exist within Europe (see table 3.2.2 in Reimann *et al.*, 1996).

## CONCLUSIONS

Snowpack cores were collected at the end of winter 1993-94 in eight selected catchments (C1 to C8) of the Barents Region. Snow meltwater (MW) and filter residue (FR) were separated by filtration (0.45 mm) and analysed independently using modern methods, including ICP-



MS, for up to 43 parameters (Ag, Al, As, B, Ba, Be, Bi, Br, Ca, Cd, Cl, Co, Cr, Cu, F, Fe, K, La, Li, Mg, Mn, Mo, Na, Ni, NO<sub>3</sub>, P, Pb, Rb, S, Sb, Sc, Se, Si, SO<sub>4</sub>, Sr, Th, Tl, U, V, Y, Zn; pH and EC).

The highest values of heavy metals and sulphur in snow are found in the catchments situated on the western Kola Peninsula, in Russia (C1-C4). C5 in Norway and, to some extent, also C6 in Finland, near the Norwegian border, are affected marginally by pollution from industrial areas of the Kola Peninsula. C7 and C8 represent unaffected background areas with respect to these anthropogenic sources. Thus, a comparison of snow composition is afforded between some of Europe's most contaminated areas when it comes to industrial airborne pollution, and some of the most pristine background areas to be found here.

There are marked differences in concentrations in snow MW and FR. For instance, Fe and Mo were found predominantly in the solid phase (FR) in most catchments. When estimating and calculating total loading (deposition) from pollution sources onto terrestrial ecosystems, both MW and FR should thus be analysed.

The Cu/Ni ratio in snow MW shows clearly the fingerprint from the sources and those fingerprints are seen quite far away from the smelters. On the other hand, by the Cu/Ni ratio is not a very sensitive indicator of pollution, since moderately polluted areas are indistinguishable from pristine ones. Using the Cu/Ni ratio in snow FR is very useful for identifying pollution in remote areas.

The relative importance of the filter residue to the total composition of snow, expressed as the ratio FR/(FR+MW), allows distinction of different industrial processes between catchments. For instance, ore smelting at Monchegorsk has a signature characterised in C2 by a low value of this ratio for Cu (15%), and high value for both Ni (70%) and As (80%). These ratios are constant with distance from the smelter in the case of Cu and Ni, as shown by results of C4; the value for As, however drops dramatically (to 30%). Ore roasting (concentration) at Zapoljarniy, near C1, has a distinct difference: Cu is predominantly solid (90%). Thus, the FR/(FR+MW) ratio is a powerful tool to: (1) distinguish different pollution sources (e.g. ore smelting vs. ore roasting), and (2) infer processes happening during airborne transport of contaminants (e.g. deposition of particular As).

#### ACKNOWLEDGEMENTS

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## TABLES

Tab. 3.2.1 Median and mean, minimum and maximum values of parameters measured in snow meltwater

Tab. 3.2.2 Median and mean, minimum and maximum values of parameters measured in snow filter residues

## FIGURES

Figure 3.2.1. Location of the regional mapping project area (frame) in NE Norway, N Finland and NW Russia, with its eight catchments (C1: Zapoljarniy, C2: Monchegorsk, C3: Kirovsk, C4: Kurka, C5: Skjellbekken, C6: Kirakka, C7: Naruska, C8: Pallas) and the major towns of Kirkenes (Ki: open-pit iron mine and mill), Nikel (N: Ni-Cu smelter), Zapoljarniy (Z: open-pit Ni-Cu mine and ore roasting), Murmansk (M), Kirovsk (K: open-pit apatite mine).

Figure 3.2.2. Boxplot representation of the Cu/Ni ratio of snow meltwater (a), filter residue (b), and the relation between the two (c) in the eight catchments (C1-C8). For clarity, the outliers are not shown. Boxplots constructed after Velleman & Hoaglin (1981).

Figure 3.2.3. Boxplot representation of the relative importance of the filter residue (FR) relative to total concentration (FR+MW) of selected elements in C1 (a), C2 (b), C3 (c), C4 (d), C5 (e), C6 (f), C7 (g), C8 (h). A low value (e.g. 0.1) of the ratio FR/(FR+MW) implies that the filter residue contributes little (e.g. 10%) to the total concentration (and deposition) of a particular element, and vice versa. Elements are ranked in all figures in order of increasing median value for C2.

Tab. 3.2.1 Median and mean, minimum and maximum values of parameters measured in snow meltwater

	Detect. Limit	C1	C2	C3	C4	C5	C6	C7	C8
Ag	0.01	<0.01	<0.01	<0.01	0.0135	<0.01	<0.01	<0.01	<0.01
		<0.01	<0.01	<0.01	0.04	<0.01	<0.01	<0.01	<0.01
Al	1	39.4	38.07	121.5	146.94	144	144.62	26.2	26.39
		21.2	45.4	88	279	96.1	189	14	46
As	0.1	0.13	0.11	1.045	1.013	0.34	0.35	0.44	0.45
		<0.1	0.16	0.18	1.76	<0.1	0.77	0.17	0.81
B	0.5	<0.5	0.28	<0.5	0.29	<0.5	0.29	<0.5	<0.5
		<0.5	0.55	<0.5	0.65	<0.5	0.73	<0.5	<0.5
Ba	0.1	0.57	0.54	1.875	1.94	1.65	1.73	0.71	0.84
		0.31	0.74	1.45	2.71	1.23	3.01	0.49	1.34
Be	0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3
		<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3
Bi	0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
		<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Br	200	<200	<200	<200	<200	<200	<200	<200	<200
		<200	<200	<200	<200	<200	<200	<200	<200
Ca	50	420	416.67	190	201	220	220	110	103.64
		310	540	150	300	110	350	<50	170
Cd	0.03	0.14	0.15	0.315	0.371	0.07	0.08	0.08	0.12
		0.1	0.25	0.24	0.76	<0.03	0.21	<0.03	0.35
Cl	100	3900	3944	1400	1220	700	746.2	600	572.7
		3200	4800	500	1700	300	1900	300	900
Co	0.03	2.59	3.03	32.95	31.984	0.14	0.15	2.45	2.80
		2.03	4.25	9.74	53	0.08	0.24	1.18	7.69
Cr	0.2	0.44	0.47	1.145	1.054	0.2	<0.2	0.33	0.36
		0.31	0.76	0.32	1.31	<0.2	0.33	0.24	0.59
Cu	0.05	35.6	37.68	555	734.1	3.93	3.62	46.2	50.02
		21.2	59.7	186	2190	1.49	7.17	19.7	131
F	50	<50	<50	<50	<50	<50	<50	<50	<50
		<50	<50	<50	<50	<50	<50	<50	<50
Fe	15	160	152.2	80	74	<15	<15	20	21.82
		80	200	20	100	<15	20	<15	40
K	10	120	116.7	85	91	130	160.8	50	55.45
		90	140	60	130	70	420	30	100
Li	0.1	0.2	0.20	0.185	0.184	<0.1	<0.1	<0.1	0.135
		0.14	0.3	<0.1	0.29	<0.1	0.14	<0.1	<0.1
Mg	50	440	425.56	105	90	<50	49.615	<50	42.273
		350	490	<50	130	<50	130	<50	70
Mn	0.03	4.71	5.31	3.355	3.605	2.35	3.03	2.02	1.97
		3.98	7.9	2.51	5.72	1.56	7.95	1	3.31
Mo	0.05	<0.05	<0.05	0.23	0.237	<0.05	<0.05	0.06	0.05
		<0.05	<0.05	0.09	0.35	<0.05	<0.05	<0.05	0.09
Na	15	2200	2133	500	440	500	538.5	200	263.6
		1800	2600	200	700	200	1200	100	500
Ni	0.1	73.7	85.36	258	336.9	2.54	2.52	21.5	23.23
		53.6	125	209	708	1.05	4.27	12	45.8
P	100	<100	<100	<100	<100	<100	<100	<100	<100
		<100	<100	<100	<100	<100	<100	<100	<100
Pb	0.03	0.08	0.09	3.375	3.378	0.45	0.76	1.24	1.40
		0.03	0.3	0.92	6.27	0.07	2.25	0.87	2.44
Rb	0.05	0.13	0.13	0.195	0.217	0.52	0.73	0.12	0.12
		0.11	0.18	0.18	0.33	0.26	2.11	0.05	0.17
S	50	890	840	825.5	891.1	576	566.1	341	343.5
		576	1020	534	1650	339	768	159	470
Sb	0.03	<0.03	<0.03	0.055	0.049	<0.03	<0.03	<0.03	<0.03
		<0.03	<0.03	<0.03	0.07	<0.03	<0.03	0.05	<0.03
Se	0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
		<0.5	<0.5	<0.5	0.54	<0.5	<0.5	<0.5	<0.5
Sr	0.1	2.47	2.47	3.805	3.66	6.91	7.69	1.42	1.48
		2.01	3.26	1.98	5.08	4.5	12.6	0.89	2.61
Th	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
		<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Tl	0.01	<0.01	<0.01	0.02	0.02	<0.01	0.01	<0.01	0.01
		<0.01	<0.01	0.01	0.04	<0.01	0.01	<0.01	0.01
U	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
		<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
V	0.05	0.06	0.09	15.7	15.691	2.58	2.77	2.09	2.32
		0.05	0.14	4	24.2	0.33	6.44	0.91	4.69
Zn	0.3	5.8	7.55	11.9	12.441	4.65	7.58	7.23	13.11
		2.91	19.4	7.51	19.8	2.87	40.6	3.05	45.1
pH		4.96	4.97	4.595	4.609	4.72	4.76	4.53	4.55
		4.86	5.09	4.38	4.83	4.33	5.04	4.38	4.83
EC		24.5	24	21.35	20.08	15.5	15.76	16.5	15.36
		20.6	28.2	11	24.7	12.1	21	9.1	20.5

Tab. 3.2.2 Median and mean, minimum and maximum values of parameters measured in snow filter residues

Detect. Limit	C1	C2	C3	C4	C5	C6	C7	C8									
Al	304 263	335.44 486	285 121	686.1 2500	164 38	423.85 1730	39 24	39.82 61	5.5 3	6.67 17	8 3	7.56 14	5 4	8.14 25	6.5 3	7.25 13	
As	0.3	3.6 1.4	3.48 5.9	4.65 2.5	6.53 16.9	<0.3 <0.3	<0.3 0.6	<0.3 <0.3	<0.3 0.5	<0.3 <0.3	<0.3 <0.3	<0.3 0.6	<0.3 <0.3	<0.3 0.3	<0.3 <0.3	<0.3 0.3	
B	0.3	0.4	0.41	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	
Ba	0.1	<0.3 0.91	0.8 0.85	<0.3 1.96	0.4 2.63	<0.3 2.51	0.4 5.70	<0.3 0.46	<0.3 0.56	<0.3 <0.1	<0.3 <0.1	<0.3 <0.1	<0.3 0.27	<0.3 0.15	<0.3 0.19	<0.3 0.125	<0.3 0.12
Ca	100	<100 <100	<100 120	256 840	150 330	<100 1350	<100 1350	<100 1350	<100 1350	<100 1350	<100 1350	<100 1350	<100 1350	<100 1350	<100 1350	<100 1350	
Cd	0.05	0.1 0.05	0.12 0.21	0.095 0.05	0.136 0.34	<0.05 <0.05	<0.05 0.05	<0.05 <0.05	<0.05 <0.05	<0.05 <0.05	<0.05 <0.05	<0.05 0.08	<0.05 <0.05	<0.05 <0.05	<0.05 <0.05	<0.05 <0.05	
Co	0.05	22.3 15.1	24.87 41.3	46.45 30.4	54.13 104	0.31 0.07	0.30 0.68	3.75 2.52	4.14 8.64	0.095 <0.05	0.08 0.12	0.09 0.44	0.14 <0.05	<0.05 <0.05	<0.05 <0.05	<0.05 <0.05	
Cr	0.05	11 9.49	12.27 18.7	5.5 3.44	9.039 20.7	0.64 0.13	0.63 0.96	0.72 0.39	0.83 1.53	0.075 <0.05	0.14 0.45	0.24 0.12	0.28 0.59	0.22 0.09	0.20 0.33	0.13 0.09	0.195 0.68
Cu	0.05	293 188	321.89 541	135.5 66	179.8 426	2.01 0.53	2.36 5.42	11.9 6.75	11.51 19	1.06 0.58	1.13 2.53	1.23 0.31	1.82 5.03	0.11 <0.05	0.17 0.53	0.14 0.08	0.19 0.46
Fe	10	3860 2180	4291 7040	1110 500	1716 4150	310 40	264.6 590	130 90	134.5 200	20 10	20.83 30	20 90	33.33 10	<10 <10	<10 30	10 <10	11.88 20
K	10	30 10	25.56 30	55 30	53 80	30 <10	131.2 690	20 <10	16.82 40	<10 <10	15.83 60	<10 <10	<10 20	10 <10	10.71 20	<10 <10	<10 10
La	0.07	0.18 0.14	0.19 0.26	0.37 0.16	0.552 1.38	1.05 0.21	2.33 8.97	<0.07 <0.07	<0.07 0.11	<0.07 <0.07	<0.07 <0.07	<0.07 <0.07	<0.07 <0.07	<0.07 <0.07	<0.07 <0.07	<0.07 <0.07	
Li	0.07	0.23 0.15	0.23 0.32	0.145 0.07	0.234 0.74	0.13 <0.07	0.12 0.22	<0.07 <0.07	<0.07 0.07	<0.07 <0.07	<0.07 <0.07	<0.07 <0.07	<0.07 <0.07	<0.07 <0.07	<0.07 <0.07	<0.07 0.08	
Mg	10	1490 1140	1619 2540	105 40	192 540	30 <10	25.77 50	10 <10	12.27 50	<10 <10	<10 <10	<10 20	<10 <10	<10 <10	<10 <10	<10 <10	
Mn		10.7 8.98	11.20 16.4	4.87 2.18	10.255 31.9	2.46 0.66	5.57 19.1	0.64 0.44	0.72 1.22	0.125 0.06	0.125 0.21	0.14 0.07	0.17 0.35	0.1 0.08	0.14 0.41	0.16 0.07	0.155 0.22
Mo	0.07	0.11 <0.07	0.12 0.19	1.49 0.93	1.902 4.36	<0.07 <0.07	<0.07 0.11	0.14 0.09	0.15 0.26	<0.07 <0.07	<0.07 <0.07	<0.07 <0.07	<0.07 0.15	<0.07 <0.07	<0.07 0.09	<0.07 <0.07	
Na	10	20 20	26.67 40	20 <10	32 100	30 <10	170.4 890	<10 <10	<10 <10	<10 <10	5.417 10	<10 <10	<10 <10	<10 <10	<10 <10	<10 <10	
Ni	0.1	627 423	708 1200	595 380	1000.7 2230	5.2 1	4.72 12.7	66.8 41.5	68.39 124	2.3 1.1	2.18 3	2.2 0.4	4.07 12.3	<0.1 <0.1	0.09 0.2	<0.1 <0.1	0.075 0.2
P	0.5	14.7 4.5	14 17.6	28.4 14.9	40.52 84.7	72.9 13.2	145.9 578	4.6 2.3	4.255 6.7	0.9 <0.5	1.192 5	0.9 2.2	1.022 2.2	0.8 <0.5	0.971 2.8	1.05 <0.5	1.169 2
Pb	0.3	1.7 1.3	1.71 2.2	5.05 3.1	7.28 17.8	1.5 0.4	1.26 2	1.1 0.6	1.06 1.8	<0.3 <0.3	<0.3 <0.3	<0.3 <0.3	<0.3 1.4	<0.3 <0.3	<0.3 <0.3	<0.3 0.4	
S	5	1490 703	1626 2870	181.5 89	234.5 568	11 <5	11.62 32	15 8	14.64 23	<5 <5	<5 17	<5 <5	<5 16	<5 <5	<5 <5	<5 <5	
Sb	0.5	0.6 <0.5	0.68 1.4	0.65 <0.5	0.93 4	<0.5 <0.5	<0.5 0.9	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 0.9	<0.5 <0.5	
Sc	0.01	0.12 0.11	0.13 0.19	0.07 0.04	0.154 0.52	0.02 <0.01	0.02 0.04	0.01 0.01	0.01 0.02	<0.01 <0.01	<0.01 <0.01	0.01 0.02	<0.01 <0.01	0.01 0.01	<0.01 <0.01	0.01 0.01	
Si	10	50 <10	41.67 70	20 20	28 40	10 <10	18.85 50	10 <10	10 20	<10 <10	<10 <10	<10 11.67	<10 <10	10 30	<10 <10	<10 20	
Sr		0.49 0.48	0.58 0.76	2.285 1.24	3.314 7.5	10.9 2.08	28.53 120	0.23 0.16	0.23 0.37	0.02 0.02	0.04 0.13	0.03 0.02	0.03 0.04	0.03 0.02	0.03 0.07	0.035 0.02	0.035 0.05
Th	0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 0.8	<0.5 <0.5	<0.5 1.1	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	<0.5 <0.5	
Ti		22.2 16.1	22.6 30.6	31.05 15.1	51.19 139	27.3 5.68	25.59 53	3.71 2.39	3.68 5.17	0.66 0.31	0.71 1.57	0.82 0.43	0.93 2.15	0.43 0.34	0.86 3.41	0.595 0.39	0.79 1.71
V	0.05	4.77 3.83	5.44 8.73	18.35 6.46	23.356 56.7	1.44 0.27	1.51 3.46	0.64 0.45	0.67 0.95	<0.05 <0.05	<0.05 0.06	0.05 <0.05	0.05 0.12	<0.05 <0.05	<0.05 0.09	<0.05 <0.05	
Y	0.01	0.11 0.09	0.11 0.15	0.12 0.06	0.233 0.7	0.19 0.04	0.42 1.54	0.02 0.01	0.02 0.03	<0.01 <0.01	0.01 0.01	<0.01 <0.01	0.01 0.02	<0.01 <0.01	0.01 0.01	<0.01 <0.01	
Zn	0.02	5.93 4.96	6.73 9.68	5.375 3.04	6.819 14.1	0.8 <0.02	1.07 2.7	1.17 0.62	1.04 1.47	0.355 <0.02	0.41 1.13	0.79 0.34	0.91 1.68	0.23 0.02	0.35 0.98	0.245 0.02	0.25 0.62

*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
 Catchment locations

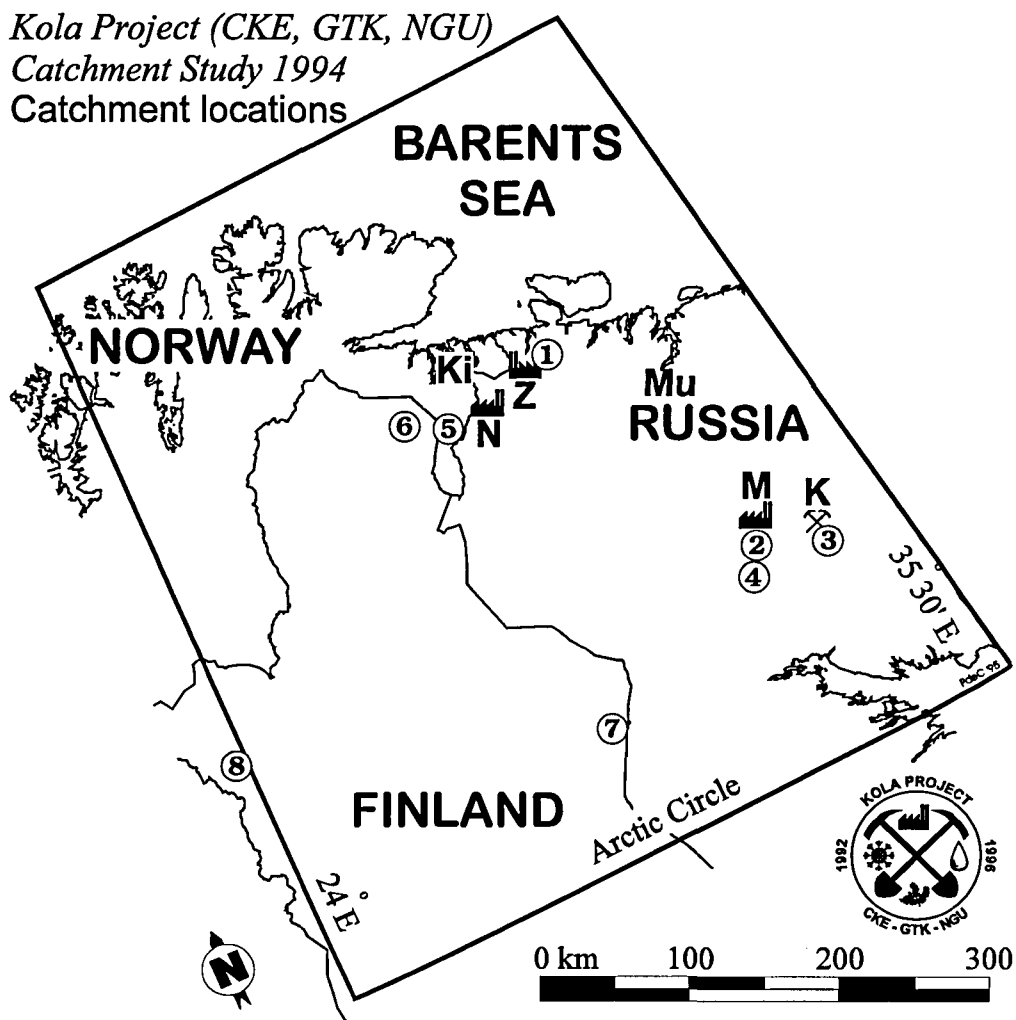


Figure 3.2.1. Location of the regional mapping project area (frame) in NE Norway, N Finland and NW Russia, with its eight catchments (C1: Zapoljarniy, C2: Monchegorsk, C3: Kirovsk, C4: Kurka, C5: Skjellbekken, C6: Kirakka, C7: Naruska, C8: Pallas) and the major towns of Kirkenes (Ki: open-pit iron mine and mill), Nickel (N: Ni-Cu smelter), Zapoljarniy (Z: open-pit Ni-Cu mine and ore roasting), Murmansk (M), Kirosvk (K: open-pit apatite mine).

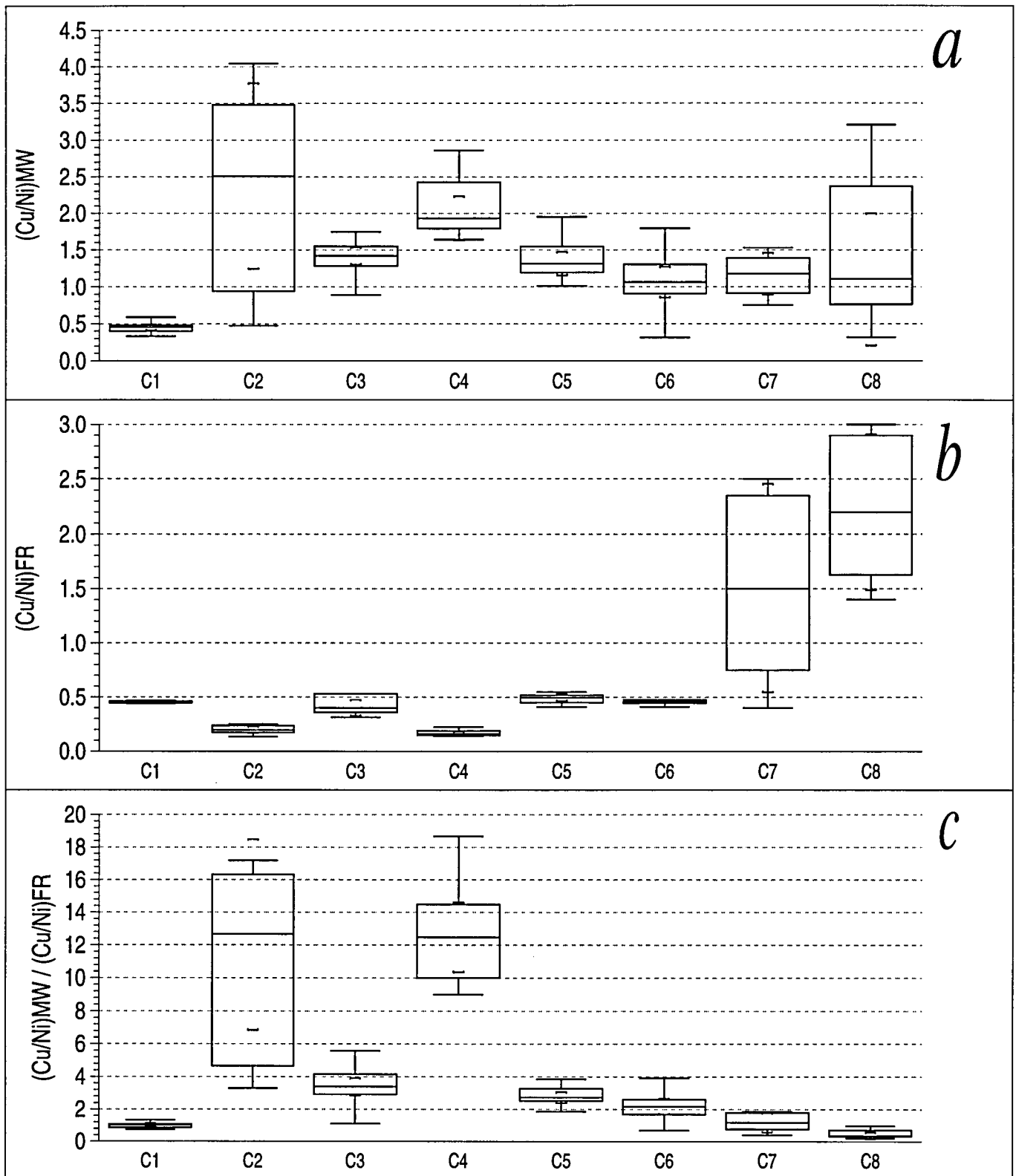


Figure 3.2.2. Boxplot representation of the Cu/Ni ratio of snow meltwater (a), filter residue (b), and the relation between the two (c) in the eight catchments (C1-C8). For clarity, the outliers are not shown. Boxplots constructed after Velleman & Hoaglin (1981).

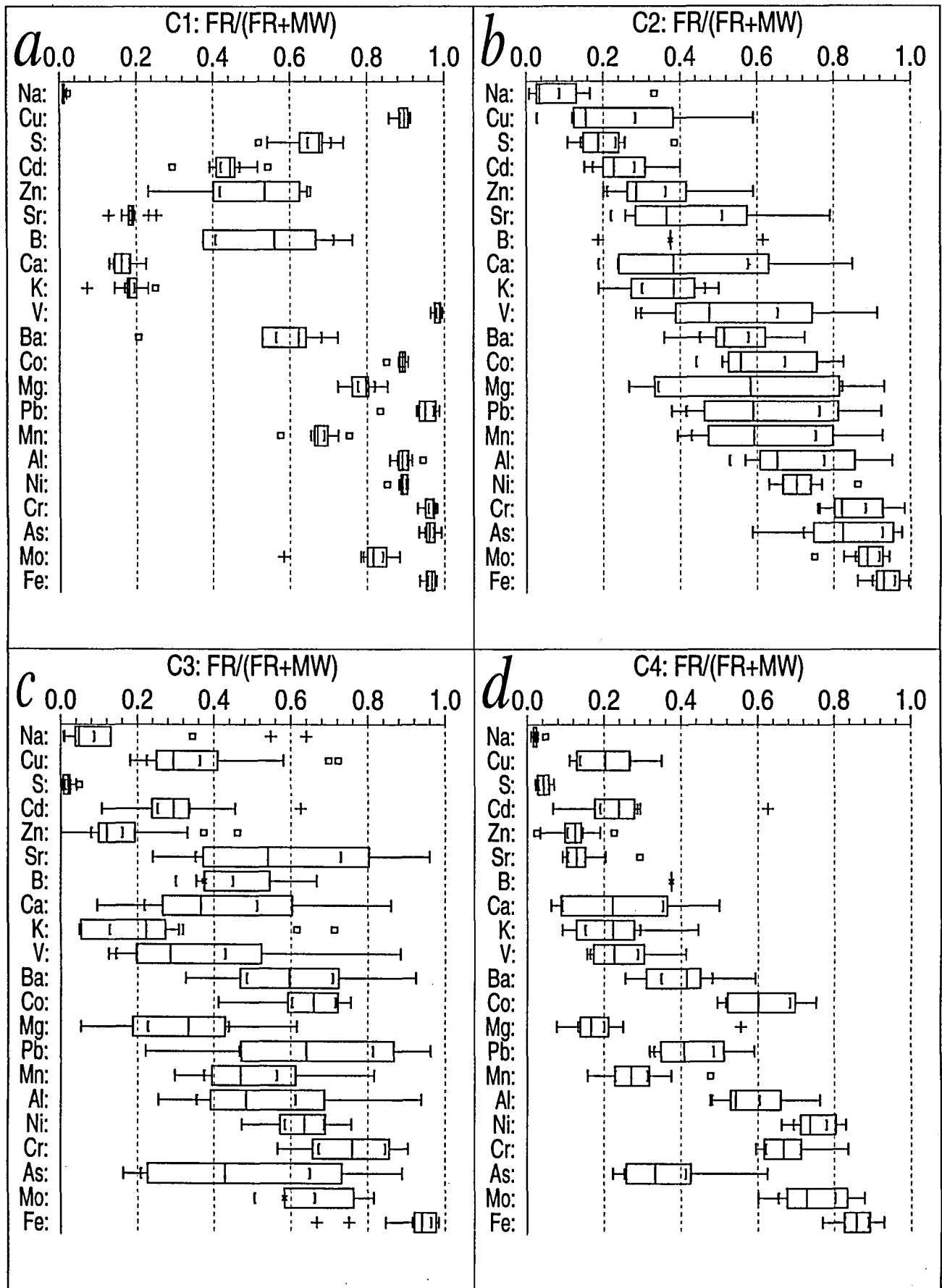
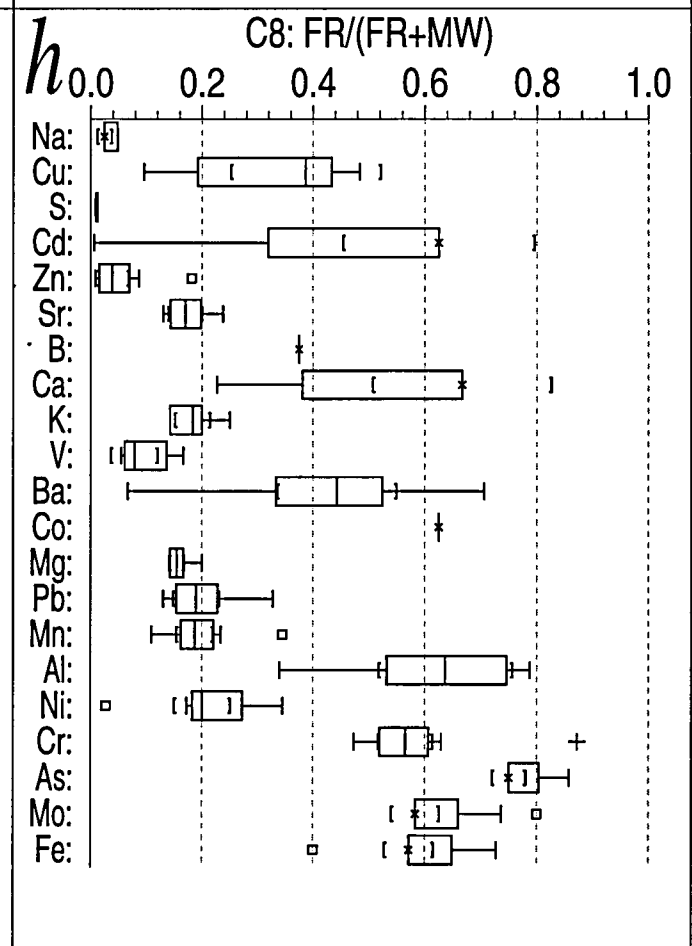
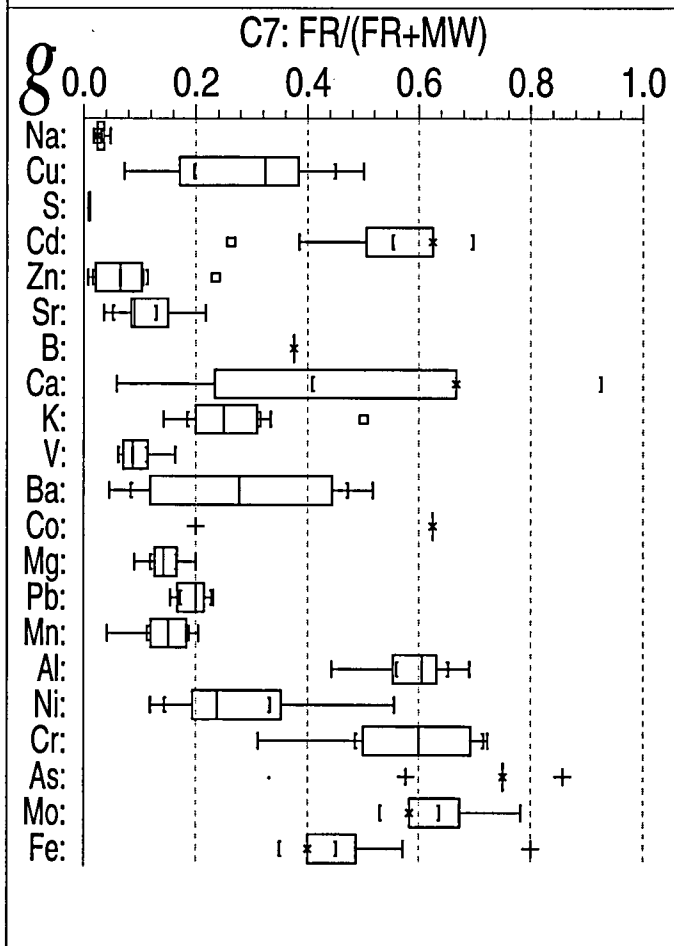
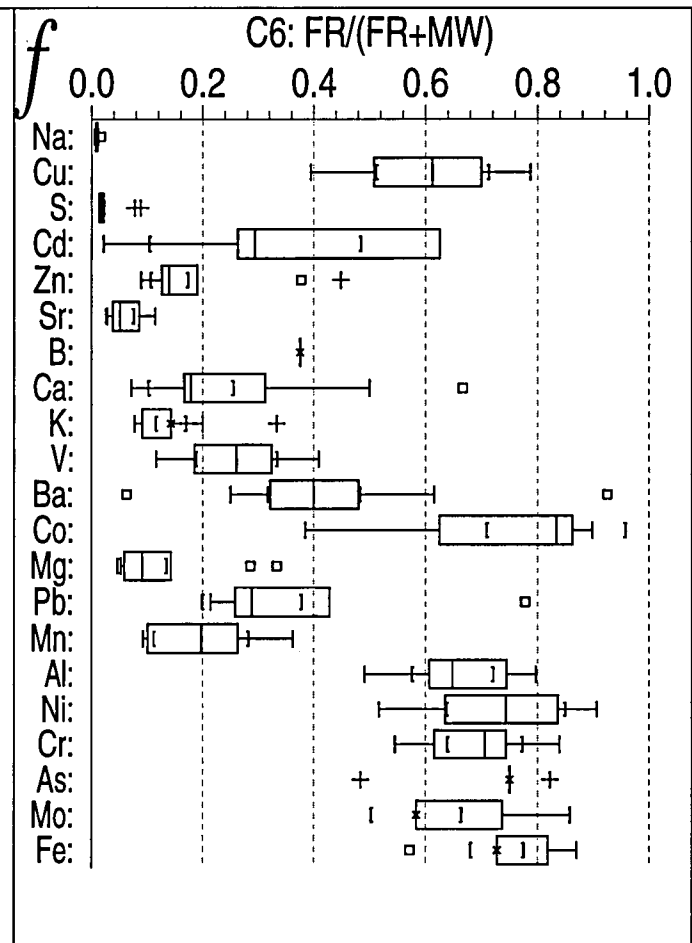
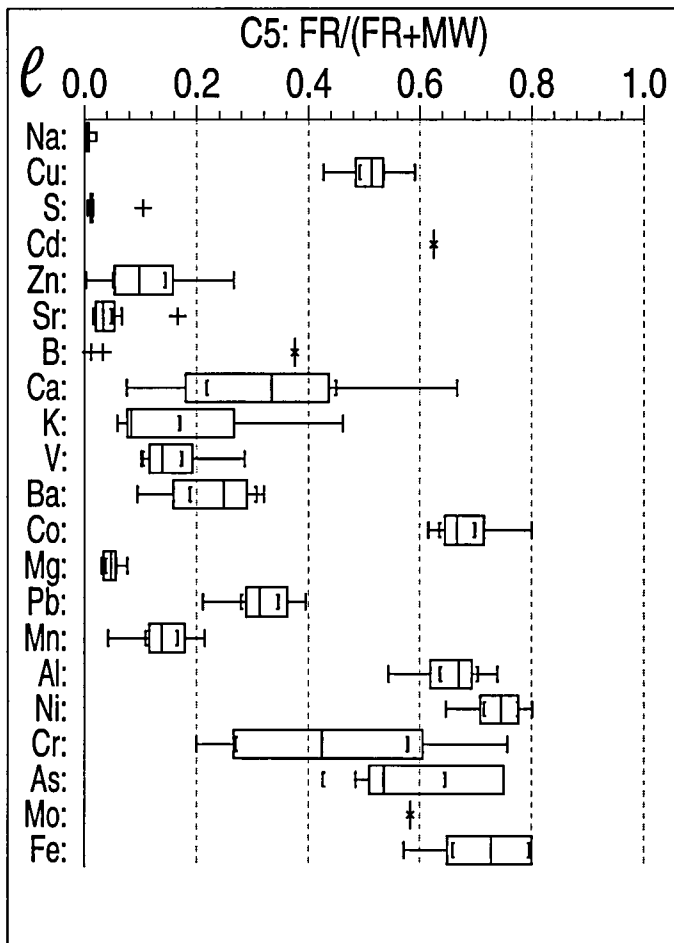


Figure 3.2.3. Boxplot representation of the relative importance of the filter residue (FR) relative to total concentration (FR+MW) of selected elements in C1 (a), C2 (b), C3 (c), C4 (d), C5 (e), C6 (f), C7 (g), C8 (h). A low value (e.g. 0.1) of the ratio FR/(FR+MW) implies that the filter residue contributes little (e.g. 10%) to the total concentration (and deposition) of a particular element, and vice versa. Elements are ranked in all figures in order of increasing median value for C2.





### **3.3 Rainwater composition in eight Arctic catchments in northern Europe (Finland, Norway and Russia)**

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#### **ABSTRACT**

Monthly rainwater samples were collected during the summer of 1994 in eight arctic catchments in northern Europe (4 in Russia, 3 in Finland, 1 in Norway), at different distances and wind directions from the emissions of the Russian nickel ore mining, roasting and smelting industry on the Kola Peninsula. Three stations consisting of 5 samplers each were placed in open areas in all the catchments.

Results show that close to the smelters in Monchegorsk, rainwater is strongly enriched in Ni (633x), Co, Cu, As, Mo, Al (36x), V, Cd, Sb, Pb (11x), Zn, Fe, Sr, Na, S/SO<sub>4</sub> (6x), Cl, Cr, Se (4x) and Ag when compared to a Finnish background catchment. Three sources of elements can be differentiated: natural dust, sea spray and anthropogenic (smokestack emissions and dust). Correlation diagrams and element ratios can be used to identify the different industrial processes and even ore feed changes at one smelter.

#### **INTRODUCTION**

The geological surveys of Finland (GTK) and Norway (NGU) and the Central Kola Expedition (CKE) in Russia are carrying out a major geochemical mapping project (see World Wide Web site <http://www.ngu.no/Kola>) in a 188,000 km<sup>2</sup> area north of the Arctic Circle, comprising the entire area between 24° and 35.5° E north to the Barents Sea (Fig. 3.3.1). As part of this project, eight catchments (hereafter abbreviated as C1-C8) widely distributed in this area (Fig. 3.3.1) were investigated in detail in 1994. Sampled media were: snow (meltwater and filter residue), rainwater, stream water, organic stream sediments, terrestrial moss, topsoil (0-5 cm), complete podzol profiles, Quaternary deposits and bedrock.

Some of the world's largest point sources of SO<sub>2</sub> emissions (Gunn et al., 1995) are located within the study area, the nickel smelter at Nikel, the ore roasting plant at Zapoljarnij and the nickel smelter at Monchegorsk (Fig. 3.3.1) together accounting for about 400,000 t of SO<sub>2</sub> emissions yearly. Table 3.3.1 gives an overview of the emission data as released by Russian authorities for all major sources in the survey area for 1993 and 1994.

The results of rainwater sampling carried out during the summer months of 1994 are presented here. A number of established rainwater monitoring stations exist in the area (e.g. at Svanvik in Norway (Hagen et al., 1995), Pesosjärvi and Vuoskojärvi in Finland (Juntto, 1992)). As a rule, the number of elements monitored there is rather limited compared to the number of elements of interest for the Kola project. Other studies carried out on the composition of precipitation from this area (mostly snow) include Derome et al. (1991, 1992), Berg et al. (1994), Makarova et al. (1994), Jaffe et al. (1995), Kelley et al. (1995), Reimann et al. (1996) and Soveri and Peltonen (1996).

The aim of this study was to improve our understanding of element sources, cycling, seasonal effects and variability on a relatively detailed scale, to aid the interpretation of the results of the planned low density (1 sample site per 100-600 km<sup>2</sup>) regional mapping project. The main characteristics of the eight catchments are summarised in Table 3.3.2.

## SAMPLING

For this project a special rainwater sampling device had to be designed and constructed (Fig. 3.3.2), allowing for rough conditions. To collect the rain, new polyethylene bags - all from a single production batch and checked for contamination - were used. Although working in Arctic areas where evaporation should not be a major problem, the PE bags were nearly closed using a plastic strip (Fig. 3.3.2) and the outer tubes of the holders were covered with aluminium foil during July and August.

Three stations, each consisting of five samplers placed about 10 m apart, were set up in all eight catchments in open areas to avoid throughfall. A composite sample from the five samplers per station was taken monthly, avoiding bags that were visibly contaminated or contained far too much or too little water compared to the others. The number of bags and amount of water were recorded. The samples were composited by pouring the rainwater from each sampler at each station into a new large PE-bag mounted in a PE-bucket. Water for cation analyses was sucked out of the bag using a PE syringe that was cleaned with rain water three times before sampling. Water for anion analysis was poured out of the plastic-lined bucket into a 500 ml PE-bottle. The total volume of precipitation was recorded after sampling, using a 2 litre PE-measurement cylinder. More details on sampling are given in Äyräs and Reimann (1995).

## ANALYSIS

Samples for cation analysis (Ag, Al, As, B, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Sb, Se, Sr, Th, Tl, U, V and Zn) were filtered through a 0.45µm membrane filter (Millipore<sup>TM</sup> Millex-HA) and acidified in the field prior to analysis using ICP-AES and ICP-MS. Unfiltered and unacidified samples were taken to analyse for Br, Cl, F, NO<sub>3</sub>, PO<sub>4</sub> and SO<sub>4</sub> by ion chromatography and to perform potentiometric determination of pH and electrical conductivity. All samples were analysed in the laboratory of the Geological Survey of Finland (GTK).

The GTK laboratory is accredited according to ISO 9001 and ISO-Guide 25. Quality control data suggest that some samples may be contaminated with Zn, the source apparently being teflon-coated rubber fittings whose coating wears off with time. Zn data presented here should thus be treated with care.

## RESULTS

Table 3.3.3 summarises the analytical results for each of the eight catchments separately (median, minimum and maximum values).

## DISCUSSION

Table 3.3.3 shows that the predominant sources of heavy metals in the whole area are the smelters in Monchegorsk (C2) and Nikel (including the ore roasting plant in Zapoljarnij (C1)).

The following suite of elements can be immediately identified as part of the emission spectrum of the Monchegorsk smelter: Ag, Al, As, B, Bi, Cd, Cl, Co, Cr, Cu, F(?), Fe, Mg, Mo, Ni, Pb, S, Sb, Se, Si, Tl, V and Zn.

Normalisation to a crustal element like Fe is often used to calculate enrichment factors in atmospheric chemistry (e.g. Galloway et al., 1982). We do not think this approach is justified as no such thing as a “crustal average” for any one element exists at a given place. The average composition of dust within an area will to a very large degree be determined by the local lithologies; element contents in different lithological domains can vary by several orders of magnitude and any calculation using a world-wide average will very likely be erroneous by at least one or two orders of magnitude.

One can, however, use a regional average for a given large area to calculate simple enrichment ratios. For our study, C7, situated in the centre of the study area and having the most widespread lithology (gneisses), should be fairly representative for the regional background. When the enrichment ratios for C2, Monchegorsk, are calculated by dividing the median observed for C2 by the median for C7, the following sequence is obtained (two values are given for those elements where the median in C7 is below the detection limit: the first value using half of the detection limit for the calculation, the second using the detection limit): Ni (633), Co (1180/590), Cu (453), As (154), Mo (107/53), Al (36), V (27), Cd (15), Sb (26/13), Pb (11), Zn (10), Fe (8), Sr (7), Na (6), S/SO<sub>4</sub> (6), Cl (6), Cr (5), Se (4) and Ag (6/3).

Ca, Cl and Na reach maximum levels in C1. This is the catchment closest to the Barents Sea and these high concentrations are at least partly due to the additional input of sea spray. The high levels of Fe, K and Mn are related to increased dust input due to the open cast mining operations near Zapoljarnij (see Reimann et al., 1996).

When our data are compared with some published data from this area (Berg, 1994, Hagen et al., 1995), it is obvious that ours are low (by a factor of 2 - 10), especially when the results from the Svanvik station are compared with those from C5 just 40 km to the south. For the majority of elements, the Svanvik data are even higher than our values from C1 near the roasting plant at Zapoljarnij. The main cause for the observed discrepancy presumably is the difference in practice when it comes to filtering or not filtering the rain water samples (we filtered at 0.45µm). Snow data (Reimann et al., 1996), where meltwater (<0.45µm) and filter residue data are compared, indicate that for Ni and Cu more than 60% of the total deposition in the vicinity of the nickel industry comes in particle size classes >0.45µm. Thus, filtering the samples explains the observed differences.

Comparison between our data and data published from northern Finland (Juntto, 1992) and other parts of the world (e.g. Galloway et al., 1982, Ross and Vermette, 1995) shows that ours fit very well within the ranges given.

Figure 3.3.3 shows boxplot comparisons of element levels and variations for Al, As, Cu, Ni, Rb, SO<sub>4</sub>, V and pH in the eight catchments studied. Note that for some elements contents were log-transformed prior to plotting.

The boxplot as used in Fig. 3.3.3 is an useful graphic originating from exploratory data analysis (EDA) (Tukey, 1977). It provides a graphical data summary, relying solely on the inherent data structure and not on any assumptions about the normality of the data distribution. It basically divides the distribution of the results into quartiles, firstly by finding the median (displayed as a

line in the box), and then doing the same for each of the remaining halves. These upper and lower points or «hinges» define the central box which thus in itself again contains 50% of all data. «Whiskers» are then drawn from the ends of the box, each extending 1.5 times the width of the box towards the maximum and the minimum (taken back to the last real data point). Any values outside of these whiskers are defined as data outliers. Important information about the dataset, e.g. median, quartiles, skewness of the distribution and the existence of data outliers can all be extracted at one glance from this simple graphic.

SO<sub>4</sub> levels in the different catchments are much more similar than those of the metals (C2 “only” by a factor of 10 above background). The SO<sub>2</sub> emissions are spread over a much larger area than the metals, thus explaining the much smaller differences between the catchments. Note, however, that the general patterns for Ni and SO<sub>4</sub> (and all the other “main pollutants”) are similar from C1 to C8, displaying the typical anthropogenic or technogenic signature for this area. A similar signature is apparent in other media, such as snow (Äyräs et al., 1995, Reimann et al., 1995), stream water (Caritat et al., in prep.) and topsoil (Reimann et al., in prep.). pH is lowest in C2 and highest in C3; all other catchments show very similar levels. A considerable input of basic rock dust buffers the pH levels in precipitation around the smelters (Reimann et al. 1996).

There is clearly an additional source for Al and V at C3. Rb shows a pattern that is typical for an element originating from natural sources (geogenic dust), with very little differences in levels between the catchments. It is interesting to note that elements mostly influenced by “geogenic” or, better, “natural” sources (e.g. Ba, K, Li, Mn, Rb) generally show a much smaller variation, while very large contrasts in concentration levels, a great variation within one catchment and generally severely skewed distributions seem to be typical of anthropogenic sources.

#### *Seasonal variation*

Variations in time and between the three stations within selected catchments are plotted in Figure 3.3.4 for the same elements as in Figure 3.3.3, but only for three catchments, C2, C4 (representing contamination at different distances from the smelter) and C7 (representing background). Note that a mixture of linear and logarithmic scales is used in these diagrams, depending on the observed element variability. For most elements, differences between catchments are much larger than seasonal effects and these again are larger than variations between the three stations per catchment. An exception is Rb, which has a “natural” source. For Rb the largest difference is observed between the three stations in C7, in August. pH also shows a great variation for the C7 August samples. Note, however, that the scale for Rb is non-logarithmic and total variation is thus very low compared to the other elements. Differences in the emission-related elements over time are especially great in the Russian catchments; for Al they can be up to one order of magnitude (C2: June vs. August). Trends differ in the various catchments and can help to improve our understanding of the sources of different elements.

Figure 3.3.5 shows the seasonal variation of the Cu/Ni ratio at C2 and C1. This ratio is of particular interest in this area as it is very different in the two ore types used in the Monchegorsk smelter, near C2. The ore roasting plant at Zapoljarnij (near C1) processes only Pechenga ore with a Cu/Ni ratio of about 0.5 whereas the Norilsk ore which, in addition to the Pechenga ore, is smelted at Monchegorsk has a Cu/Ni ratio of about 2 (Pechenga Nickel Combinat, pers. comm.). When this ratio is plotted, as it is revealed by the rainwater samples month after month, it remains very constant in C1 at around 1 (median: 0.8), but C2 shows a

sharp drop from 6.5 to 2.5 in August, and throughout the autumn, indicating a change from the use of Norilsk ore to Pechenga ore.

### *Correlations*

XY diagrams (Fig. 3.3.6) show a generally very good correlation for samples from the Russian catchments, with the highest values in C2 lying on one trend with C4 and C3 (distance from source). C1 often shows an offset from the Monchegorsk trend due to the difference in ore feed (compare Cu/Ni, As/Ni, Pb/Ni, V/Ni) as well as the difference in the technological process (roasting vs. smelting). The differences in the ore feed at Monchegorsk are clearly displayed in two slightly offset trends for the C2 samples in these diagrams, one set coming close to the C1 trends.

Samples from the Norwegian catchment take an intermediate position, but are often closer to the C2 than to the C1 trend. This could be caused by mixed inputs from both smelting in Nikel and roasting in Zapoljarnij, or fractionation of the different elements with transport distance in the atmosphere, which would, for example, mean that Ni is deposited faster than Cu (differences in particle size distributions in the aerosol?). Such a fractionation can be clearly seen in the Ni/Pb diagram, where Pb is strongly enriched in relation to Ni in C5. The Finnish background samples, however, do not usually display any correlation or trend other than an enrichment/depletion in one or other element.

The Ni/V diagram shows that there must be a local source of V in, or close to, C3 because there is about 10 times more V than expected from the Ni content and the trend of the other catchments. This V source could be the coal-fired power plant in Apatity, exhaust from diesel trucks in the opencast mine (road within the catchment), or simply lithology (local dust).

The Al contents in C3 show a strong offset in the Al/Ni diagram, clearly demonstrating that local dust from opencast mining is an important source of elements observed in rain in C3. Otherwise the correlation of Al with Ni for C1, C2, C4 and C5 is surprisingly good, whereas that between Al and Rb is poor, arguing against a purely dust-related origin for the high Al levels. Al thus seems to have a fourfold origin: natural dust, anthropogenic "natural" dust originating from the mining of alkaline rocks in C3, smokestack emissions from the smelters, and wind blown "anthropogenic" soil dust due to the severely damaged vegetation cover around the smelters in C2, C4, C1 and C5.

## CONCLUSIONS

The values obtained from the rainwater samples are generally very low for the 38 analysed elements/parameters and, even with advanced analytical techniques (ICP-MS), results for many elements were at or below the detection limit. The highest levels for almost all elements can be observed in C2, followed by C1, C4 and C3, this sequence reflecting the distance from industrial plants. Most of the heavy metals (Co, As, Cu, Mo, Ni, Sb) show enrichments of two to three orders of magnitude in their median levels close to industry in Russia, compared to background levels in Finland. C5 in Norway has an intermediate position with regard to pollution. There are a few short-lived events, dependent on the wind direction, when very high inputs of contaminants were registered here.

Regional variation in element content was found to be much greater than temporal variations or variations between the three stations per catchment. When time variations in rain chemistry

in C2 are examined in more detail, drastic changes in element ratios can be detected, reflecting changes in the ore fed into the smelter (Pechenga vs. Norilsk ore).

The following elements reach maximum concentrations in rainwater at C2: Ag, Al, As, B, Ba, Bi, Cd, Co, Cr (highest median, highest single value in C8), Cu, Li (highest median, highest single value in C1), Mo, Ni, Pb, S, Sb, Se, Si, Tl, V and Zn (highest median, highest single value in C6). Electrical conductivity also shows maximum values in C2, pH is lowest. Furthermore, clearly elevated levels can be observed for Ca, Cl, Fe, Mg, Na and Sr. All these elements can be attributed to industrial activity and can thus be classified as “anthropogenic”. Some (Al, Ba, Ca, Fe(?), Mg, Li, Sr) are probably further enriched or solely caused by increased dust input due to industry-related activities and/or destroyed vegetation cover in the surroundings of the smelters.

Monitoring rainwater chemistry in 8 catchments at different distances from major, well-defined pollution sources in an otherwise nearly pristine area proved to be very successful in determining the degree of contamination, the large number of pollutant elements, the very sharp drop of almost all element levels with distance from the smelters, and in improving our understanding of the various sources of element input to the catchments studied.

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## TABLES

Table 3.3.1: Official Russian emission data for 1993 and 1994 for the main sources of heavy metals and sulphur in the survey area (Committee for ecology and natural resources CENR, 1995).

Table 3.3.2: Summary of some characteristics of the different catchments.

Table 3.3.3: Summary statistics of rainwater analysis for the eight catchments investigated. The highest median value and the highest maximum value for any one element are printed in bold type. Samples are 30-day composite samples for the summer (May to September) of 1994. All samples for cation analysis were filtered at  $<0.45\mu\text{m}$  and acidified with ultrapure  $\text{HNO}_3$  in the field; samples for anion analysis and measurement of pH and EC were left untreated.

## FIGURES

Fig. 3.3.1: Location of the study area for the regional mapping project and the catchments from which data are presented here. Svanvik (NILU's official rainwater monitoring station) is located halfway between Kirkenes and C5, close to the Russian border.

Fig. 3.3.2: Construction drawing of the rainwater bulk sampler used for this project.

Fig. 3.3.3: Boxplot comparison of element levels and variations in the eight catchments (C1 to C8) studied.

Fig. 3.3.4: Seasonal variations in rainwater samples as observed in C2, C4 and C7.

Fig. 3.3.5: Seasonal variations in the Cu/Ni ratio in rainwater from C2 (Monchegorsk, where Pechenga ore and ore from Norilsk are used, which have different Cu/Ni ratios) and C1 (Zapoljarnij, where only Pechenga ore is roasted).

Fig. 3.3.6: XY diagrams for selected elements. Lines indicate element ratios as given in the figures.

Table 3.3.1 Official emission data 1993/94 (CENR, Murmansk)

Location	Year	t/y									
		Ni	Cu	Co	V <sub>2</sub> O <sub>5</sub>	HF	Cl	H <sub>2</sub> S	SO <sub>2</sub>	CO <sub>2</sub>	NO <sub>2</sub>
Murmansk	1993	-	-	-	98.4	-	-	-	30991	4420	1651
	1994	0.0	0.4	-	91.3	0.172	0.03	-	26587	3540	1266
Nikel	1993	129.8	87.1	5.2	13.4	-	-	-	160629	267	167
	1994	136.1	81.9	5.2	12.9	-	-	-	129160	244	158
Zapoljarnyi	1993	152.3	74.8	5.4	23.1	-	-	-	66629	499	286
	1994	161.1	81.0	5.4	21.4	-	-	-	69208	463	285
Monchegorsk	1993	1960.3	1049.1	89.2	57.2	-	365.4	27.7	136880	1294	5117
	1994	1618.8	933.7	81.5	59.8	-	341.3	18.1	97715	917	1267
Olenegorsk	1993	-	-	-	0.3	-	-	-	3519	419	468
	1994	-	0.0	-	0.4	-	-	-	3508	869	610
Apatity	1993	-	-	-	26.1	-	-	-	21406	683	6927
	1994	-	-	-	0.4	-	-	-	14576	271	5141
Kirovsk	1993	-	-	-	51.7	-	-	-	3753	811	248
	1994	-	-	-	43.6	-	-	-	4041	554	1358

Table 3.3.2. Overview of the main characteristics of the catchments

No. Name	Coordinates of catchment outlet	Size (km <sup>2</sup> )	Elevation (m a.s.l.)	Annual precip. (mm)*	Vegetation	Bedrock	Surface cover, peculiarities
<b>RUSSIA</b>							
C1 Zapoljarniy	69°27'01"N 31°03'49"E	19.02	25-373	454	birch forest tundra	gneiss	till, fluvioglacial, outcrop
C2 Monchegorsk	67°50'30"N 32°54'48"E	22.38	128-507	391	technogenic desert, birch shrubs	dacite & andesite & tuffs, gabbro/norite	till, prone to erosion
C3 Kirovsk	67°32'50"N 33°48'55"E	20.01	240-1075	502	spruce forest, mountain tundra birch forest	nephelinite	till, diluvial/eluvial
C4 Kurka	67°41'25"N 32°50'14"E	20.49	152-466	502	north taiga spruce forest, birch; incipient deterioration	amphibolite, gneiss	till, fluvio-glacial
<b>NORWAY</b>							
C5 Skjellbekken	69°21'25"N 29°27'25"E	34.56	80-297	422	north taiga pine forest, birch	andesite, basalt & tuffs, 'black shale'	till, esker
<b>FINLAND</b>							
C6 Kirakka	69°35'12"N 28°51'46"E	11.86	110-200	386	north taiga pine forest	granite	outcrop, till, moraine ridge
C7 Naruska	67°21'44"N 29°22'05"E	20.16	263-490	513	north taiga spruce forest	gneiss	till, peat, outcrop
C8 Pallas	68°09'14"N 23°52'50"E	24.42	303-500	405	north taiga spruce forest	quartzite	till, peat

\* from the closest meteorological station (data from 1994)

Table 3.3.3 summary statistics RAIN									
Elem.	Unit	C1	C2	C3	C4	C5	C6	C7	C8
		median	median	median	median	median	median	median	median
		(range)	(range)	(range)	(range)	(range)	(range)	(range)	(range)
Ag	µg/L	<0.01	<b>0.03</b>	<0.01	0.01	all <0.01	all <0.01	all <0.01	all <0.01
		<0.01-0.05	<0.01- <b>0.14</b>	<0.01-0.05	<0.01-0.06				
Al	µg/L	12.4	<b>105</b>	51	10.3	5.2	2.5	2.9	2.9
		6.0-45	13.4- <b>272</b>	30-171	4.7-68	2.8-11.1	1.6-25.7	0.9-8.6	1.7-7.4
As	µg/L	0.58	<b>12.3</b>	0.22	2.7	0.26	0.11	0.08	0.07
		0.18-1.34	3.6- <b>84.4</b>	0.11-2.44	1-9.3	0.15-2.6	0.08-0.16	<0.05-0.18	<0.05-0.1
B	µg/L	<0.5	<b>0.73</b>	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
		<0.5-2.11	<0.5-2.36	<0.5-0.79	<0.5-0.78	<0.5-0.75	<0.5-3.5	<0.5-1.17	<0.5- <b>16.9</b>
Ba	µg/L	0.83	<b>1.07</b>	0.99	0.51	0.58	0.58	0.47	0.71
		0.6-2.3	0.6-1.9	<b>0.5-2.9</b>	0.3-1.2	0.3-0.7	0.4-0.7	0.4-1.1	0.3-1.5
Be	µg/L	all <0.1	all <0.1	all <0.1	all <0.1	all <0.1	<0.1	all <0.1	<0.1
							<0.1-0.11		<0.1- <b>0.12</b>
Bi	µg/L	all <0.03	<0.03	all <0.03	<0.03	all <0.03	all <0.03	all <0.03	all <0.03
			<0.03- <b>0.08</b>		<0.03-0.03				
Br	µg/L	all <0.2	all <0.2	all <0.2	all <0.2	all <0.2	all <0.2	all <0.2	all <0.2
Ca	mg/L	<b>0.15</b>	0.1	0.12	0.04	0.1	0.07	0.05	0.1
		0.08- <b>0.4</b>	0.05-0.2	0.09-0.32	0.02-0.13	0.02-0.16	0.05-0.1	0.02-0.14	0.02-0.26
Cd	µg/L	0.07	<b>0.89</b>	0.03	0.13	0.03	<0.02	0.06	<0.02
		0.03-0.16	0.32- <b>5.11</b>	<0.02-0.16	0.06-0.96	<0.02-0.16	<0.02-0.05	<0.02-0.33	<0.02-0.64
Cl	mg/L	<b>0.9</b>	0.6	0.2	0.25	0.4	0.2	0.1	0.15
		0.3- <b>3.7</b>	0.3-2.1	0.05-0.7	0.1-1.1	0.3-1.8	0.2-0.6	<0.1-0.3	0.1-1.1
Co	µg/L	0.36	<b>11.8</b>	0.03	1.07	0.04	0.03	<0.02	<0.02
		0.14-1.52	2.17- <b>68.9</b>	<0.02-0.07	0.24-3.32	<0.02-0.19	<0.02-0.04	<0.02-0.03	<0.02-0.04
Cr	µg/L	<0.2	<b>0.47</b>	all <0.2	<0.2	all <0.2	<0.2	all <0.2	<0.2
		<0.2-0.8	0.25-0.95		<0.2-0.3		<0.2-0.26		<0.2- <b>2.04</b>
Cu	µg/L	8.3	<b>231</b>	1.2	27	1.8	0.7	0.51	0.4
		4.4-38	86- <b>848</b>	0.6-4.4	7-89.5	0.76-9	0.25-1.44	0.24-1.75	0.2-2.1
F	mg/L	all <0.05	<0.05	all <0.05	all <0.05	all <0.05	<0.05	all <0.05	<0.05
			<0.05-0.08				<0.05-0.18		<0.05- <b>1.53</b>
Fe	mg/L	<b>0.04</b>	<b>0.04</b>	<0.01	0.01	<0.01	<0.01	<0.01	<0.01
		0.01- <b>0.13</b>	<0.01-0.09	<0.01-0.01	<0.01-0.03	<0.01-0.01	<0.01-0.04	<0.01-0.01	<0.01-0.02
K	mg/L	<b>0.17</b>	0.09	0.11	0.06	0.08	0.12	0.05	0.14
		0.04-0.82	0.02-0.36	0.02-0.72	0.03-0.2	0.03-0.33	0.06-0.33	0.02-0.26	0.03- <b>1.53</b>
Li	µg/L	<0.1	<b>0.12</b>	<0.1	<0.1	all <0.1	<0.1	<0.1	<0.1
		<0.1- <b>0.5</b>	<0.1-0.28	<0.1-0.24	<0.1-0.22		<0.1-0.25	<0.1-0.45	<0.1-0.41
Mg	mg/L	<b>0.1</b>	0.04	0.01	<0.01	0.01	0.03	0.02	<0.01
		<0.01-0.27	<0.01-0.1	<0.01-0.13	<0.01-0.06	<0.01-0.2	<0.01-0.06	<0.01-0.04	<0.01- <b>0.34</b>
Mn	µg/L	<b>3.6</b>	2	1.6	1.2	1.4	1.5	2.1	<b>3.6</b>
		1.5-18.1	1.0-4.4	0.6-4.7	0.5-2.2	0.7-4.5	1.2-2.9	0.45-6.11	0.6- <b>166</b>
Mo	µg/L	all <0.03	<b>1.6</b>	0.04	0.33	<0.03	all <0.03	all <0.03	all <0.03
			0.46- <b>7.36</b>	<0.03-0.14	0.12-0.97	<0.03-0.05			
Na	mg/L	<b>0.4</b>	0.3	0.2	<0.1	0.2	0.2	<0.1	<0.1
		0.2- <b>1.9</b>	<0.1-0.72	<0.1-0.61	<0.1-0.56	<0.1-1.1	<0.1-0.58	<0.1-0.13	<0.1-0.19
Ni	µg/L	10.3	<b>57</b>	0.72	9	1.31	0.36	0.09	0.21
		3.3-42.2	24- <b>132</b>	0.35-1.26	1.7-20.3	0.8-4.6	0.16-0.77	<0.06-0.21	0.08-0.49
NO3	mg/L	0.45	0.5	0.5	0.4	0.5	<b>0.6</b>	0.5	0.4
		0.3-0.8	0.3-0.8	0.3-0.7	0.2-0.8	0.2-0.8	0.2-0.7	0.3-1	0.1-0.7
P	mg/L	all <0.05	all <0.05	all <0.05	all <0.05	all <0.05	all <0.05	all <0.05	all <0.05
Pb	µg/L	0.94	<b>6.3</b>	0.61	2	2.42	0.75	0.56	0.84
		0.61-1.63	2.1- <b>40.5</b>	0.38-1.85	0.51-9.1	0.53-6.8	0.27-1.7	0.1-1.4	0.26-6.93

PO4	mg/L	0.04	0.03	<0.02	<0.02	<0.02	<b>0.06</b>	<0.02	<0.02
		<0.02-0.94	<0.02-0.55	<0.02- <b>2.15</b>	<0.02-0.39	<0.02-0.63	<0.02-0.88	<0.02-0.55	<0.02-0.71
Rb	μg/L	0.19	0.19	0.42	0.12	0.09	0.27	0.17	<b>0.47</b>
S	mg/L	0.07-0.69	0.06-0.52	0.07-1.06	0.05-0.39	0.05-0.47	0.11-0.41	0.05-0.68	0.08- <b>2.31</b>
		0.75	<b>1.63</b>	0.51	0.61	0.35	0.44	0.29	0.31
Sb	μg/L	0.52-1.45	0.87- <b>3.92</b>	0.39-0.96	0.25-1.55	0.27-1.11	0.32-0.5	0.09-0.56	0.16-0.49
		<0.025	<b>0.32</b>	0.03	0.09	<0.025	<0.025	<0.025	<0.025
Se	μg/L	<0.025-0.12	0.12- <b>1.78</b>	0.025-0.07	0.03-0.23	0.025-0.05	0.025-0.04	0.025-0.36	0.025-0.03
		all <0.5	<b>0.93</b>	all <0.5	<0.5	all <0.5	all <0.5	all <0.5	all <0.5
			<0.5- <b>6.6</b>		<0.5-0.7				
Si	mg/L	all <0.1	<0.1	< 0.1	all <0.1	all <0.1	all <0.1	all <0.1	<0.1
SO4	mg/L	2.25	<0.1-0.2	<0.1- <b>0.23</b>	1.5	1.8	1	1.3	<0.1-0.13
		1.6-4.5	2.7- <b>18.2</b>	1.1-2.9	0.7-4.7	0.8-3.2	0.7-1.4	0.2-1.7	0.8
Sr	μg/L	0.83	1.37	<b>2.31</b>	0.7	0.35	0.24	0.2	0.27
		0.47-2.24	0.43-4.44	1.26- <b>9.8</b>	0.35-2.3	0.21-1.3	0.17-0.44	0.1-0.44	0.13-0.62
Th	μg/L	all <0.02	all <0.02	all <0.02	all <0.02	all <0.02	all <0.02	all <0.02	all <0.02
Tl	μg/L	all <0.02	<b>0.02</b>	all <0.02	<0.02	<0.02	all <0.02	all <0.02	all <0.02
U	μg/L	all <0.01	<0.02- <b>0.14</b>	all <0.01	<0.02-0.02	<0.02-0.02	all <0.01	<0.01	all <0.01
			<0.01		all <0.01	all <0.01		<0.01- <b>0.04</b>	
V	μg/L	0.47	<b>3.51</b>	1	0.78	0.2	0.14	0.13	0.13
		0.22-0.65	1.36- <b>15.1</b>	0.35-1.88	0.32-2.78	0.14-1.12	0.11-0.25	0.07-0.23	0.08-0.36
Zn	μg/L	32	<b>56</b>	7.9	35	3.5	19.4	5.7	11.1
		31-634	33-197	3.7-99.8	17-53	1.8-9.5	3.9- <b>1810</b>	2.54-14.1	4.5-68.4
EC	mS/m	1.85	<b>4.6</b>	1.1	1.7	1.1	1	0.8	0.7
		1.1-3.1	2.0- <b>9.0</b>	0.7-3.4	0.9-4.4	0.8-2.8	0.7-1.9	0.4-1.7	0.4-2.8
pH		4.7	4	<b>5</b>	4.5	4.6	4.7	4.7	4.8
		4.1-6.3	3.7-4.3	4.3- <b>6.5</b>	4.1-5.0	4.4-5.2	4.4-5.4	4.3-6	4.5-5.3

*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
 Catchment locations

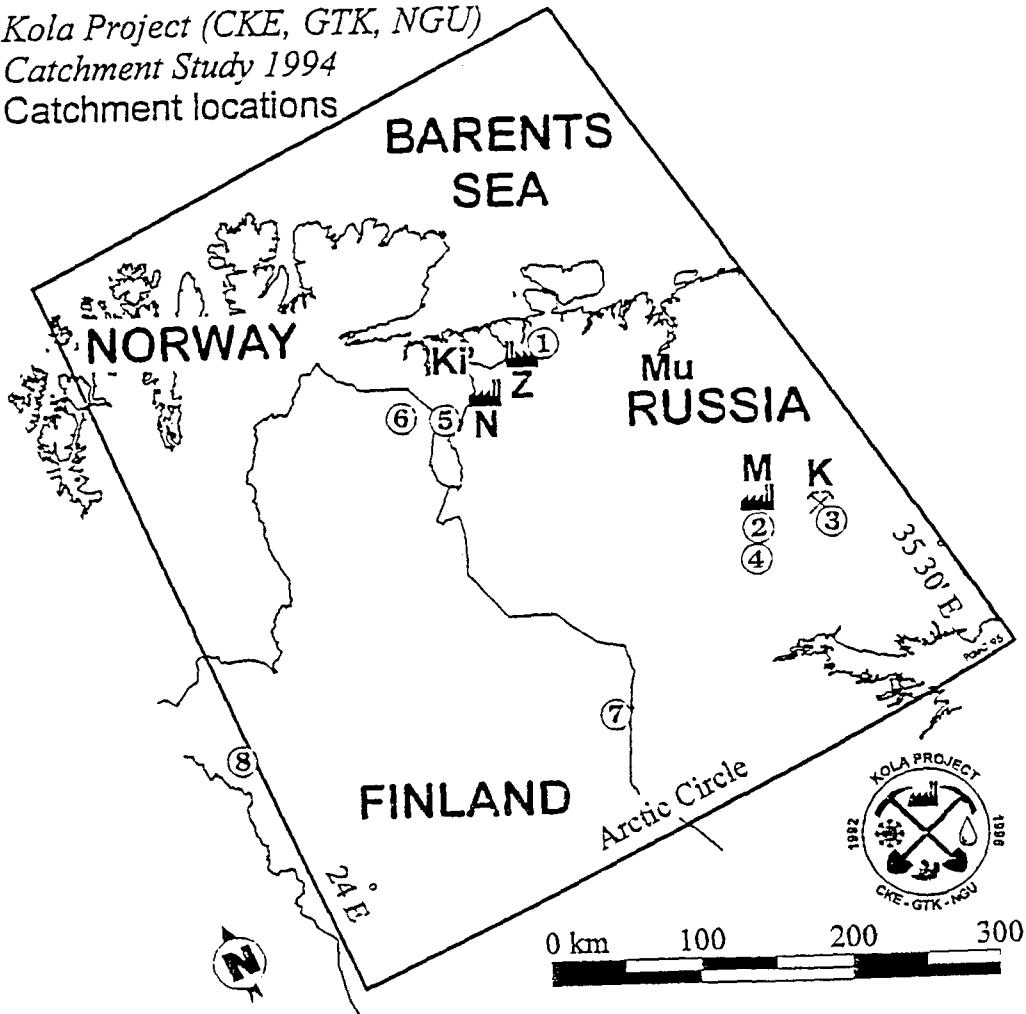


Fig. 3.3.1: Location of the study area for the regional mapping project and the catchments from which data are presented here. Svanvik (NILU's official rainwater monitoring station) is located halfway between Kirkenes and C5, close to the Russian border.

RAINWATER SAMPLER KOLA PROJECT

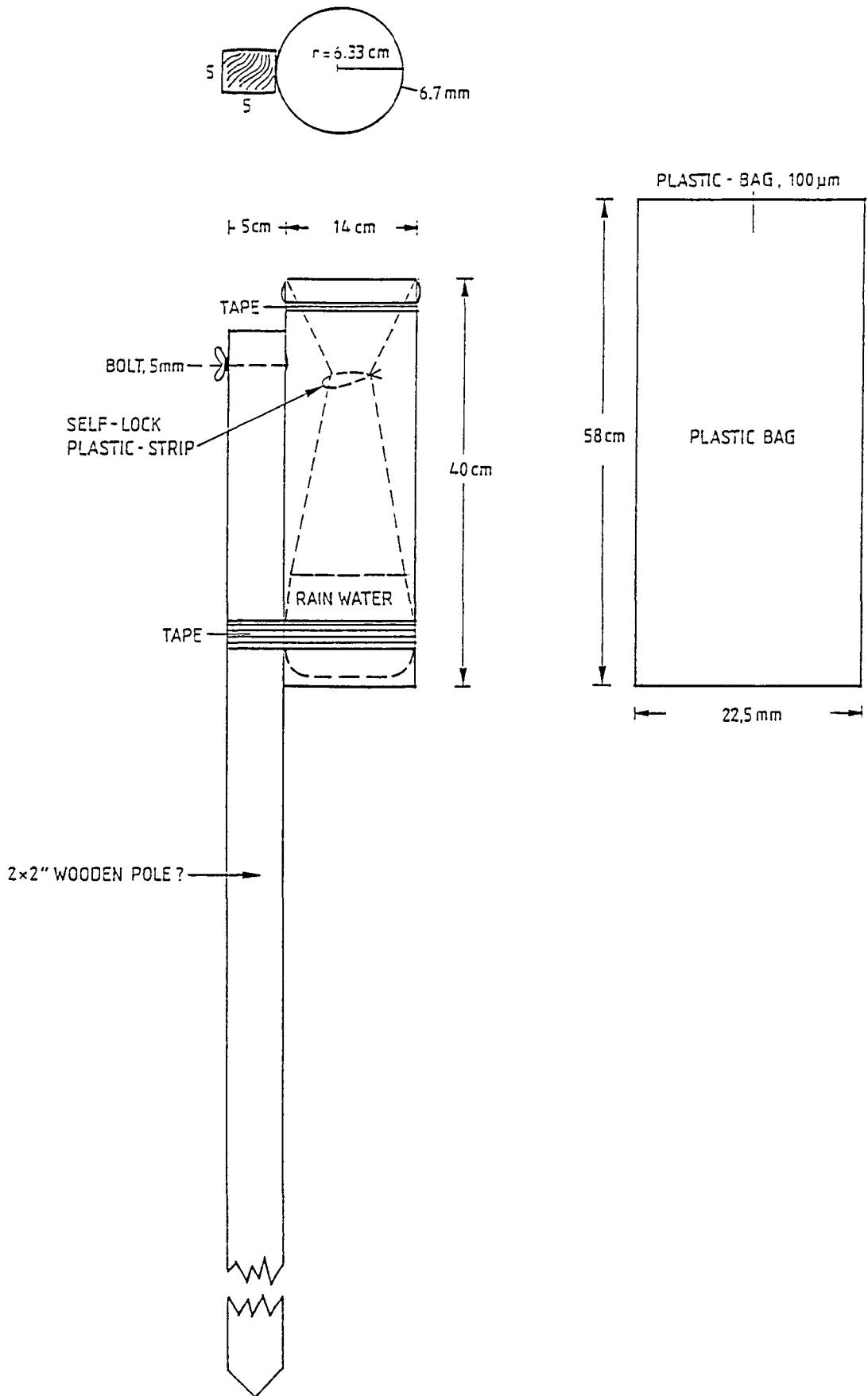


Fig. 3.3.2: Construction drawing of the rainwater bulk sampler used for this project.



Fig. 3.3.3: Boxplot comparison of element levels and variations in the eight catchments (C1 to C8) studied.

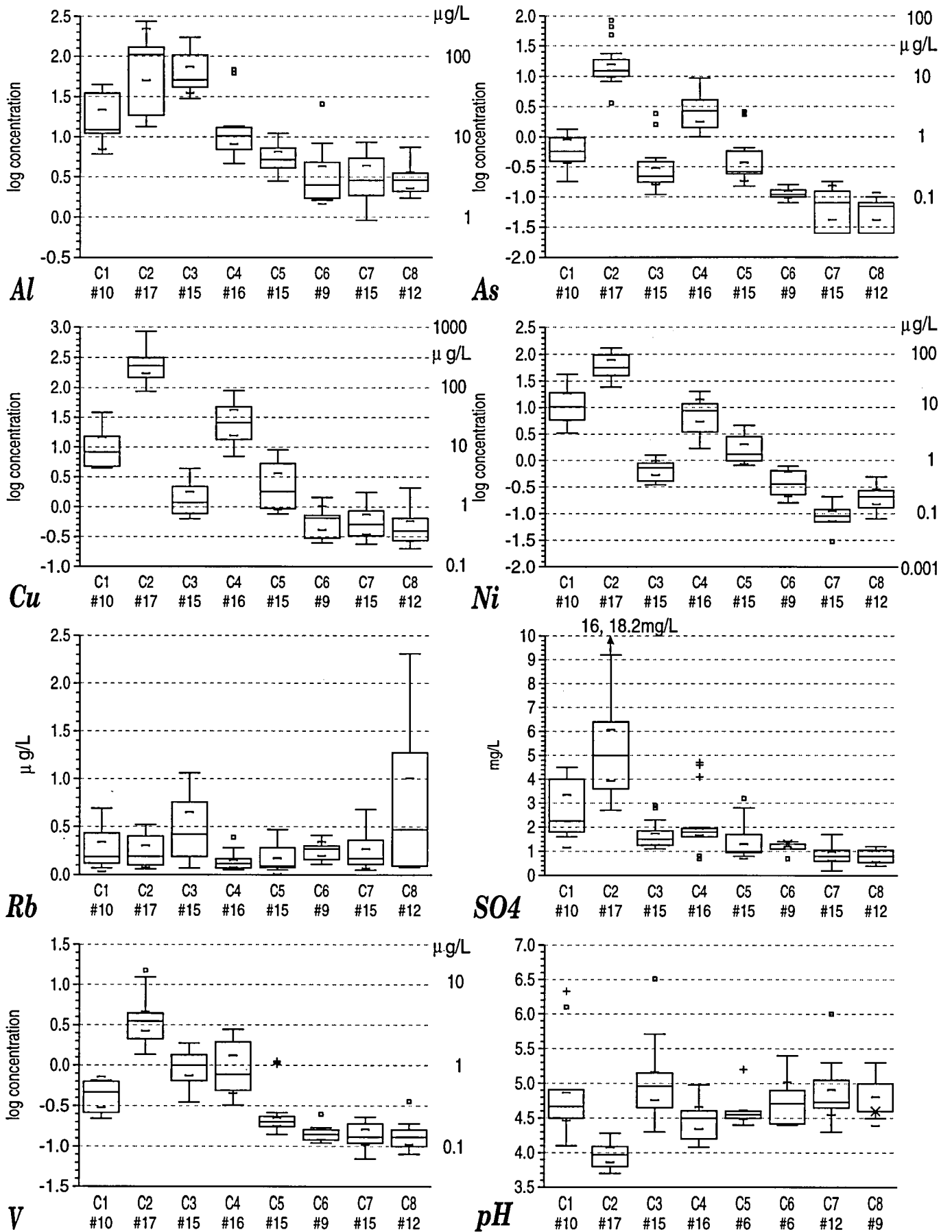
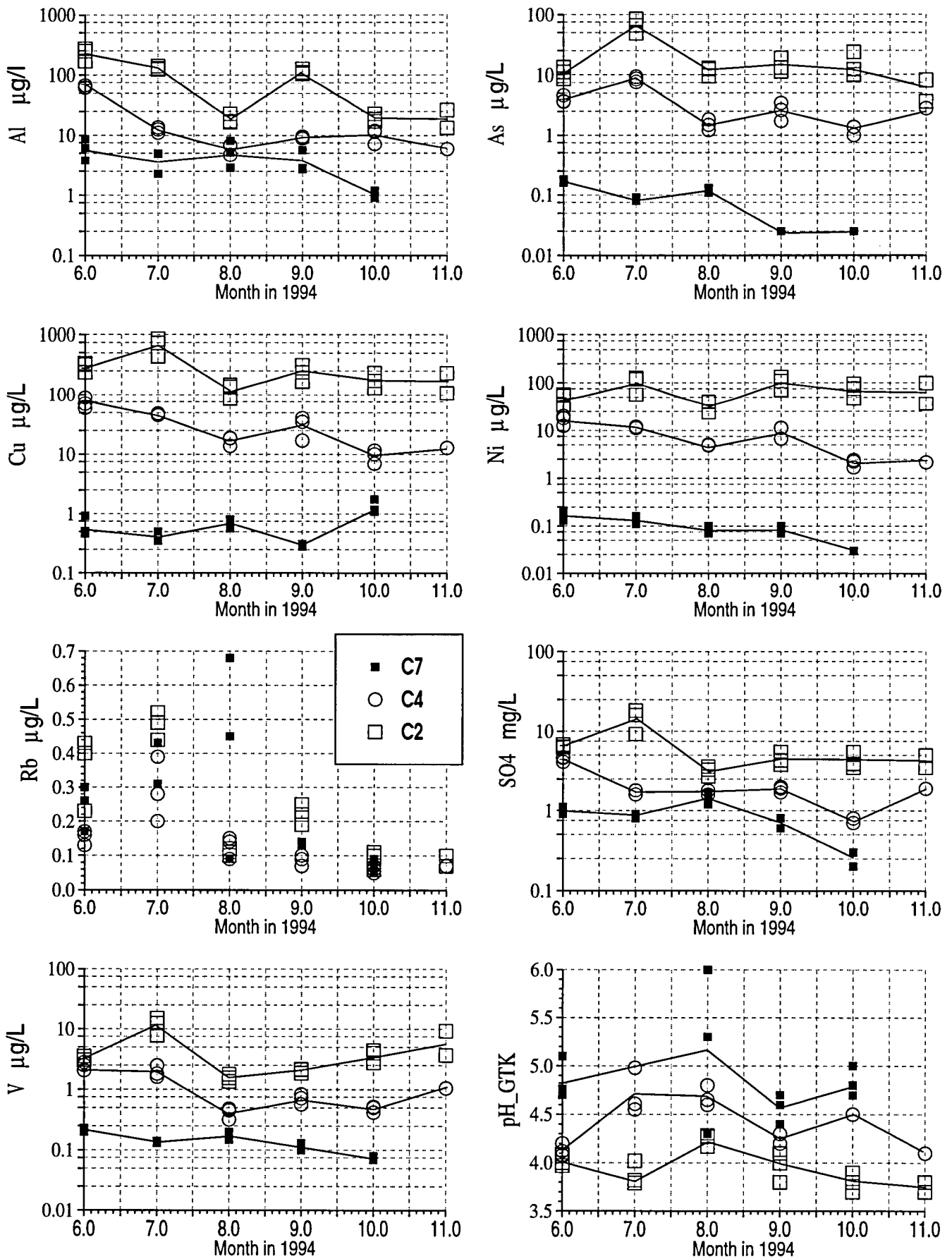


Fig. 3.3.4: Seasonal variations in rainwater samples as observed in C2, C4 and C7.



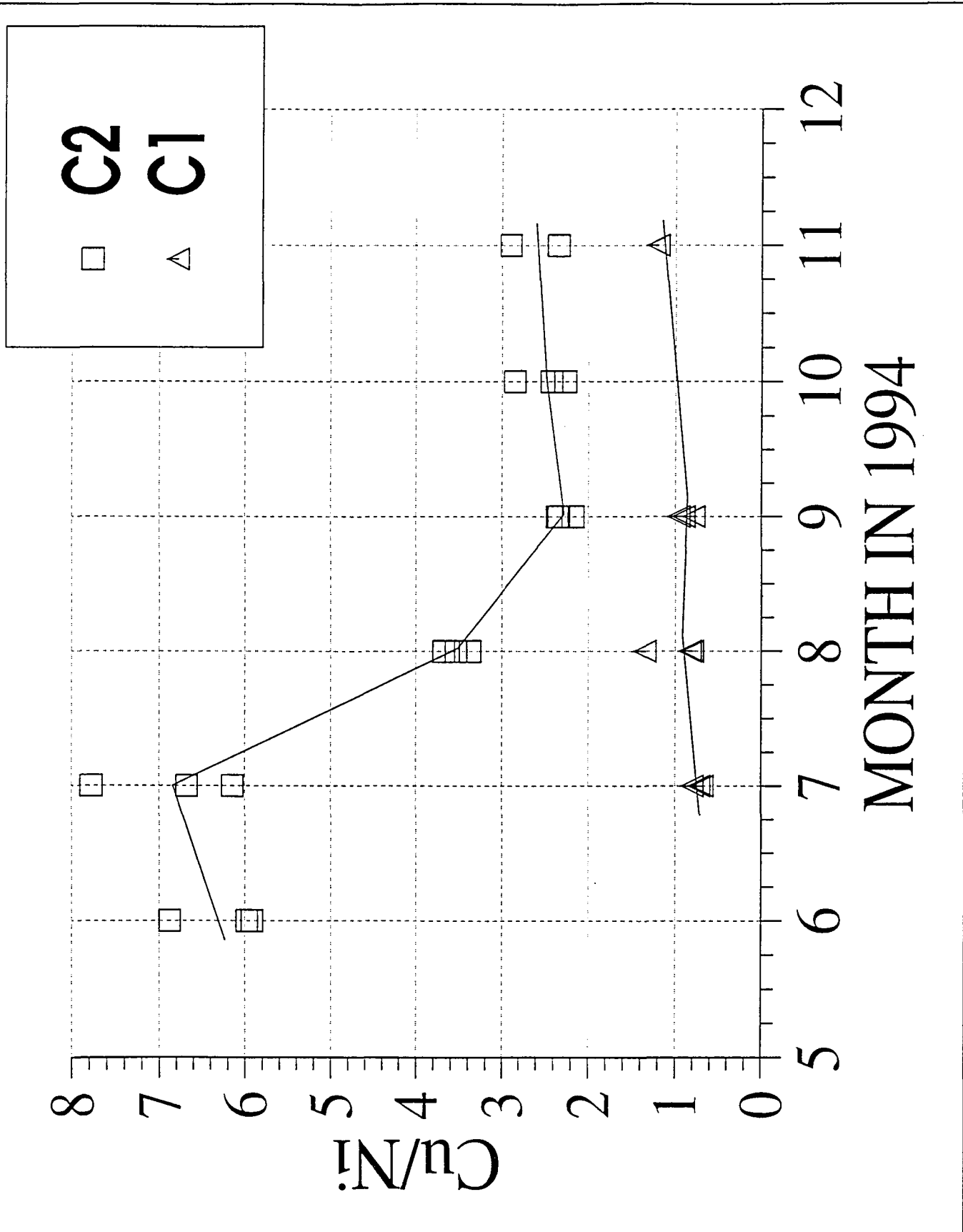
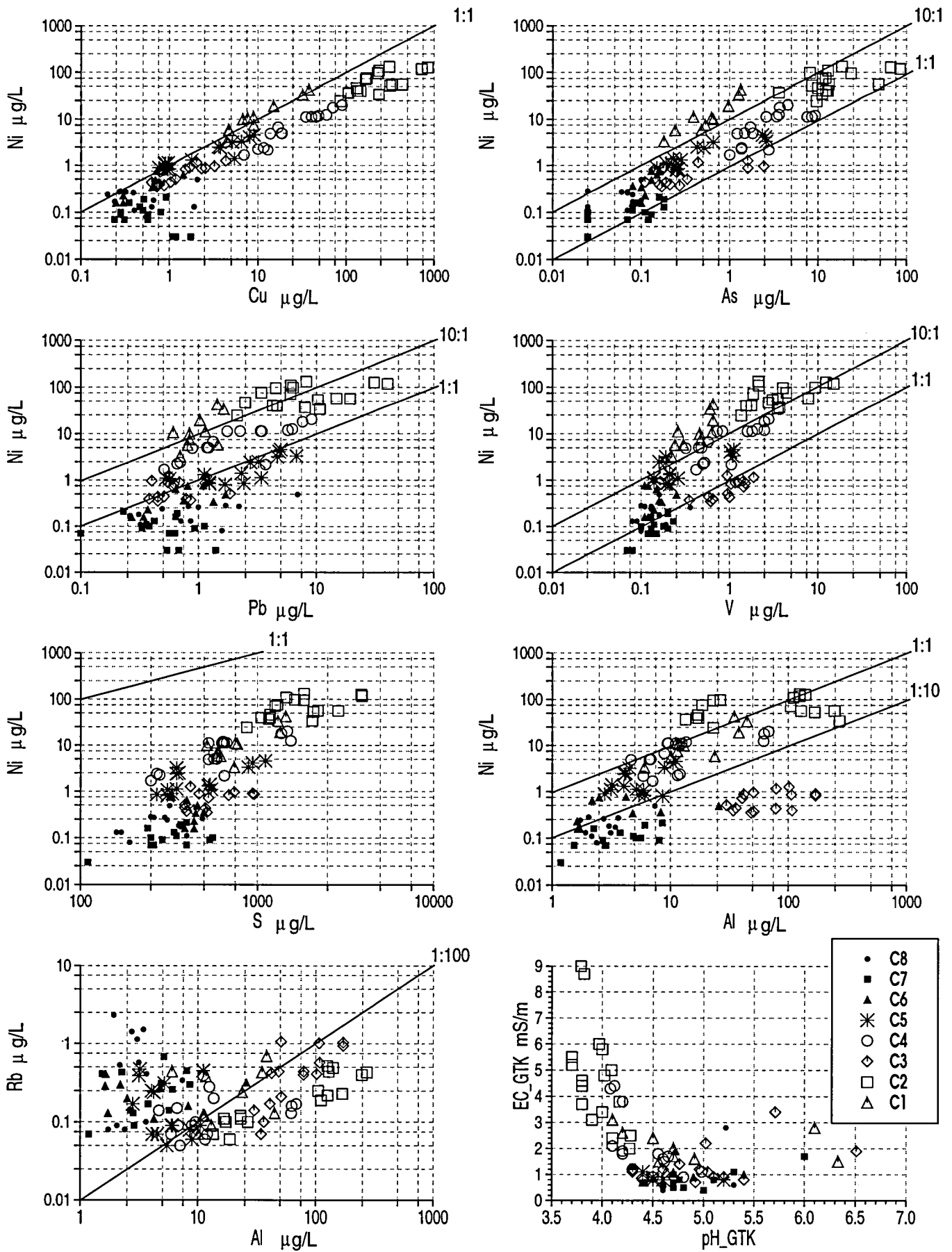


Fig. 3.3.5: Seasonal variations in the Cu/Ni ratio in rainwater from C2 (Monchegorsk, where Pechenga ore and ore from Norilsk are used, which have different Cu/Ni ratios) and C1 (Zapoljarnij, where only Pechenga ore is roasted).

Fig. 3.3.6: XY diagrams for selected elements. Lines indicate element ratios as given in the figures.



### 3.4 Annual atmospheric deposition of 17 elements in eight catchments of the Barents region

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#### ABSTRACT

Snow cover and rainwater samples were used to calculate total yearly (1994) deposition rates for Al, As, Ca, Cd, Co, Cr, Cu, Fe, Mg, Mo, Ni, Pb, S, Sb, Sr, V and Zn in eight catchments located in the Barents region, Finland, Norway and Russia. Wet and dry (>0.45 mm) deposition was determined separately for the snowpack samples representative of the winter 1993/94. Filtered rainwater sampled during the summer months of 1994 was used to obtain monthly wet deposition data.

The snow samples were used to calculate characteristic parameters for the dispersion of pollutants. These parameters are constant for any given industrial source. To calculate these, the element ratio of water soluble to particulate deposition was expressed as a function of distance from source. This function can then be used to calculate total (dry + wet) summer deposition data from the wet deposition data collected. Even superposition of pollution from two different sources can be separated using this technique.

#### INTRODUCTION

The Central Kola Expedition (CKE) and the Geological Surveys of Finland (GTK) and Norway (NGU) are carrying out a joint ecogeochemical mapping and monitoring project at the scale of 1:1 million in the Western Murmansk region and contiguous areas of Finland and Norway, covering 188,000 km<sup>2</sup> (see <http://www.ngu.no/Kola>). As one part of this cooperation, multimedia sampling in eight catchments (hereafter abbreviated as C1 to C8) distributed widely throughout the area (Figure 3.4.1) was carried out in 1994.

Some of the world's largest point sources of SO<sub>2</sub> emissions are located within the study area (Gunn et al., 1995): the nickel smelter at Nikel, the ore roasting plant at Zapoljarnij and the nickel smelter at Monchegorsk (Figure 3.4.1) account together for about 300,000 t of SO<sub>2</sub>, 1900 t of Ni, 1100 t of Cu and 94 t of V<sub>2</sub>O<sub>5</sub> emissions yearly (Murmansk Regional Committee for Ecology and Natural Resources - see Reimann et al., in press).

In terms of closeness to pollution sources, C2, Monchegorsk, Russia, is situated only 5 km S of the smelter in Monchegorsk, large parts of the vegetation are completely destroyed in this catchment. C1, Zapoljarnij, Russia, is located 10 km NE of the ore roasting plant at Zapoljarnij, and vegetation is severely damaged here. C4, Kurka, Russia, lies 25 km SW of the Monchegorsk plants at a distance where vegetation damage due to anthropogenic pollution is obvious even to the untrained eye (increase in the number of dead trees, underbrush damaged, moss missing). C3, Kirovsk, Russia, lies 50 km to the SE of Monchegorsk and borders on a large open cast mine for apatite ore. No vegetation damage is seen here. C5, Skjellbekken, Norway, lies 35 km to the SW (off-wind) of the smelter in Nikel, some vegetation damage is detectable for the trained eye only. C6 (Kirakka), C7 (Naruska) and C8 (Pallas) in Finland can be taken as typical background catchments for the whole survey area.

The main wind directions in the area are S-N, and the main topographic features follow this general direction as well.

Results obtained from snow cover for both meltwater (MW) and filter residue (FR), as well as from rainwater (RW) sampling are used here to evaluate levels of anthropogenic pollution at different distances to the main sources of contamination. The measurement of dry deposition data during summer time is not easy. Here we will demonstrate that the knowledge of the distribution of wet and dry deposition as measured in snowpack samples, can be used to estimate total yearly deposition data even when collecting wet deposition data only during summertime.

## SAMPLING

### *Snow*

Snowpack samples were collected in March/April 1994 at 10-12 stations per catchment. Sample sites were preferably located in level (20 x 20 m) areas with uniform snow depth. Composite snowpack samples were taken from this area, the number of subsamples depending on the thickness of snow cover. The minimum number of subsamples was three and the maximum number such that the weight of the composite sample was 4-6 kg. The entire snowpack thickness was sampled, with the exception of the lowermost 5 cm, to avoid mixing with vegetation and soil. A clear Plexiglas tube (Soveri, 1985) with Plexiglas closing mechanism was used for sampling. The inside diameter of the tube was 92.5 mm and the length 100 cm. The snow depth at the sampling sites was registered. Further field information included date of sampling, number of subsamples, weight of composite samples and average depth of snow cover on the sample plot (average of 20 measurements).

### *Rain*

For this project a special rainwater sampling device had to be designed and constructed (a construction drawing is given in Reimann et al., in press, Figure 3.4.2), allowing for rough conditions. To collect the rain, new polyethylene bags - all from a single production batch and checked for contamination - were used. The inner diameter of the rainwater collector is 12.66 cm (Reimann et al., in press a).

Three stations, each consisting of five samplers placed about 10 m apart, were set up in all eight catchments in open areas to avoid throughfall. A field composite sample from the five samplers per station was taken monthly, avoiding bags that were visibly contaminated or contained far too much or too little water compared to the others. A large subsample (500 ml) of the field composite sample was left unfiltered and unacidified for the determination of anions, pH and electrical conductivity. A small subsample (100 ml) of the field composite sample was filtered (Millipore™ filter, <0.45 mm) and acidified with ultrapure HNO<sub>3</sub> in the field. Great care was taken to avoid contamination. The number of bags used and the total amount of water (after the two subsamples had been subtracted from the field composite sample) were recorded.

For more details on sampling see Äyräs and Reimann (1995).

## ANALYSIS

All samples were analysed at the GTK laboratory. The GTK laboratory is accredited according to EN45001 and ISO-Guide 25.

### *Snow*

Snow samples were kept frozen at all times after sampling and stored in a freezer after arrival at GTK's laboratory. Each day five samples were taken out of the freezer and hung up in a fume hood. Here a small hole was made in the bottom of the bags and the samples were allowed to melt at room temperature. Meltwater was collected in a Nalgene filter unit, equipped with replaceable cellulose acetate membrane filters (0.45 mm).

The total volume of meltwater was recorded. Then the filtrate was divided into two subsamples. One subsample was used for the determination of pH, electric conductivity (EC, at 25°C), F (by potentiometry), PO<sub>4</sub> (by spectrophotometry) and Br, Cl, NO<sub>3</sub>, SO<sub>4</sub> (by ion chromatography). The other subsample was acidified with suprapure nitric acid (0.5 ml/100 ml sample) and used for cation analysis (Ag, Al, As, B, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Sb, Se, Sr, Th, Tl, U, V and Zn) by ICP-MS and ICP-AES.

The filter paper was digested with 10 ml of concentrated nitric acid in a microwave oven, diluted to 50 ml with water and analysed with ICP-AES. Results were then recalculated to the original sample volume.

### *Rain*

The unfiltered and unacidified samples were used to analyse for Br, Cl, NO<sub>3</sub> and SO<sub>4</sub> by ion chromatography, and for PO<sub>4</sub> by spectrophotometry, and to perform potentiometric determinations of F, pH and EC. The filtered and acidified samples as received from the field teams were analysed by ICP-MS and ICP-AES for Ag, Al, As, B, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Sb, Se, Sr, Th, Tl, U, V and Zn.

## CALCULATION OF DEPOSITION DATA FROM CONCENTRATIONS

The analytical results of the snow and rainwater samples were recalculated to deposition data per area (g/km<sup>2</sup> or kg/km<sup>2</sup>).

For this purpose the following parameters and formulas were used:

### *Snow*

Density of snow cover ( $D_{SN}$ ):

$$D_{SN} \text{ (kg/dm}^3\text{)} = \frac{W_{MW}}{\text{Sum}_L \times S}, \quad (1)$$

where

- $W_{MW}$  - is the weight of meltwater (kg), which, assuming a unit density, is equivalent to its volume (l),
- $\text{Sum}_L$  - is the sum of the core's lengths (dm),
- $S$  - is the cross section of the sampler (dm<sup>2</sup>) =  $\pi \times (0.4625)^2 = 0.67 \text{ dm}^2$ .

Moisture reserve of the snow cover ( $M_{SN}$ ):

$$M_{SN} \text{ (mm/m}^2\text{)} = H_{SN} \times D_{SN} \times 100, \quad (2)$$

where  $H_{SN}$  - is the average snow thickness (dm),  
100 - is the conversion factor to given units.

### Rain

Quantity of precipitation ( $Q_P$ ):

$$Q_P \text{ (l/m}^2\text{)} = \frac{(V_{RW} + 0.6) \times 100}{n \times S}, \quad (3)$$

where  $V_{RW}$  - is the recorded volume of the composite rainwater sample (l),  
0.6 - is the volume of sample subtracted in the field for analysis (l),  
100 - is the conversion factor to given units,  
 $S$  - is the cross section of the rainwater sampler ( $\text{dm}^2$ ) =  $\pi \times (0.633)^2 = 1.26 \text{ dm}^2$ ,  
 $n$  - is the number of subsamples accepted (max. 5).

### Snow & rain

The average monthly deposition data for snow meltwater ( $Q_{MW}$ ), snow filter residue ( $Q_{FR}$ ) and rain ( $Q_{RW}$ ) can then be calculated as:

$$Q_{i,MW} \text{ (g/km}^2\text{ or kg/km}^2\text{)} = M_{SN} \times C_{i,MW}, \quad (4a)$$

$$Q_{i,FR} \text{ (g/km}^2\text{ or kg/km}^2\text{)} = M_{SN} \times C_{i,FR}, \quad (4b)$$

where  $C_{i,MW \text{ or } FR}$  - is the concentration of element  $i$  in MW or FR (mg/l or mg/l, depending on  $i$ );

$$Q_{i,RW} \text{ (g/km}^2\text{ or kg/km}^2\text{)} = Q_P \times C_{i,RW}, \quad (5)$$

where  $C_{i,RW}$  - is the concentration of element  $i$  in RW (mg/l or mg/l, depending on  $i$ ).

Kriging (Matheron, 1967) with a search radius of 2000 m was used to calculate average deposition data per catchment for snow. The monthly element depositions with rainfall were calculated as a mean value for the period of June-October.

Of all the elements analysed for, Fe, S, Ni, Cu, Co, Cr, Al, Mg, Ca, Sr, V, Pb, Zn, As, Cd, Sb and Mo show the strongest influence of anthropogenic contamination and the results obtained for these elements are discussed in the following in more detail.

## RESULTS

### Snow

Average monthly «dry» (FR) and «wet» (MW) deposition for 17 elements during winter 1993-94 is given in Table 3.4.1 for all eight catchments. Both water soluble and particulate deposition data cover several orders of magnitude for most elements between the different



catchments. Figure 3.4.2 compares the monthly deposition values per catchment derived from snow meltwater, snow filter residue and rainwater data.

Dry S deposition is highest in C1, followed by C2. For C5 through to C8, S deposition data are equal to the regional background. Generally, the water-soluble part of the S-deposition is higher than the particulate input, with the exception of C1, where the particulate S-input prevails (see also Åyräs et al. in prep.).

C1 and C2 show strong enrichments in Al (also in C3), As, Cd, Co, Cr, Cu, Fe, Mg, Mo (not in C1), Ni, (Sb), V, Zn in both FR and MW phases when compared to the other catchments. C1 shows the highest deposition figures for Fe, Cr, Ca and Mg. These elements (with the exception of Ca), and, in addition, Cu occur mostly in particulate form here. The highest values for Al, As, Cd, Co, Cu, Mo, Ni, Pb, Sb, V and Zn are detected in C2. Cu, Cd, Mo, Sr and Zn are predominantly in the water soluble phase here. These obvious differences between C1 and C2 reflect differences in industrial activity (C1: roasting and smelting of local Cu-Ni ores, C2: mostly smelting of ores imported from Norilsk in Siberia) and size of the industrial emissions (Monchegorsk >> Zapoljarnij).

The spectrum of elements showing elevated deposition data as observed in C4 (25 km SW of Monchegorsk), is just the same as in C2, but the values are considerably lower here and the proportions between water soluble and particulate phases change in favour of the water soluble phase.

C5 through to C8 are at background levels for all elements, with the exception of Cu, Co and partly Ni, which show more elevated deposition in C5 and C6 than in C7 and C8.

#### *Rain*

Average monthly deposition data for rainwater for the summer of 1994 are given in Table 3.4.2. Figure 3.4.2 gives a graphical comparison between catchments as well as with the winter deposition data. The values of monthly wet deposition of most elements are at a regional background level in C6 through to C8. Compared to these areas, the Russian catchments show very high values for the major ore and accompanying trace elements.

S-deposition during the summer month is highest in C2 (74.9 kg/km<sup>2</sup>) and C1 (44.9 kg/km<sup>2</sup>) followed by C3 and C4 (both ca. 33 kg/km<sup>2</sup>).

The highest deposition data for Ni, Cu, Co, As, Pb, Cd, Mo, Sb and V in rainfall were registered in C2. These are clearly enriched in C1 as well. Mg, Ca, Fe and Zn show the strongest enrichment in C1. This can again be explained by differences in type and size of industry in the two areas.

In C4 the levels of wet deposition for the main ore elements are comparable to the level of these elements in C1, despite of the significant difference in their distance to pollution sources (25 vs. 10 km). Deposition levels for the elements As, Pb, Cd, Co, Cu, Mo and Sb are actually higher in C4 than in C1. This can be explained by the two different ore types used (Pechenga ore at Zapoljarnij and the richer Norilsk ore at Monchegorsk) and is in addition enhanced by the different amounts of ore processed.

Results obtained at C3 (50 km SE from Monchegorsk) are special in so far as the influence of the smelter in Monchegorsk is noticeable (enrichments in Ni, Cu, Co, As, (V?)) and, in

addition there are separate contamination sources close by (huge apatite open cast mine and large coal fired central heating plants near Kirovsk) with their own typical element spectrum (Al, Sr, S, V).

In C5 the influence of the smelter in Nickel can be detected in enhanced deposition levels of Ni, Cu, Co and As in rain when compared to the Finnish catchments.

#### TOTAL YEARLY DEPOSITION

Snow sampling gave the opportunity to estimate both water soluble and particulate (Table 3.4.1) monthly deposition, which is accumulated and preserved in the snowpack during winter time. Element ratios of meltwater to (meltwater + filter) residue as introduced in Reimann et al. (1996) and detailed in Äyräs et al. (in prep.) gave distinct signatures («fingerprints») for each industrial source. The most important feature of these ratios is the consistency with which they change with distance from the smelters. A very similar feature is well known in exploration geochemistry as «zonality of ore deposits». There, it is caused by differences in the dispersion of different elements from their sources during the formation of the deposit. For the snow deposition data we observe a surprisingly comparable situation here, not only with regards to the dependency of total deposition from distance to source, but also with regards to the fractionation of elements.

The change of an element ratio of meltwater (MW) to filter residue (FR) can be described as a function of distance from source:

$$Q_{x,MW}/Q_{x,FR} = (Q_{0,MW}/Q_{0,FR}) \times \exp(dK_{FR-MW} \times X), \quad (6)$$

where

$Q_{0,MW}/Q_{0,FR}$	- is the deposition ratio of MW to FR as observed in snowpack close to source (dimensionless),
$Q_{x,MW}/Q_{x,FR}$	- is the deposition ratio between two phases at a distance $x$ from source (dimensionless),
$dK_{FR-MW}$	- is a measure of the inequality of the coefficients of dispersion ( $K_{FR}$ and $K_{MW}$ ) of element deposition in FR and MW ( $\text{km}^{-1}$ ), and
$X$	- is the distance from the pollution source (km).

The ratio  $Q_{0,MW}/Q_{0,FR}$  is a characteristic parameter of the pollution source itself. The value of  $dK_{FR-MW}$  is a constant parameter for each element and does not depend on other conditions (e.g. reiteration and average monthly wind speeds) of dispersion (Dzagalovskaja and Nazarov, 1981).

Regression analysis (Krumbein and Graybill, 1965) based on diagrams of  $\ln(Q_{MW}/Q_{FR})$  vs.  $x$  was used to calculate  $Q_{0,MW}/Q_{0,FR}$  (Y-intercept) and  $dK_{FR-MW}$  (slope) for two groups of catchments: one under the influence of industrial emissions from Nickel/Zapoljarnij (C1, C5, C6 and C8, the latter serving as background), the other affected by industrial emissions from Monchegorsk (C2, C4, C3 and C7, the latter serving as background). An example of this is shown for Ni in Figure 3.4.3.

In addition, calculations of  $Q_{0,MW}/Q_{0,FR}$  and  $dK_{FR-MW}$  were performed separately for the smelter in Nickel (based on C1 and C5) and ore roasting plant in Zapoljarnij (C1, C5, C6, C8). For two

elements (Al, Sr) individual estimations of these parameters were calculated for C3 (influence of the Kirovsk industrial centre) as well.

All results are presented in Table 3.4.3. A high or low value of the ratio  $Q_{0,MW}/Q_{0,FR}$  indicates the phase (wet or particulate) predominating in the elements dispersion from pollution source. From Table 3.4.3 it can be seen that  $Q_{0,MW}/Q_{0,FR}$  ratios are very different for the sources studied. Elements such as S, Ca, Mg, Sr and Zn occur preferably in water soluble form. Characteristic for the Zaplojarnij industrial district are the lowest ratios for almost all heavy metals and other emitted elements, compared to Nickel. The largest differences between the Monchegorsk and Nickel smelters can be observed for the distribution of Cu, Co, Cd, Sb, (Ni, Fe partly) between the soluble and solid phases of deposition. Kirovsk is marked by a low  $Q_{0,MW}/Q_{0,FR}$  ratio for Sr when compared to Monchegorsk. The constant  $dK_{FR-MW}$  could not be calculated separately for the different industrial districts within each catchment group due to too few data, thus the same value was used in all cases within each group.

With the help of these parameters it is now possible (see Figure 3.4.3) to determine the ratio of water soluble to particular deposition in snow for any of the 17 elements at any point  $x$  ( $Q_{x,MW}/Q_{x,FR}$ ) in the area of influence of the above mentioned industrial centres. This is achievable by solving the linear Equation (6) based on  $Q_{0,MW}/Q_{0,FR}$  and  $dK_{FR-MW}$  given in Table 3.4.3. This ratio will remain constant over time, as long as the technological processes used at the plants are not changed dramatically.

There exists very little information on the comparison of the seasonal winter and summer deposition in the vicinity of the Russian nickel smelters. Data obtained from snow and rainfall sampling in the eight catchments were used to calculate element ratios of deposition in snow meltwater to those observed in rainwater (Reimann et al. 1995, Chekushin et al. 1995). The majority of elements could be divided into two groups, showing different behaviour. The first group of elements (Ni, Cu, Co, Cr, Fe, Sr, Ca, Mg, Ba, Al) shows a strong enrichment in snow meltwater close to the smelters when compared to the rain data. The second group of elements (As, Cd, Mo, Pb, Sb and S) displays an opposite trend in a zone of 50-60 km around the smelters. Deposition figures for these elements are low here in snow when compared to rain, and reach comparable levels in the background areas. This can be explained by the predominance of dry deposition close to the pollution sources and differences in particle size of the different elements emitted to the atmosphere and thus different settling characteristics in winter and summer.

Regression analysis performed on these data (e.g. Figure 3.4.4) shows that, in general, the peculiarities of the element ratios of meltwater (MW) to rainwater (RW) can be satisfactorily described by the following function:

$$Q_{x,MW}/Q_{x,RW} = (Q_{0,MW}/Q_{0,RW}) \times X^B, \quad (7)$$

where

$Q_{0,MW}/Q_{0,RW}$	- is the element ratio of deposition in MW to RW close to a smelter (dimensionless),
$Q_{x,MW}/Q_{x,RW}$	- is the element ratio of deposition in MW to RW at a distance $x$ from a smelter (dimensionless), and
$B$	- is a coefficient, dependent on differences of the element dispersion in MW and RW at the distance $x$ from a smelter.

Results of the calculation of  $Q_{0,MW}/Q_{0,RW}$  and the coefficient B for the Nikel/Zapoljarnij and the Monchegorsk industrial centres are given in Table 3.4.3. The data presented in this table confirm the above mentioned conclusions about the element ratios in MW to RW collected close to the smelters. The physical significance of these parameters is similar to those estimated for the element ratios in meltwater to filter residue of snowpack samples and they can be used as coefficients for the recalculation of deposition data from rainwater to meltwater and vice versa.

It is now possible to estimate total (wet + dry) deposition for any element during summer at a distance  $x$  from source, based only on information about wet deposition and snow deposition:

$$Q_{x,TOT} = Q_{x,wet} + Q_{x,dry}, \quad (8)$$

where  $Q_{x,wet}$  - is wet summer deposition; by definition  $Q_{x,wet} = Q_{x,RW}$ , and  $Q_{x,dry}$  - is dry summer deposition (unknown).

Since  $Q_{x,dry}$  is not known, we use an estimate of wet and dry deposition in summer as equivalents of winter deposition, recalculated as to depend only on previously established constants. Winter equivalent of summer wet deposition ( $Q_{x,RW}$ ) is the value in snow meltwater ( $Q'_{x,MW}$ ), which can be recalculated from  $Q_{x,RW}$  using Equation (7):

$$Q'_{x,MW} = Q_{x,RW} \times (Q_{0,MW}/Q_{0,RW}) \times X^B. \quad (9)$$

Winter equivalent of summer dry deposition ( $Q_{x,dry}$ ) is derived from the calculated filter residue value in winter ( $Q'_{x,FR}$ ) recalculated from  $Q'_{x,MW}$  using Equation (6):

$$Q'_{x,FR} = Q'_{x,MW} \times (Q_{0,FR}/Q_{0,MW}) \times \exp(-dK_{FR-MW} \times X). \quad (10)$$

Combining wet and dry deposition, one obtains total monthly deposition in summer:

$$Q_{x,TOT} = Q'_{x,MW} + Q'_{x,FR} \quad (11)$$

$$Q_{x,TOT} = Q_{x,RW} \times (Q_{0,MW}/Q_{0,RW}) \times X^B \times [1 + (Q_{0,MW}/Q_{0,FR})^{-1} \times \exp(-dK_{FR-MW} \times X)]. \quad (12)$$

Estimations of total monthly element deposition at the eight catchments during the summer of 1994 are presented in Table 3.4.4. For C1, the element deposition originating from the Zapoljarnij ore roasting plant and that originating from the nickel smelter in Nikel (30 km to the SW) were estimated separately. It can be seen that the contribution of the Nikel smelter to the total deposition of S, some trace elements (As, Cd, Sb, Mo, Zn, V) as well as to that of Ca and Mg in C1 is substantial. For C2, C3 and C4 separate estimations were performed for Al and Sr, which originate from both the Monchegorsk smelter and the Kirovsk industrial centre. Most of the observed deposition of Al and Sr in C3 is explained by the industry in the Kirovsk district. In C4 about equal amounts of the observed Sr and Al deposition originates from Monchegorsk and Kirovsk.

Annual deposition data for the studied catchments are calculated as the sum of total element depositions during 5 months of winter (snow sampling) and 7 months of summer (as estimated using Equation [12]). Results are presented in Table 3.4.5.

C1 receives the highest yearly input of S, Fe, Cr, Mg and Ca. In contrast, in C2 yearly input of Cu, Ni, As, Cd, Mo, Pb, Sb and V is highest. C4 takes an intermediate position for almost all elements. C3, C5 and C6 show elevated deposition data for Co, Cu, Fe, Mg and Ni. Annual deposition of Al, Sr and S is high in C3, close to the Kirovsk industrial centre. C7 and C8 in Finland show yearly deposition figures that are well comparable with data from other remote areas throughout the world (Galloway et al., 1982).

## CONCLUSIONS

Analysis of water soluble (meltwater), particulate (filter residues) and total element concentrations in snowpack samples are used to calculate seasonal total deposition data for the eight catchments investigated. Element ratios of water soluble to particulate phases of snow samples are characteristic for each industrial centre and can be used to determine the parameters of fallout dispersion, which are constants for each individual source, as long as the technological process is not changed.

Rainwater sampling («wet only» deposition) is the only practicable approach for a direct measurement of summer deposition in the survey area, considering location, size and remoteness. Results obtained from snow meltwater and rainwater sampling are well comparable in the Finnish background areas, but not, however, in the surroundings of the Russian nickel industry where dry deposition predominates.

The ratio between snow meltwater and rainwater is again a constant for any one element for each pollution source, and changes with distance from source. A power function can be used for modelling the ratio, and, in combination with results obtained from snow sampling, where wet and particulate deposition were determined separately, to predict total deposition data, based on wet only results, for the summer month as well.

Annual deposition loadings have been calculated (up to and can then be used for comparison) and interpretation of results of multimedia sampling, carried out in the catchments studied.

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## TABLES

Table 3.4.1. Average monthly winter atmospheric deposition figures for 17 elements as determined from snowpack samples taken at the end of the winter 1993/94, separated after water soluble (meltwater, MW) and particular (filter residue, FR) deposition.

Table 3.4.2. Average monthly summer atmospheric deposition figures for 17 elements as observed from (filtered) rainwater sampled from June to November 1994 in the eight catchments investigated.

Table 3.4.3. Parameters characterising the dispersion of pollutants from Nikel (Nk), Zaplojarnij (Zp), Monchegorsk (Mn) and Kirovsk (Kr). See text for calculation details.

Table 3.4.4. Average monthly atmospheric deposition figures during the summer of 1994 differentiated after source: Nikel (Nk), Zaplojarnij (Zp), Monchegorsk (Mn) and Kirovsk (Kr).

Table 3.4.5. Total winter, summer and annual atmospheric deposition values for 17 elements in the eight catchments studied.

## FIGURES

Figure 3.4.1. Location of the study area for regional geochemical mapping (frame) and of the eight catchments discussed herein (1: Zaplojarnij, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska and 8: Pallas). K: Kirovsk, Ki: Kirkenes (open-pit iron mine and mill), M: Monchegorsk (nickel-copper smelter, cobalt smelter, etc.), Mu: Murmansk, N: Nikel (nickel-copper smelter), Z: Zaplojarnij (nickel-copper ore roasting plant).

Figure 3.4.2. Comparison of the average monthly atmospheric deposition values in summer (rain water) and winter (differentiated after filter residue, i.e. particular deposition, and meltwater) in the eight catchments studied.

Figure 3.4.3. Scatter diagram of  $\ln(Q_{MW}/Q_{FR})$  vs. distance from source for the two groups of catchments: C1, C5, C6 and C8 (solid squares), and C2, C3, C4 and C7 (open circles), showing the results of linear regressions for Ni.

Figure 3.4.4. Scatter diagram of  $Q_{MW}/Q_{RW}$  vs. distance from source for the two groups of catchments: C1, C5, C6 and C8 (solid squares), and C2, C3, C4 and C7 (open circles), showing the results of power-law regressions for Ni.

*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
 Catchment locations

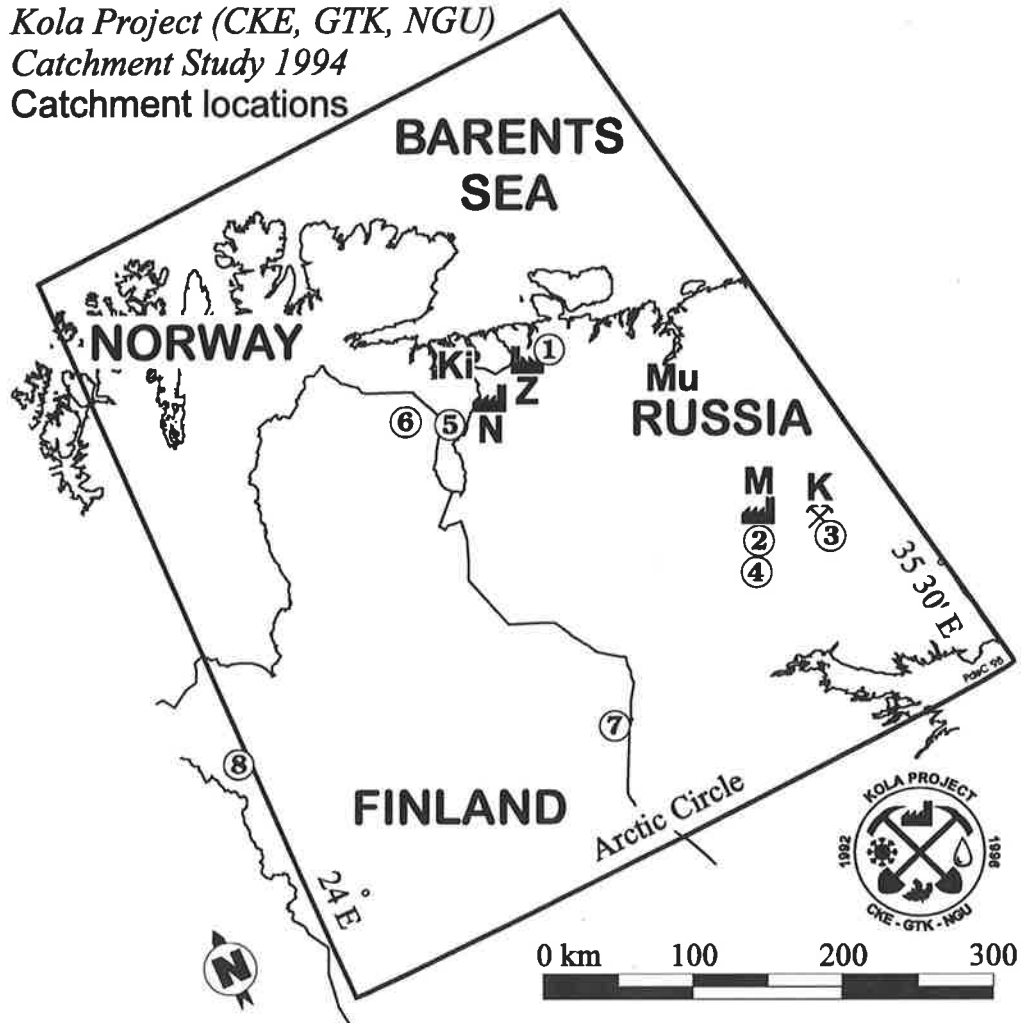


Figure 3.4.1. Location of the study area for regional geochemical mapping (frame) and of the eight catchments discussed herein (1: Zaplojarnij, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska and 8: Pallas). K: Kirovsk, Ki: Kirkenes (open-pit iron mine and mill), M: Monchegorsk (nickel-copper smelter, cobalt smelter, etc.), Mu: Murmansk, N: Nikel (nickel-copper smelter), Z: Zapoljarnij (nickel-copper ore roasting plant).



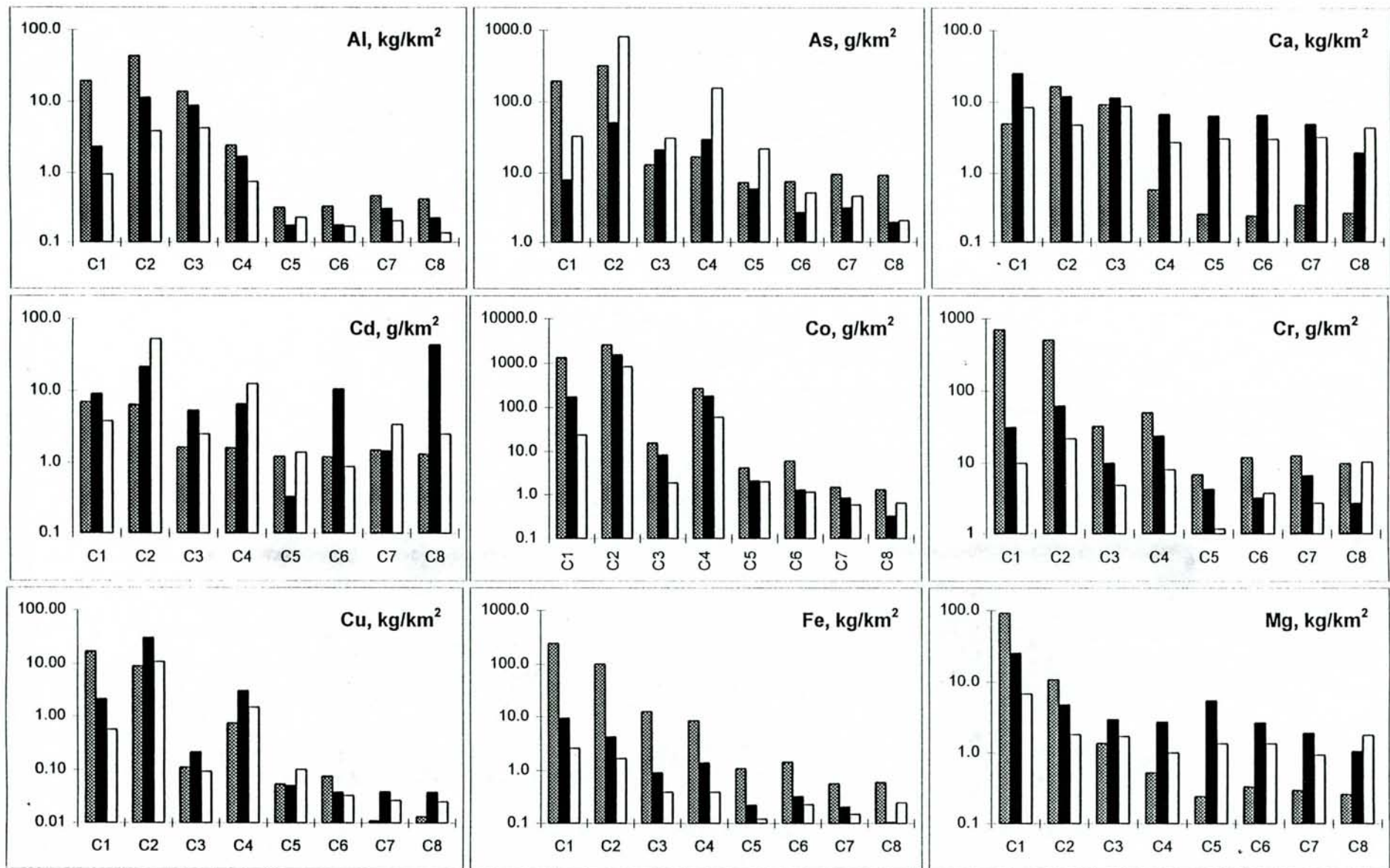


Figure 3.4.2. Comparison of the average monthly atmospheric deposition values in summer (rain water) and winter (differentiated after filter residue, i.e. particular deposition, and meltwater) in the eight catchments studied.

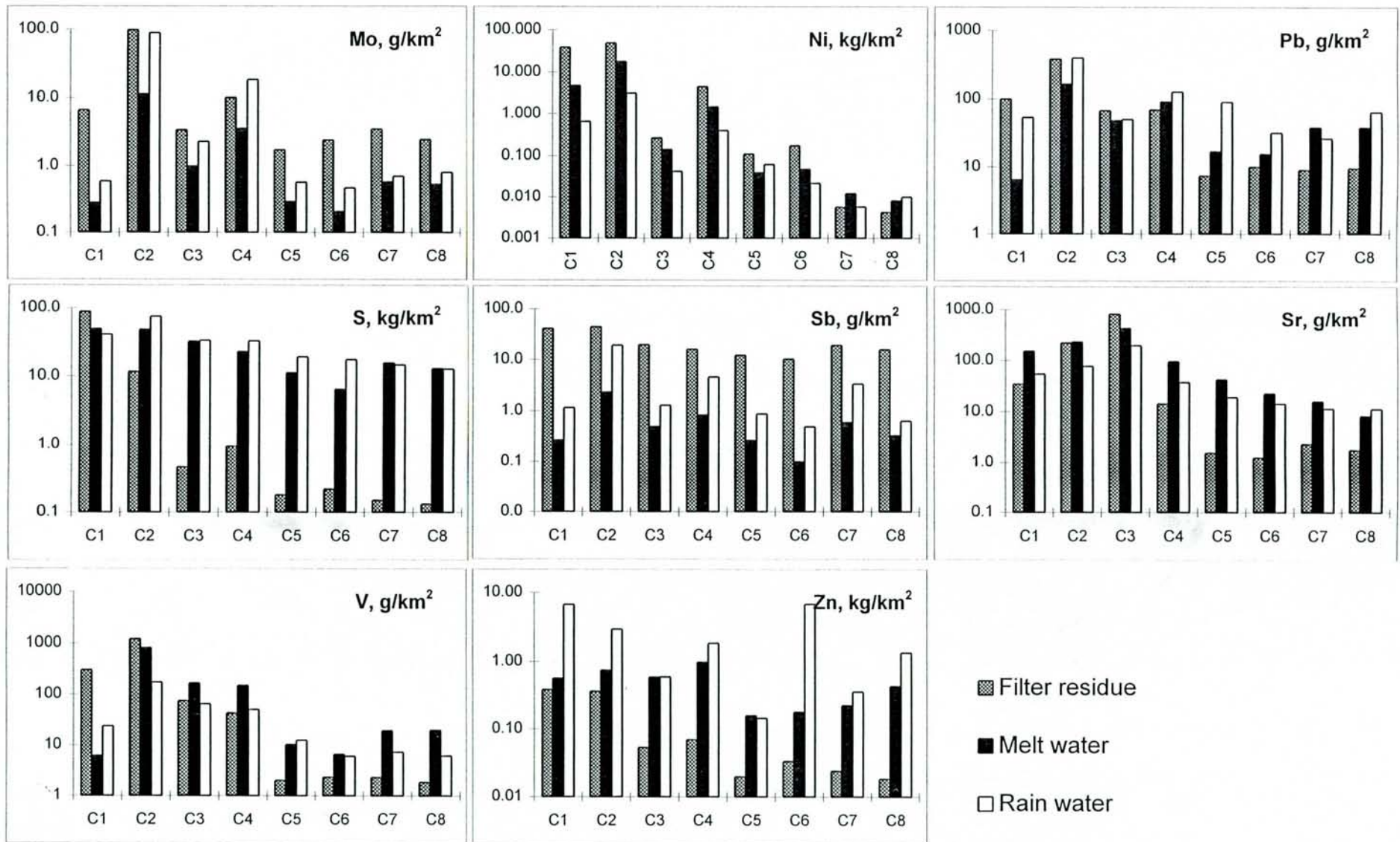


Figure2

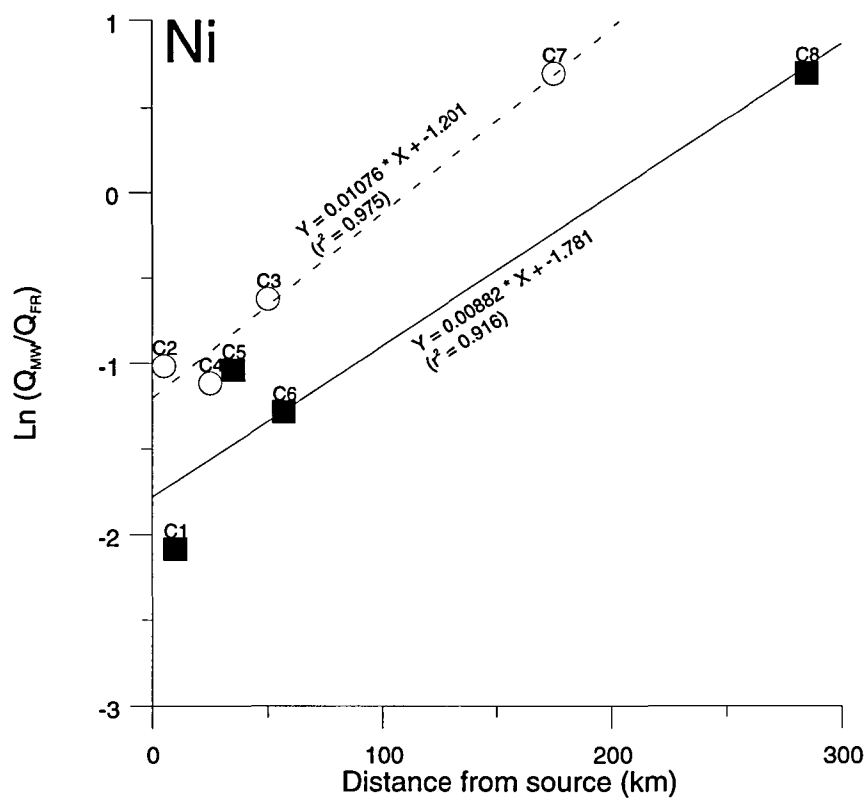


Figure 3.4.3. Scatter diagram of  $\ln(Q_{MW}/Q_{FR})$  vs. distance from source for the two groups of catchments: C1, C5, C6 and C8 (solid squares), and C2, C3, C4 and C7 (open circles), showing the results of linear regressions for Ni.

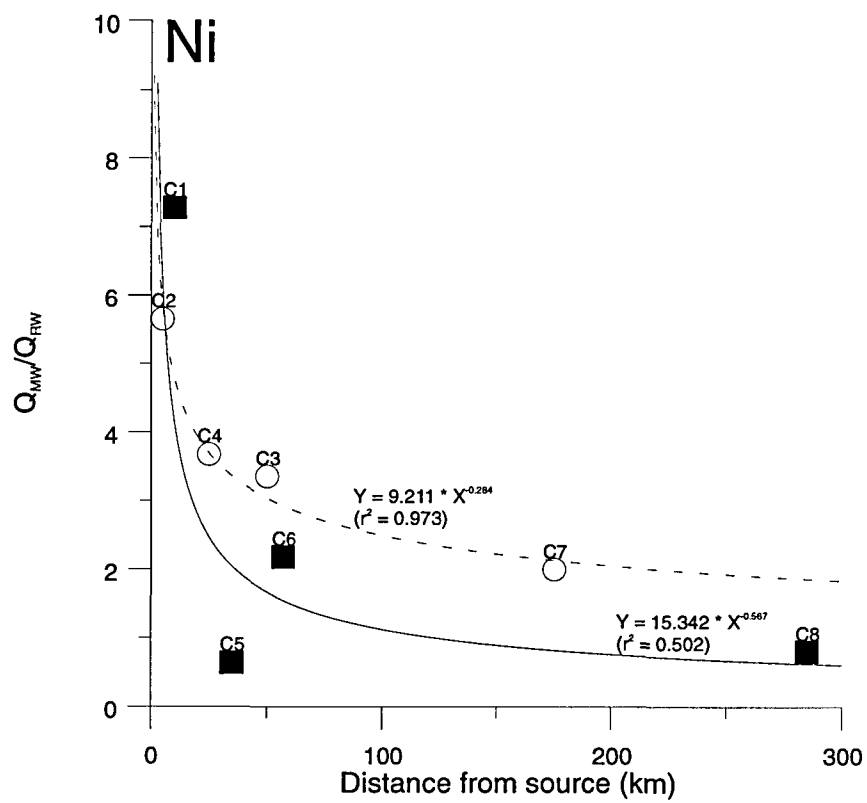


Figure 3.4.4. Scatter diagram of  $Q_{MW}/Q_{RW}$  vs. distance from source for the two groups of catchments: C1, C5, C6 and C8 (solid squares), and C2, C3, C4 and C7 (open circles), showing the results of power-law regressions for Ni.

Table 3.4.1. Average monthly winter atmospheric deposition figures for 17 elements as determined from snowpack samples taken at the end of the winter 1993/94, separated after water soluble (meltwater, MW) and particular (filter residue, FR) deposition.

Catchment		C1	C2	C3	C4	C5	C6	C7	C8
Al	MW	2.30	11.4	8.82	1.68	0.174	0.175	0.305	0.224
	kg/km <sup>2</sup> FR	19.5	43.6	13.9	2.40	0.316	0.329	0.470	0.421
As	MW	7.95	51.0	21.0	29.2	5.94	2.69	3.13	1.92
	g/km <sup>2</sup> FR	195	322	13.0	16.8	7.31	7.56	9.54	9.28
Ca	MW	25.1	12.0	11.5	6.73	6.39	6.61	4.91	1.92
	kg/km <sup>2</sup> FR	4.94	16.6	9.18	0.581	0.254	0.239	0.344	0.262
Cd	MW	9.08	21.5	5.32	6.52	0.326	10.6	1.45	43.5
	g/km <sup>2</sup> FR	6.93	6.40	1.63	1.60	1.22	1.20	1.49	1.31
Co	MW	170	1556	8.26	178	2.11	1.29	0.834	0.322
	g/km <sup>2</sup> FR	1338	2588	15.6	266	4.23	6.08	1.49	1.31
Cr	MW	31.4	61.9	10.0	24.0	4.23	3.17	6.65	2.65
	g/km <sup>2</sup> FR	703	510	32.6	50.2	6.86	12.0	12.6	9.95
Cu	MW	2.12	30.8	0.212	3.09	0.051	0.038	0.039	0.038
	kg/km <sup>2</sup> FR	17.2	9.07	0.111	0.738	0.054	0.076	0.010	0.013
Fe	MW	9.32	4.15	0.901	1.39	0.220	0.325	0.204	0.105
	kg/km <sup>2</sup> FR	243	100	12.4	8.35	1.09	1.44	0.561	0.598
Mg	MW	25.5	4.70	2.88	2.65	5.38	2.60	1.87	1.04
	kg/km <sup>2</sup> FR	91.1	10.8	1.34	0.525	0.244	0.334	0.299	0.262
Mo	MW	0.280	11.4	0.988	3.52	0.289	0.206	0.576	0.530
	g/km <sup>2</sup> FR	6.63	97.3	3.35	10.0	1.71	2.38	3.43	2.44
Ni	MW	4.72	17.4	0.141	1.46	0.039	0.048	0.012	0.008
	kg/km <sup>2</sup> FR	38.0	48.0	0.263	4.45	0.110	0.173	0.006	0.004
Pb	MW	6.46	164	47.6	90.8	16.8	15.4	37.6	37.7
	g/km <sup>2</sup> FR	98.7	381	66.2	68.4	7.31	9.94	8.96	9.47
S	MW	49.5	48.4	32.0	22.8	11.1	6.43	15.6	13.0
	kg/km <sup>2</sup> FR	87.9	11.7	0.465	0.949	0.179	0.217	0.149	0.131
Sb	MW	0.268	2.29	0.487	0.823	0.264	0.099	0.594	0.334
	g/km <sup>2</sup> FR	41.2	44.3	19.6	16.0	12.2	10.3	19.0	15.8
Sr	MW	151	230	425	95.8	42.3	22.5	15.7	8.19
	g/km <sup>2</sup> FR	34.2	220	817	14.1	1.55	1.24	2.34	1.78
V	MW	6.18	816	165	149	10.2	6.53	19.2	19.7
	g/km <sup>2</sup> FR	303	1213	73.9	42.5	2.01	2.34	2.32	1.83
Zn	MW	0.557	0.735	0.580	0.975	0.157	0.178	0.224	0.427
	kg/km <sup>2</sup> FR	0.379	0.357	0.053	0.070	0.020	0.033	0.024	0.018

Table 3.4.2. Average monthly summer atmospheric deposition figures for 17 elements as observed from (filtered) rainwater sampled from June to November 1994 in the eight catchments investigated.

		C a t c h m e n t N (number of samples)							
		C1(10)	C2(17)	C3(15)	C4(16)	C5(15)	C6(9)	C7(15)	C8(12)
<b>Al</b>	kg/km <sup>2</sup>	0.951	3.84	4.25	0.750	0.226	0.168	0.202	0.135
<b>As</b>	g/km <sup>2</sup>	32.6	806	30.6	157	21.6	5.21	4.67	2.04
<b>Ca</b>	kg/km <sup>2</sup>	8.43	4.77	8.75	2.67	3.03	2.98	3.18	4.35
<b>Cd</b>	g/km <sup>2</sup>	3.79	52.7	2.5	12.5	1.39	0.880	3.42	2.5
<b>Co</b>	g/km <sup>2</sup>	23.6	829	1.89	59.5	2.01	1.14	0.580	0.640
<b>Cr</b>	g/km <sup>2</sup>	9.95	21.9	4.8	8.06	1.16	3.68	2.65	10.4
<b>Cu</b>	kg/km <sup>2</sup>	0.560	11.0	0.093	1.49	0.102	0.033	0.027	0.025
<b>Fe</b>	kg/km <sup>2</sup>	2.55	1.65	0.390	0.390	0.120	0.225	0.148	0.245
<b>Mg</b>	kg/km <sup>2</sup>	6.69	1.77	1.67	0.990	1.32	1.32	0.930	1.74
<b>Mo</b>	g/km <sup>2</sup>	0.590	88.6	2.26	18.4	0.565	0.468	0.696	0.802
<b>Ni</b>	kg/km <sup>2</sup>	0.649	3.08	0.042	0.397	0.061	0.022	0.006	0.010
<b>Pb</b>	g/km <sup>2</sup>	52.9	395	49.5	126	88.8	31.5	25.9	62.9
<b>S</b>	kg/km <sup>2</sup>	41.1	74.9	33.2	32.7	19.1	17.5	14.7	12.8
<b>Sb</b>	g/km <sup>2</sup>	1.14	19.1	1.28	4.57	0.867	0.488	3.35	0.630
<b>Sr</b>	g/km <sup>2</sup>	53.6	76.8	195	37.3	18.9	14.2	11.3	11.2
<b>V</b>	g/km <sup>2</sup>	23.6	173	64.3	49.8	12.4	6.04	7.29	6.13
<b>Zn</b>	kg/km <sup>2</sup>	6.67	2.93	0.588	1.84	0.144	6.75	0.353	1.33

Table 3.4.3. Parameters characterising the dispersion of pollutants from Nikel (Nk), Zaplojarnij (Zp), Monchegorsk (Mn) and Kirovsk (Kr). See text for calculation details.

		C1, C5, C6, C8				C2, C3, C4, C7				
		$Q_{0,mw}/Q_{0,rw}$	B	$Q_{0,mw}/Q_{0,fr}$	$dK_{fr-mw}$ (km <sup>-1</sup> )	$Q_{0,mw}/Q_{0,rw}$	B	$Q_{0,mw}/Q_{0,fr}$	$dK_{fr-mw}$ (km <sup>-1</sup> )	
Al	Zp	1.7701	-0.0715	0.1212	0.00266	Kr	4.0635	-0.185	0.8179	0.00291
	Nk	1.7701	-0.0715	0.2848	0.00266	Mn	4.0635	-0.185	0.4356	0.0291
As	Zp	0.0809	0.4268	0.0400	0.000714	Kr				
	Nk	0.0809	0.4268	0.2074	0.000714	Mn	0.0221	0.7208	0.2645	0.00337
Ca	Zp					Kr				
	Nk	14.5312	-0.5714	15.4097	0.002	Mn	3.3791	-0.167	1.5857	0.0124
Cd	Zp	0.1557	0.7851	1.1163	0.013	Kr				
	Nk	0.1557	0.7851	0.9017	0.013	Mn	0.4540	0.1087	4.3623	0.00829
Co	Zp	26.8782	-0.7529	0.1225	0.00041	Kr				
	Nk	26.8782	-0.7529	0.2306	0.00041	Mn	2.7114	-0.0306	0.8245	0.0006
Cr	Zp	29.6756	-0.8123	0.0793	0.00212	Kr				
	Nk	29.6756	-0.8123	0.1710	0.00212	Mn	3.0931	-0.0525	0.2181	0.00558
Cu	Zp	2.8655	-0.1946	0.0659	0.00856	Kr				
	Nk	2.8655	-0.1946	0.2536	0.00856	Mn	3.7914	-0.17	3.0253	0.00094
Fe	Zp	17.3221	-0.6425	0.0420	0.00235	Kr				
	Nk	17.3221	-0.6425	0.1052	0.00235	Mn	4.2209	-0.1736	0.0598	0.01031
Mg	Zp	20.3798	-0.5887	0.7634	0.00202	Kr				
	Nk	20.3798	-0.5887	3.0557	0.00202	Mn	3.1477	-0.0999	1.2141	0.0103
Mo	Zp	0.3567	0.09476	0.0729	0.00402	Kr				
	Nk	0.3567	0.09476	0.1225	0.00402	Mn	0.0468	0.5412	0.1444	0.00086
Ni	Zp	17.3420	-0.5806	0.0907	0.00865	Kr				
	Nk	17.3420	-0.5806	0.1655	0.00865	Mn	8.8982	-0.2693	0.2982	0.01122
Pb	Zp	0.0413	0.4988	0.0648	0.00887	Kr				
	Nk	0.0413	0.4988	0.4156	0.00887	Mn	0.1491	0.5357	0.5488	0.01156
S	Zp	0.8441	-0.0426	0.4916	0.01086	Kr				
	Nk	0.8441	-0.0426	6.2651	0.01086	Mn	0.4897	0.1497	11.4730	0.01438
Sb	Zp	0.1210	0.2302	0.0096	0.00252	Kr				
	Nk	0.1210	0.2302	0.0142	0.00252	Mn	0.1120	0.1601	0.0452	0.00271
Sr	Zp	8.2262	-0.4144	13.8184	0.00333	Kr	4.5719	-0.2109	3.6219	0.00829
	Nk	8.2262	-0.4144	13.8184	0.00333	Mn	4.5719	-0.2109	20.5529	0.00829
V	Zp	19.0634	0.7397	0.0739	0.01333	Kr				
	Nk	19.0634	0.7397	0.3631	0.01333	Mn	5.6859	-0.1724	1.2448	0.01137
Zn	Zp	0.0340	0.2308	4.4282	0.00759	Kr				
	Nk	0.0340	0.2308	2.9683	0.00759	Mn	0.1904	0.2186	5.6916	0.004

Catchments		C1	C5	C6	C8		C2	C3	C4	C7
<b>Al</b> kg/km <sup>2</sup>	Zp	12.9				Kr	15.6	31.2	3.04	
	Nk	5.60	1.30	0.892	0.422	Mn	34.6	12.9	3.54	0.320
<b>As</b> g/km <sup>2</sup>	Zp	182				Kr				
	Nk	64.4	45.4	13.3	9.09	Mn	269	47.7	158	13.3
<b>Ca</b> kg/km <sup>2</sup>	Zp	16.9				Kr				
	Nk	18.6	6.12	4.55	2.59	Mn	19.6	20.6	7.71	4.86
<b>Cd</b> g/km <sup>2</sup>	Zp	6.43				Kr				
	Nk	14.9	6.01	5.01	33.8	Mn	34.8	2.00	9.55	2.87
<b>Co</b> g/km <sup>2</sup>	Zp	1023				Kr				
	Nk	259	19.6	7.64	1.19	Mn	4728	9.90	321	2.81
<b>Cr</b> g/km <sup>2</sup>	Zp	607				Kr				
	Nk	121	12.3	25.3	13.1	Mn	340	54.0	105	17.0
<b>Cu</b> kg/km <sup>2</sup>	Zp	15.3				Kr				
	Nk	3.35	0.573	0.149	0.032	Mn	42.2	0.238	4.32	0.054
<b>Fe</b> kg/km <sup>2</sup>	Zp	244				Kr				
	Nk	49.0	2.07	2.70	0.659	Mn	88.9	9.16	13.1	0.956
<b>Mg</b> kg/km <sup>2</sup>	Zp	80.3				Kr				
	Nk	24.1	4.34	3.22	1.51	Mn	8.4744	5.31	3.70	1.98
<b>Mo</b> g/km <sup>2</sup>	Zp	3.71				Kr				
	Nk	2.39	2.28	1.84	1.76	Mn	78.3	6.71	38.3	3.71
<b>Ni</b> kg/km <sup>2</sup>	Zp	32.8				Kr				
	Nk	8.85	0.735	0.167	0.010	Mn	74.0	0.380	5.24	0.019
<b>Pb</b> g/km <sup>2</sup>	Zp	104				Kr				
	Nk	33.9	59.7	23.9	51.9	Mn	380	121	250	76.2
<b>S</b> kg/km <sup>2</sup>	Zp	88.8				Kr				
	Nk	33.5	15.4	13.5	8.55	Mn	50.5	30.4	27.5	15.7
<b>Sb</b> g/km <sup>2</sup>	Zp	24.1				Kr				
	Nk	20.0	15.6	9.30	9.91	Mn	63.2	5.45	18.6	12.7
<b>Sr</b> g/km <sup>2</sup>	Zp	182				Kr	182	899	88.4	
	Nk	115	37.9	23.2	9.10	Mn	262	404	89.9	17.6
<b>V</b> g/km <sup>2</sup>	Zp	87.3				Kr				
	Nk	43.6	24.6	14.4	22.3	Mn	1308	271	261	18.9
<b>Zn</b> kg/km <sup>2</sup>	Zp	0.466				Kr				
	Nk	0.630	0.014	0.711	0.173	Mn	0.930	0.301	0.819	0.226

Table 3.4.4. Average monthly atmospheric deposition figures during the summer of 1994 differentiated after source: Nikel (Nk), Zaplojarnij (Zp), Monchegorsk (Mn) and Kirovsk (Kr).



Table 3.4.5. Total winter, summer and annual atmospheric deposition values for 17 elements in the eight catchments studied.

Catchment		C1	C2	C3	C4	C5	C6	C7	C8
Area	km <sup>2</sup>	19.02	22.38	20.01	20.49	34.56	11.86	20.15	24.42
Al	Winter	109	275	113	20.4	2.45	2.52	3.87	3.22
	Summer	129	351	308	46.1	9.12	6.24	2.24	2.95
	Annual	238	626	421	66.5	11.6	8.76	6.12	6.18
As	Winter	1016	1863	170	230	66.3	51.3	63.3	56.0
	Summer	1726	1881	334	1105	318	93.3	92.8	63.6
	Annual	2741	3744	504	1335	384	145	156	120
Ca	Winter	150	143	103	36.5	33.2	34.3	26.3	10.9
	Summer	248	137	144	54.0	42.9	31.8	34.0	18.1
	Annual	399	280	248	90.5	76.1	66.1	60.3	29.1
Cd	Winter	80.0	140	34.7	40.6	7.73	58.9	14.7	224
	Summer	149	243	14.0	66.9	42.1	35.1	20.1	237
	Annual	230	383	48.7	107	49.8	94.0	34.8	461
Co	Winter	7537	20719	119	2220	31.7	36.8	11.6	8.16
	Summer	8975	33095	69.3	2246	137	53.5	19.7	8.30
	Annual	16512	53814	189	4466	169	90.4	31.3	16.5
Cr	Winter	3670	2861	213	371	55.5	76.0	96.3	63.0
	Summer	5094	2379	378	735	86.3	177	119	91.4
	Annual	8764	5240	592	1106	142	253	216	154
Cu	Winter	96.3	199	1.62	19.1	0.52	0.57	0.244	0.257
	Summer	130	295	1.66	30.2	4.01	1.04	0.381	0.226
	Annual	227	494	3.28	49.4	4.53	1.62	0.626	0.483
Fe	Winter	1264	522	66.7	48.7	6.54	8.80	3.82	3.51
	Summer	2051	622	64.1	91.7	14.5	18.9	6.69	4.61
	Annual	3315	1144	131	140	21.0	27.7	10.5	8.13
Mg	Winter	583	77.4	21.1	15.9	28.1	14.7	10.8	6.50
	Summer	730	59.3	37.1	25.9	30.3	22.5	13.9	10.5
	Annual	1313	137	58.2	41.8	58.4	37.2	24.7	17.0
Mo	Winter	34.5	543	21.7	67.7	10.0	12.9	20.0	14.9
	Summer	42.7	548	47.0	268	16.0	12.8	26.0	12.3
	Annual	77.3	1091	68.7	336	26.0	25.8	46.0	27.2
Ni	Winter	214	327	2.02	29.6	0.75	1.10	0.089	0.407
	Summer	292	518	2.66	36.7	5.14	1.17	0.131	0.069
	Annual	506	845	4.68	66.3	5.89	2.27	0.220	0.475
Pb	Winter	526	2727	569	796	121	126	233	236
	Summer	966	2657	850	1749	418	168	534	363
	Annual	1492	5384	1418	2544	538	294	767	599
S	Winter	687	300	162	119	56.5	33.3	78.6	65.5
	Summer	856	353	213	193	108	94.5	110	59.9
	Annual	1543	654	375	311	164	128	189	125
Sb	Winter	207	233	100	83.9	62.3	51.9	98.2	80.7
	Summer	309	442	38.1	130	109	65.1	88.6	69.4
	Annual	517	675	139	214	171	117	187	150
Sr	Winter	924	2248	6212	549	219	119	90.3	49.8
	Summer	2075	3106	9120	1248	265	162	123	63.7
	Annual	2999	5353	15332	1797	485	281	213	114
V	Winter	1545	10144	1193	955	60.9	44.4	108	108
	Summer	916	9156	1897	1826	172	101	132	156
	Annual	2461	19300	3090	2781	233	145	240	264
Zn	Winter	4.68	5.46	3.17	5.23	0.89	1.06	1.24	2.23
	Summer	7.67	6.51	2.11	5.73	0.098	4.98	1.58	1.21
	Annual	12.4	12.0	5.27	11.0	0.98	6.03	2.82	3.44

## 4. STREAM WATER

### 4.1 Stream water geochemistry from selected catchments on the Kola Peninsula (NW Russia) and in neighbouring areas of Finland and Norway: 1. Element levels and sources

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#### ABSTRACT

The pH, electrical conductance and concentration of six anions and 33 cations were determined in stream waters sampled in 1994 from eight arctic catchments on and adjacent to the Kola Peninsula (Russia, Finland and Norway). The catchments are located at various distances (5-300 km) from the major industrial emissions centres of Monchegorsk, Nickel and Zapoljarniy in Russia. The median concentration of any constituent can vary by up to three orders of magnitude from polluted to remote locations. The source of these elements can be (1) natural, marine (sea spray), (2) natural, geogenic (blowing soil/dust, till, bedrock), and/or (3) anthropogenic, industrial (emissions from smelters, dust from mining, etc.), each source showing a distinct signature. Ni is strongly correlated to As, Cd, Co and Cu in the most polluted stream, suggesting a common origin (industrial emissions). Elevated trace metal contents in the most polluted streams indicates that deposited contaminants are transferred to the aquatic ecosystem, and eventually can reach the ocean. Factor analysis distinguished three geogenic-marine factors and one, dominant technogenic factor, together explaining 81% of the total variance.

#### INTRODUCTION

The western half of the Kola Peninsula (NW Russia) is a heavily industrialised area, and has common borders with northernmost Finland and Norway. Among other industries, nickel-copper ore smelters are found at Monchegorsk and Nickel, and an ore roasting plant at Zapoljarniy (Fig. 4.1.1). These industrial activities have released large amounts of various pollutants (SO<sub>2</sub>, NO<sub>x</sub>, CO, Ni, Cu, Co, Mn, As, Cr, Zn, etc.) (Baklanov et al., 1994) into the atmosphere during the last few decades, leading to locally severe environmental degradation (e.g. Kozlov et al., 1993). In 1994, the year relevant for the present investigation, ca. 98,000 tonnes SO<sub>2</sub> were emitted from the Monchegorsk smelters, ca. 129,000 tonnes from Nickel, and ca. 69,000 tonnes from Zapoljarniy; Cu and Ni emissions were of the order of 934 and 1619 tonnes, respectively, from Monchegorsk (or a value of US \$ 13 million for Ni alone!), and 82 and 136 tonnes from Nickel, and 81 and 161 tonnes from Zapoljarniy (MRCENR, 1995). The Monchegorsk smelter complex started production in 1938 using only local, low-sulphur (ca. 6.5% S) ore, and from 1968 onward, sulphur-rich (up to 30% S) ore from Norilsk has been smelted there also (see Alexeyev (1993) and Pozniakov (1993)). In a cumulative perspective, historical emissions to air from Monchegorsk amount to a staggering 6.5 million tonnes of SO<sub>2</sub> from 1960 to 1994 (data collated from Alexeyev (1993), Pozniakov (1993), Baklanov et al. (1994), and MRCENR (1995)). Emissions have varied over the production years and, in addition, available estimates differ somewhat (compare data in Alexeyev (1993), Pozniakov (1993), Sivertsen (1994a), Baklanov et al. (1994), Jaffe et al. (1995) and the official

MRCENR (1995) release). In any case, atmospheric pollution by industry in the area is one of the worst in the world. It is not surprising, therefore, that trace metal and sulphur contamination of ecosystems in arctic western Russia and neighbouring countries is a problem currently receiving much attention. This is particularly true when it comes to surface waters (e.g. Lahermo et al., 1994, Moiseenko et al., 1995, Rodushkin et al., 1995), which are used as the main drinking water source in the region.

Following the completion of a pilot project (1992-1993) focusing on environmental pollution around Nickel (Niskavaara et al., 1996, Väisänen et al., in prep.), the Central Kola Expedition (CKE) and the Geological Surveys of Finland (GTK) and Norway (NGU) are carrying out (1994-1996) a major, co-operative regional geochemical mapping project of a 188,000 km<sup>2</sup> area north of the Arctic Circle on the Kola Peninsula (NW Russia) and in neighbouring areas of Finland and Norway (Fig. 4.1.1). More information about the «Kola Ecogeochemistry Project» is posted on the world wide web (<http://www.ngu.no/Kola>). The aims of the main project are: (1) to establish regional geochemical maps of the distribution of about 40 elements and radionuclides in an ecologically vulnerable area, (2) to build up a soil sample data bank for future study, and (3) to assess the environmental impact of the nickel smelting and other heavy industry in the survey area. Here and in the companion paper (Caritat et al., in prep. a), part of the results of a detailed study and monitoring of eight different areas (catchments) within the regional geochemical mapping area is presented. The catchment study was undertaken to investigate in greater detail the source, residence and fate of elements cycling through catchments that differ greatly in degree of contamination, in proximity to the sea, and nature of underlying bedrock. In this paper, we address specifically the differences in stream water composition between these eight catchments and the possible sources for various elements.

## STUDY SITES AND SAMPLING

As part of the main geochemical project, element content in various sampling media (rain water, snow, moss, organic stream sediments, stream water, ground water, overbank sediments, different soil horizons, Quaternary deposits and bedrock) was determined and/or monitored during 1994 in eight catchments (watersheds or drainage basins) located at various distances from the smelters (5-300 km), and in different geological settings. Four of these catchments are situated in Russia (*C1*: Zapoljarniy, *C2*: Monchegorsk, *C3*: Kirovsk, *C4*: Kurka), one in Norway (*C5*: Skjellbekken) and three in Finland (*C6*: Kirakka, *C7*: Naruska, *C8*: Pallas) (Fig. 4.1.1, Tab. 4.1.1).

*C1* is located in the vicinity of a nickel-copper ore roasting plant (Zapoljarniy), and is the closest catchment to the sea. *C2* and *C4* are found adjacent (5 km) to, and slightly more distant (25 km) from, respectively, the major sulphur dioxide and trace metal emission point-source of the whole study area (the nickel-copper ore smelter in Monchegorsk). *C3* is off-wind from Monchegorsk, but near a large open-pit apatite mine and processing plant (Kirovsk). *C5* is relatively close to the Nickel smelter centre, but off the predominant wind direction; it is relatively close to the sea. *C6* is located a little further away from Nickel than *C5* and has a very different lithology. *C7* and *C8* are remote from any major point-source emitter of contamination, and can be considered the most background catchments of this study. An overview of the major characteristics of the catchments is given in Table 4.1.1. In Russia, main wind directions are from the north and south (Mäkinen, 1994), spreading most contaminated air primarily in these directions. The stream water samples discussed here were collected near the outlet of each watershed approx. every 4-6 weeks in five catchments and weekly in *C2*, *C5* and *C8*, between March and November 1994.

## METHODS

Two subsamples of stream water were taken in the field at each site, one (500 ml) unacidified and unfiltered, the other (60 ml) filtered (0.45 µm) and acidified with suprapure nitric acid. At the GTK laboratory, the unacidified subsamples were used for the determination of pH, electrical conductance (EC), five anions (Br, Cl, F, NO<sub>3</sub> and SO<sub>4</sub>) by ion chromatography (IC) and PO<sub>4</sub> by spectrophotometry. The acidified subsamples were used for the determination of six major cations (Ca, Mg, Na, P, S and Si) by inductively coupled plasma-atomic emission spectrometry (ICP-AES) and of 27 trace cations (Ag, Al, As, B, Ba, Be, Bi, Cd, Co, Cr, Cu, Fe, K, Li, Mn, Mo, Ni, Pb, Rb, Sb, Se, Sr, Th, Tl, U, V and Zn) by inductively coupled plasma-mass spectrometry (ICP-MS). Detection limits are given in Table 4.1.2. For more information about the methods, accuracy and precision, see Äyräs and Reimann (1995) and Niskavaara (1995). In addition to the presently reported analytical round, the sulphur isotopic composition of stream water sulphate was determined once in each catchment (Caritat et al., in prep. b).

## QUALITY ASSURANCE/QUALITY CONTROL

The chemical laboratory of the Geological Survey of Finland is accredited to meet the requirements of the EN 45001 standard and the ISO Guide 25 (Niskavaara, 1995).

## RESULTS AND PRESENTATION

Results are summarised in Table 4.1.2, and illustrated for a selection of elements in Figure 4.1.2 using boxplots. A boxplot shows the distribution or structure of the data: the closed box, delimited by a lower hinge, LH (the 25th percentile), and an upper hinge, UH (the 75th percentile), contains the two inner quartiles or 50% of the data; the hinge spread HS is equivalent to (UH - LH); the median (the 50th percentile) is represented by the strike inside the box (or a cross when it coincides with one of the hinges); the lower and upper whiskers ('T' bars), in combination with the outlier symbols where applicable, show the spread of the outer quartiles of the data; the near outliers (squares) and far outliers (plus signs) are data points respectively more than 1.5 and 3 hinge-spreads away from a hinge; the notches (square brackets), placed at  $1.58 \times (HS / \sqrt{n})$  on either side of the median, are a test of significance of medians from different populations or boxes (for more details, see Tuckey, 1977, Reimann and Wurzer, 1986, and O'Connor and Reimann, 1993). Data analysis is performed with values below detection set to one half the detection limit.

## DISCUSSION

### *Concentration levels*

The range of stream water concentrations found between catchments (i.e. as a function of space) for many elements, particularly those that are part of the industrial emission spectrum, is very large (medians can vary by up to three orders of magnitude for a given element), while the range found within catchments (i.e. as a function of time) is much more limited (Fig. 4.1.2). For instance, median Ni and Cu are respectively 1115 times and 303 times higher in C2 than in C7, whilst *minimum* Ni and Cu values in C2 are respectively 252 times and 75 times greater than *maximum* values in C7, i.e. populations do not overlap at all (Fig. 4.1.2, Tab. 4.1.2). This indicates that: (1) the areal extent of intense environmental pollution is limited and restricted to the vicinity of the point-source emitters, and (2) a profound and lasting disturbance has been created by atmospheric pollution and ensuing ecosystem contamination in some places.

Highest median values (Fig. 4.1.2, Tab. 4.1.2) were recorded at *C1* for Cl and Na, *C2* for Ag, B, Cd, Co, Cr, Cu, K, Mg, Mn, Ni, NO<sub>3</sub>, Pb, S, Sb, SO<sub>4</sub>, V and Zn, *C3* for F, Mo, Rb and Sr, *C4* for Ba, *C5* for As and Ca, *C7* for Al and *C8* for Fe, Li, Pb, Si and U. Highest and lowest median pH values are from *C5* and *C7*, respectively, while highest and lowest median EC values are from *C2* and *C7*, respectively.

High median concentrations of Na and Cl in stream water at *C1* are sourced mostly from the nearby Barents Sea, and transported to the catchment as sea spray, probably to a large extent as a result of winter storm events. These elements can, however, also be part of the emission spectrum of the smelting industry at Nikel (Reimann et al., 1996), and Cl is reported as an emitted pollutant from Monchegorsk (341 tonnes in 1994, MRCENR, 1995).

The highest median levels of Cd, Co, Cr, Cu, Ni, S, SO<sub>4</sub> and V, in stream water from *C2*, and the generally unusually high content in these elements in *C1* and *C4* as well, are interpreted to result mainly from pollution from the nearby nickel-copper ore smelting/roasting industry in Monchegorsk, Nikel and Zapoljarniy. However, a minor, natural contribution from the regional bedrock/Quaternary deposits, which in places contains nickel ore, can not be ruled out categorically. In most cases, though, it is clear from the comparison of concentrations in A<sub>0</sub> and C-horizons (Reimann et al., 1995, Niskavaara et al., 1996), that the above-ground, anthropogenic source in these elements in surface environments is much more significant than the subsurface, geogenic source. These elements are also extremely enriched in both snow (Äyräs et al., 1995) and rain water (Reimann et al., in prep. a). The remaining elements with highest median at *C2* are likely to also find their origin in one part or another of the industrial process of ore smelting at Monchegorsk.

Comparing with water data from 'Stream 2' (ca. 30 km southwest of Monchegorsk) of Moiseenko et al. (1995) collected in 1993, the presently reported 1994 median values from *C2* for Co, Cu, Ni and Zn are 5 to 10 times higher, those for Mn and Sr are ca. 2 times higher, and those for Al are approximately similar. We attribute the differences mainly to the closer distance of *C2* to the smelters ( $\leq 5$  km south of Monchegorsk).

Ni (median content) in the *C2* stream is 720 times greater than in 'background' stream water as reported by Hart and Hines (1995), Zn up to 70 times, Cu 40 times, Cd 7 times; surprisingly, Pb in *C2* stream water is about one third of 'background' levels as postulated by Hart and Hines (1995).

Compared with average stream water composition from Finland (in Koljonen, 1992), the stream sampled at *C2* is enriched 743 times in Ni, 14 times in Cu, 7.5 times in Cd, 2.5 times in Zn, and is 43 times depleted in Pb!

However, it may be more meaningful to compare trace element levels in *C2* with a local background area for anthropogenic fallout, for instance *C7*. Comparing median values, Ni is 1115 times higher in *C2* than in *C7*, Cu 303 times, Cd >7.5 times, Zn 6 times, and Pb 1.6 times.

The stream water from *C3* is rather unusual in comparison with the three other Russian catchments in that it has concentration levels for the main anthropogenic metals and sulphur that are similar to the remote streams (i.e. low), yet has remarkably elevated F, Mo, Rb and Sr levels. In the town of Kirovsk there is an apatite open-cut mine and processing plant, and the

local bedrock (alkaline rocks) and overlying glacial deposits are enriched in these elements (e.g. median Sr content in Quaternary deposits is more than 20 times higher at C3 than in any of the other catchments). Therefore, these elements ultimately have a geogenic origin here, though their transfer to the streams is likely to be exacerbated by human activity (open-pit mine, stock piling of ore). Indeed, based on results of snow analysis (Äyräs et al., 1995), we believe that these elements are transferred to the stream water partly directly (ground water, soil water) and partly indirectly (via blowing dust from the mine area, for instance). Interestingly, neither Ca nor P (PO<sub>4</sub>), which are major constituents of apatite, reach particularly elevated concentrations in the stream water here.

The high level of stream water Ba at C4 is attributed to a geogenic source, on the basis of its anomalous enrichment in till and bedrock here relative to the other catchments.

The presence of the highest Ca concentration in the stream water at C5 is related to the presence of minor carbonate bedrock and the dominance of andesitic and basaltic bedrock in the catchment area. This is again reflected in the high Ca content in Quaternary deposits relative to other catchments. The relatively high As concentrations in stream water are also best explained as originating primarily from natural, geogenic sources (the Quaternary deposits in C5 have the highest median As content of all the catchments studied here). The content of As in air (Sivertsen, 1994b), rain (Reimann et al., in prep. a) and snow (Äyräs et al., 1995) are only slightly elevated in the area.

The high levels of stream water Al at C7 and of Fe, Li, Pb, Si, U at C8 are attributed to a geogenic source, as none of these elements is particularly abundant in rain or snow there, and most of them are unusually enriched in Quaternary deposits and/or bedrock. C-horizons from C7 have relatively high total Al content, and the Al is unusually mobile (up to 31% of total Al is aqua regia extractable, up to 1% is ammonium acetate extractable) (Kashulina et al., in prep.).

In summary, on the basis of comparison with element contents in Quaternary deposits, bedrock, A<sub>0</sub> and C soil horizons, rain and snow (Reimann et al., 1995, Äyräs et al., 1995, Reimann et al., in prep. a, b), we interpret the above highest median element contents to *primarily* have either (1) a natural, marine source (Cl and Na at C1), (2) a natural, geogenic source (K at C2; Sr at C3; Ba at C4; As and Ca at C5; Al at C7; Li, Pb, Si and U at C8), or (3) an anthropogenic, industrial source (Ag, B, Cd, Co, Cr, Cu, Ni, Pb, V, S, Sb, SO<sub>4</sub> and Zn at C2). However, many elements have both a natural and an anthropogenic origin in the region as a whole. In general, it may only be possible to determine with certainty the principal source of an element in either strongly polluted or, conversely, clearly pristine areas.

Those elements that are an important part of the emission spectrum from the industrial activities in the area display a characteristic, technogenic signature on the boxplots of Figure 4.1.2 (see for instance Cu, Ni, SO<sub>4</sub>, etc.). The pattern described by the medians (a relatively high value for C1, the highest peak for C2, a trough for C3, then a decrease from relatively high C4 to low C8) can be identified easily, and is different from the geogenic (see Li, Si, etc.) or the marine (Cl, Na) signatures. Again, many elements have signatures that are mixed to some degree.

## CORRELATIONS

Figure 4.1.3 shows selected XY-correlation diagrams. Na vs. Cl indicates that stream water from *C1*, nearest to the sea and with the highest Na and Cl values, *C5*, *C4* and *C6* plot on a trend roughly parallel to, and about 500 µg/l Na above, the sea water dilution line. The additional Na apparently originates from weathering in the bedrock and overburden (e.g. dissolution of Na-plagioclase). *C2* falls on the sea water dilution line, despite its great distance from the sea, and this is attributed to the likely addition of salt (NaCl) in the industrial process at the nearby Monchegorsk smelter (MRCENR (1995) reports Cl emission to air from this smelter amounting to 341 tonnes in 1994, and Pozniakov (1993) notes that sodium chloride discharges to the neighbouring lake, Lake Imandra, are of the order of 17,000 tonnes per annum). The very peculiar lithological substrate at *C3* (alkaline rocks) results in a clear separation of this data group from the others: Na is enriched (ca. 3500 µg/l) to levels similar to the catchment most influenced by sea spray (i.e. *C1*), yet Cl levels remain low (ca. 1000 µg/l). *C7* and *C8* clearly also are influenced by the contribution of Na from silicate weathering, and have low Cl levels.

In terms of Ca and SO<sub>4</sub>, the different catchments form distinct groups. *C5* is most noticeable, with its high Ca and low SO<sub>4</sub> values resulting from dissolution of relatively abundant Ca-bearing minerals (carbonates, plagioclases, etc.). The departure from the sea water dilution trend suggests that neither of the two elements are controlled by sea spray here. *C2*, *C4* and *C1* have the highest SO<sub>4</sub> levels as a result of the massive deposition of airborne sulphur compounds, and intermediate Ca levels resulting probably both from deposition from industrial emissions (carbonates are used as flux material in the smelters) and from leaching of Ca-minerals due to very acid conditions. Industrial processes of ore smelting (*C2*, *C4*) and ore roasting (*C1*) yield different signature in terms of Ca emissions (higher and lower, respectively). *C3* and *C6-8* have all low SO<sub>4</sub> values and relatively low Ca levels, with a trend suggesting some contribution of Ca without contribution of SO<sub>4</sub> (parallel to the trend for *C5*).

Correlation of a heavy metal typical of the emission spectrum, e.g. Ni, with S or Cu yields clear groups for the different catchments, with *C2*, *C1* and *C4* being most influenced by the emissions, *C5* being intermediate, and *C3* and *C6-8* being least affected and showing natural, background variability. For *C2*, very strong correlations ( $r > 0.9$ ) exist between the content in streamwater of Ni on the one hand and As, Cd, Co (not shown) and Cu on the other hand. For the other catchments, this correlations is considerably weaker. Points from *C4* and *C1*, and to some extent from *C5*, commonly are aligned upon the *C2* regression, but at lower concentration levels. This suggests common source(s) and progressive dilution with increasing transport distance. In the case of Cu vs. Ni, the whole data set (eight catchments) clusters close to a line with slope  $\approx 1:1$ . Similar slope is also observed in snow, both for melt water and filter residue (Äyräs et al., 1995).

The Si vs. EC diagram illustrates the relationship between silicate weathering and salinity of stream water. In the background catchments (*C6-8* and *C3*) where weathering dominates, Si reaches the highest levels (up to 8000 µg/l), yet EC remains relatively low (<6 mS/m). In *C1*, *C2* and *C4*, the more polluted watersheds, EC can be quite high (up to 15 mS/m) and Si also relatively high (ca. 5000 µg/l). In *C5*, EC can also be high (up to 13 mS/m), but the low Si content (<3000 µg/l) suggests that silicate framework weathering is not the main cause for this; we have previously shown that Ca from carbonate and calc-silicate dissolution was an important weathering product in *C5*.

## FACTOR ANALYSIS

Factor analysis was performed in order to gain insight into the associations of elements, and, by inference, to put forward hypotheses as to their origin. Principal factor analysis (SMC) with varimax rotation was applied on a selection of log-transformed parameters (Tab. 4.1.3). The obtained factors have the following loadings (elements ranked in order of decreasing importance within each factor): F1 (Ca, K, As, Mg, Ba, Cl), F2 (Li, Fe, Si, U), F3 (Na, Mo, Sr) and F4 (Co, Ni, Cu, Mn, V, Al, S, Zn, B, Rb). Together, the four factors account for 81% of the variance of the total dataset. F1-F3 have negative loadings, F4 has positive loadings. F1 is interpreted to represent a mixed geogenic-marine source, including base cations and As from weathering of unusual rocks and overburden (C5). F2 is a geogenic factor, especially characteristic of the acid (Si) lithology and U content of C8. F3 is another geogenic group representing the alkaline rocks of C3. F4 contains many of the elements known to make up the industrial emission spectrum and therefore is a technogenic factor depicting contamination.

Figure 4.1.4 shows how pairs of these factor covary. F1 vs. F2 nicely separates the trends from the three countries. The Finnish catchments (C6-8) have the lowest F2 values and high F1 values. C8 is even distinguishable from C6-7, having even lower F2 values. The Norwegian catchment (C5) scores the lowest F1 factors, and intermediate F2 factors. The Russian catchments (C1-4) have an intermediate location on the diagram, which is expected from the interpretation of F1 (reflecting especially C5) and F2 (reflecting especially C8). Plotted against the third 'geogenic' factor (F3), F1 affords a clear separation of C3 from C1 and C6. C7 and C8 overlap somewhat, as do C2 and C4 to a greater extent. C5 again is quite distinct in character. The technogenic factor (F4) in combination with F3, for instance, clearly indicates that C2, C1 and C4 are the most affected by airborne contamination. C3, C5 and C6-8 are approximately on the same level of F4-value, with the first one being clearly separated from the others due to its low F3 score.

## CONCLUSIONS

Water from streams draining eight catchments (C1-C8) on the western Kola Peninsula (Russia) and in neighbouring areas of Finland and Norway (see Fig. 4.1.1, Tab. 4.1.1) were collected in 1994 and analysed for major, minor and trace constituents to determine the levels of element content (from heavily contaminated to pristine drainage basins) and their origin. From this study, a number of conclusions can be drawn.

- (1) Regional variation in element content was found to be much greater than temporal variation: most fallout elements show a large variation in concentration (up to three orders of magnitude for medians) from the polluted to the remote streams, with no overlap of data ranges (see Fig. 4.1.2, Tab. 4.1.2). This implies that the extent of the most polluted areas is restricted, but that the perturbation within these is quite profound and lasting.
- (2) Highest median values (see Fig. 4.1.2) were recorded at C1 for Cl and Na, C2 for Ag, B, Cd, Co, Cr, Cu, K, Mg, Mn, Ni, NO<sub>3</sub>, Pb, S, Sb, SO<sub>4</sub>, V and Zn, C3 for F, Mo, Rb and Sr, C4 for Ba, C5 for As and Ca, C7 for Al and C8 for Fe, Li, Pb, Si and U. Highest and lowest median pH values are from C5 and C7, respectively, while highest and lowest median EC values are from C2 and C7, respectively.
- (3) Based upon element contents in Quaternary deposits, bedrock, O- and C-soil horizons, rain and snow in those same catchments, the above highest median element contents in stream water are interpreted to *primarily* have either: (1) a natural, marine source (Cl and Na in C1),



(2) a natural, geogenic source (K in C2; Sr in C3; Ba in C4; As and Ca in C5; Al in C7; Li, Pb, Si and U in C8), or (3) an anthropogenic, industrial source (Ag, B, Cd, Co, Cr, Cu, Ni, Pb, V, S, Sb, SO<sub>4</sub> and Zn in C2). However, many elements have both a natural and an anthropogenic origin in the region as a whole.

(4) Typical technogenic (Cu, Ni, ...), geogenic (Li, Si, ...) and marine (Cl, Na) signatures can be differentiated (see Fig. 4.1.2). Many elements show somewhat mixed signatures.

(5) Strong positive correlations ( $r > 0.9$ ) between Ni and As, Cd, Co, Cu in polluted areas (see Fig. 4.1.3) suggest that these trace metals have a common origin, industrial emissions, and are progressively diluted with increasing transport distance from the sources. Ni and Cu are present in all stream waters in approximately equal amounts. In C5, As has a dominant geogenic source.

(6) High element content in stream waters from the most polluted areas indicates that contaminants are constantly and effectively being washed out of the catchment area in which they initially were deposited. Therefore, stream, lake and, ultimately, ocean waters and sediments can become the final repository of anthropogenic pollutants.

(7) Factor analysis revealed the importance of geogenic-marine sources (three distinct factors, underscoring the complex and substantial influence of substrate on stream water chemistry), and of the technogenic influence (one, but dominant, factor). Data from the three countries and from most catchments form distinct cluster on factor-factor diagrams, as a result of the large range in variation in terms of airborne contamination, geology and maritime influence considered in this comparison of eight catchments. It is interesting to note that even in an area with extreme airborne contamination, the geogenic factors still emerge as subtle and important controls on the chemistry of stream water.

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## TABLES

Table 4.1.1 Overview of the main characteristics of the catchment

Table 4.1.2 Median, minimum and maximum values of stream water data from the eight catchments

Table 4.1.3 Factor loadings obtained by principal factor method after varimax rotation of log-transformed data

## FIGURES

Figure 4.1.1. Location of study area for regional geochemical mapping (frame) and of the eight catchments discussed herein (1: Zapoljarniy, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, and 8: Pallas), and selected townships/industrial centres (K: Kirovsk, Ki: Kirkenes, M: Monchegorsk, Mu: Murmansk, N: Nikel, Z: Zapoljarniy).

Figure 4.1.2. Boxplot diagrams showing the variation in pH, EC (mS/m) and 14 selected elements ( $\mu\text{g/l}$ ) in stream water from the eight catchments (C1 to C8 on X-axis). Please note that the concentrations are log-transformed (base 10) to accommodate the large concentration differences reported. The values on the right are the actual concentrations in  $\mu\text{g/l}$ . For an explanation of the boxplot representation, see text.

Figure 4.1.3. Diagrams showing the correlations between Na and Cl, Ca and  $\text{SO}_4$ , S and Ni, Cu and Ni, and Si and EC in all catchments (all concentrations in  $\mu\text{g/l}$ , EC in mS/m). The sea water dilution line (SW) is shown for the first two diagrams.

Figure 4.1.4. Results of factor analysis of the stream water data: F1 vs. F2 and F3, F4 vs. F3, for all catchments.

Table 4.1.1 Overview of the main characteristics of the catchments

No.	Name	Coordinates of catchment outlet	Size (km <sup>2</sup> )	Elevation (m a.s.l.)	Annual precip. (mm)*	Vegetation	Bedrock	Surface cover, peculiarities
RUSSIA								
C1	Zapoljarniy	69°27'01"N 31°03'49"E	19.02	25-373	454	birch forest tundra	gneiss	till, fluvio-glacial, outcrop
C2	Monchegorsk	67°50'30"N 32°54'48"E	22.38	128-507	391	technogenic desert, birch shrubs	dacite & andesite & tuffs, gabbro/norite	till, prone to erosion
C3	Kirovsk	67°32'50"N 33°48'55"E	20.01	240-1075	502	spruce forest, mountain tundra birch forest	nephelinite	till, diluvial/eluvial
C4	Kurka	67°41'25"N 32°50'14"E	20.49	152-466	502	north taiga spruce forest, birch; incipient deterioration	amphibolite, gneiss	till, fluvio-glacial
NORWAY								
C5	Skjellbekken	69°21'25"N 29°27'25"E	34.56	80-297	422	north taiga pine forest, birch	andesite, basalt & tuffs, 'black shale'	till, esker
FINLAND								
C6	Kirakka	69°35'12"N 28°51'46"E	11.86	110-200	386	north taiga pine forest	granite	outcrop, till, moraine ridge
C7	Naruska	67°21'44"N 29°22'05"E	20.16	263-490	513	north taiga spruce forest	gneiss	till, peat, outcrop
C8	Pallas	68°09'14"N 23°52'50"E	24.42	303-500	405	north taiga spruce forest	quartzite	till, peat

\* from the closest meteorological station (data from 1994)

Table 4.1.2 Median, minimum and maximum values of stream water data from the eight catchments

	Detec. limit	Catch. 1 (n=6) Apr - Sep 1994			Catch. 2 (n=32) Mar - Nov 1994			Catch. 3 (n=7) Mar - Oct 1994			Catch. 4 (n=8) Mar - Nov 1994			Catch. 5 (n=34) Mar - Nov 1994			Catch. 6 (n=5) Apr - Aug 1994			Catch. 7 (n=8) Mar - Oct 1994			Catch. 8 (n=30) Mar - Oct 1994		
		Med	Min	Max	Med	Min	Max	Med	Min	Max	Med	Min	Max	Med	Min	Max	Med	Min	Max	Med	Min	Max	Med	Min	Max
Ag	0.01	<0.01	<0.01	0.03	0.02	<0.01	0.08	<0.01	<0.01	<0.01	<0.01	<0.01	0.03	<0.01	<0.01	0.03	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.03
Al	0.1	14.75	7.92	74.7	31.1	6.02	895	7.85	5.66	19	27.25	9.76	84.7	7.6	1.9	24.5	22.1	15.5	52.5	39.9	10.9	83.5	18	7.84	97.9
As	0.05	0.18	0.15	0.32	0.43	0.23	2.39	0.17	0.1	0.28	0.21	0.12	0.35	0.58	0.35	0.83	0.06	0.06	0.09	0.09	0.07	0.15	0.11	0.06	0.26
B	0.5	2.32	1.82	3.16	2.62	0.6	5.27	<0.5	<0.5	1.08	1.16	0.96	4.14	1.125	0.52	2.24	<0.5	<0.5	1.88	<0.5	<0.5	0.92	<0.5	<0.5	1.78
Ba	0.04	4.96	4.83	6.06	18.9	13.9	26.9	3.75	2.91	3.83	19.85	11.3	27.1	11.3	7.41	19.7	1.35	1.19	2.17	2.785	1.79	3.06	15.2	8.12	19.2
Be	0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.22	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.14
Bi	0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	0.05	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03
Br	200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200	<200
Ca	10	3995	2200	6100	12000	4900	14600	2620	1330	4100	7000	2820	9800	16350	8250	19400	1140	1090	1800	2400	1260	3300	4200	1060	6600
Cd	0.02	<0.02	<0.02	0.03	0.15	0.05	2.51	<0.02	<0.02	<0.02	<0.02	<0.02	0.22	<0.02	<0.02	0.03	<0.02	<0.02	0.03	<0.02	<0.02	0.08	<0.02	<0.02	0.03
Cl	100	5300	4900	6500	4000	1880	5500	900	800	1100	2450	1300	3400	2500	1700	3300	1900	1600	2300	600	400	800	600	300	1200
Co	0.02	0.06	<0.02	0.34	6.81	1.57	83.8	<0.02	<0.02	<0.02	0.065	0.05	0.32	<0.02	<0.02	0.03	<0.02	<0.02	0.04	0.045	0.03	0.06	0.05	0.03	0.17
Cr	0.2	<0.2	<0.2	0.37	0.5	<0.2	1.98	<0.2	<0.2	<0.2	0.355	<0.2	0.74	<0.2	<0.2	0.21	<0.2	<0.2	<0.2	0.395	0.34	0.59	<0.2	<0.2	0.49
Cu	0.04	4.26	3.63	6.08	41	16.5	1100	0.16	0.08	0.6	3.69	2.41	12.1	0.945	0.25	2.24	0.29	0.27	0.37	0.135	<0.04	0.22	0.18	<0.04	0.71
F	50	<50	<50	<50	<50	<50	80	150	80	180	37.5	<50	60	<50	<50	50	125	110	140	<50	<50	<50	<50	<50	50
Fe	10	25	10	150	40	10	240	<10	<10	<10	55	30	290	40	10	130	10	<10	20	120	70	200	280	140	1240
K	10	680	630	920	1080	730	2600	710	570	900	1010	620	1890	910	530	1180	140	120	210	405	240	740	360	120	1330
Li	0.1	0.34	0.24	0.41	0.56	0.15	1.99	0.14	<0.1	0.28	0.605	0.38	1.21	0.305	0.11	1.04	0.29	0.15	0.47	0.895	0.52	1.43	1.79	0.68	3
Mg	10	1500	1200	2000	4150	1640	4940	300	170	500	1590	690	2100	1175	600	1500	340	300	500	595	330	800	950	320	1400
Mn	0.02	1.285	0.45	14.3	21.8	9.91	166	0.1	0.03	1.15	14.55	7.73	41.8	2.595	1.05	5.95	0.75	0.52	4.12	2.24	1.44	3.77	7.94	3.57	37.8
Mo	0.03	0.39	0.08	0.62	0.46	0.15	0.79	1.29	0.72	1.88	0.215	0.05	0.54	0.165	0.09	0.21	0.55	0.28	0.66	0.08	0.04	0.13	0.31	0.05	0.54
Na	100	3850	3300	4700	2500	1000	3700	3500	2540	3700	2200	1110	3800	2100	1300	2900	1620	1480	2100	1750	1090	2400	2000	830	2700
Ni	0.06	27	24.9	37.4	223	96	1100	0.09	<0.06	0.37	7.27	4.94	13.9	0.915	0.41	1.59	0.29	0.2	0.41	0.2	<0.06	0.38	0.26	<0.06	0.47
NO3	200	<200	<200	400	2430	1400	5500	1100	900	1800	<200	<200	200	<200	<200	1470	<200	<200	<200	<200	<200	<200	<200	<200	300
P	50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50
Pb	0.03	<0.03	<0.03	0.06	0.07	<0.03	0.57	<0.03	<0.03	0.14	0.04	<0.03	0.25	0.05	<0.03	0.19	0.04	<0.03	0.04	0.045	<0.03	0.14	0.06	<0.03	0.34
PO4	20	<20	<20	210	<20	<20	20	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20	20	<20	<20	30	<20	<20	20
Rb	0.01	1.105	1.04	1.29	0.96	0.47	2.15	1.45	1.12	1.68	0.605	0.52	1.35	0.66	0.45	0.85	0.46	0.42	0.61	0.325	0.24	0.53	0.54	0.37	3.53
S	50	3230	2250	3630	9230	5600	12500	920	790	1420	3255	2590	3890	2175	1450	2980	640	550	770	545	410	660	370	300	730
Sb	0.025	<0.025	<0.025	<0.025	0.06	<0.025	0.15	<0.025	<0.025	0.03	<0.025	<0.025	0.03	<0.025	<0.025	0.03	<0.025	<0.025	<0.025	<0.025	<0.025	0.04	<0.025	<0.025	0.13
Se	0.5	<0.5	<0.5	<0.5	<0.5	<0.5	1.4	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	0.6	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Si	100	2710	2480	4600	4200	1600	5500	3000	2090	3700	4550	2160	6500	1900	1170	3400	1100	900	1500	4250	2890	6800	5090	2390	8500
SO4	200	10000	6800	11000	27500	14600	39000	2800	2200	4300	10100	7800	12000	6600	4900	9100	1800	1600	2300	1400	600	1900	900	600	1900
Sr	0.1	17.9	10.6	23.8	38	20.2	48.5	128	111	141	22.65	11.2	37.1	31.55	14.1	37.4	6.17	5.27	9.34	16	8.33	22.1	13.8	4.44	20.5
Th	0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	0.02	<0.02	<0.02	<0.02	<0.02	<0.02	0.02
Ti	5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Tl	0.02	<0.02	<0.02	<0.02	<0.02	<0.02	0.07	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
U	0.01	0.06	0.02	0.07	0.02	<0.01	0.05	<0.01	<0.01	0.01	0.06	0.03	0.15	0.06	0.02	0.07	0.43	0.31	0.61	0.03	0.01	0.05	2.61	1.69	4.99
V	0.02	0.195	0.15	0.25	0.64	0.47	1.86	0.19	0.12	0.56	0.23	0.14	0.36	0.205	0.11	0.32	0.07	0.05	0.08	0.315	0.22	0.37	0.13	0.07	0.24
Zn	0.1	29.85	2.93	36.6	38.1	6.45	3820	1.46	0.93	7.27	32.05	3.68	1280	1.55	0.42	15.9	9.27	1	159	2.625	0.81	6.89	14.6	0.76	2390
pH		6.82	6.03	7.12	7	5.78	7.6	6.9	6.63	7.61	6.8	6.4	7.3	7.2	6.65	7.6	6.45	4.58	7.81	6.69	6.4	7.7	6.8	4.4	7.9
EC		5.95	4.5	8.1	12.1	6	14.8	3.5	2.4	4.8	6.85	3.2	8.7	10.5	5.5	13.3	1.9	1.8	2.6	2.4	1.5	3.5	3.8	1.2	5.7

Table 4.1.3 Factor loadings obtained by principal factor method after varimax rotation of log-transformed data

	F1	F2	F3	F4
Al	0.424	-0.139	0.425	<b>0.706</b>
As	<b>-0.728</b>	0.381	0.218	0.338
B	-0.549	0.172	-0.159	<b>0.573</b>
Ba	<b>-0.646</b>	-0.502	-0.016	0.375
Ca	<b>-0.979</b>	0.040	-0.130	0.007
Cl	<b>-0.633</b>	0.427	-0.241	0.425
Co	-0.160	-0.133	-0.120	<b>0.939</b>
Cu	-0.431	0.197	-0.078	<b>0.861</b>
Fe	0.087	<b>-0.792</b>	0.403	-0.029
K	<b>-0.774</b>	0.135	-0.384	0.294
Li	0.087	<b>-0.870</b>	0.060	0.103
Mg	<b>-0.665</b>	-0.212	-0.288	0.593
Mn	-0.282	-0.441	0.234	<b>0.754</b>
Mo	0.033	0.022	<b>-0.829</b>	0.226
Na	-0.246	0.081	<b>-0.870</b>	-0.046
Ni	-0.414	0.105	-0.132	<b>0.865</b>
Rb	-0.171	0.270	-0.248	<b>0.523</b>
S	-0.621	0.337	-0.220	<b>0.640</b>
Si	0.014	<b>-0.743</b>	-0.521	0.173
Sr	-0.471	0.362	<b>-0.660</b>	0.101
U	0.340	<b>-0.716</b>	0.216	-0.369
V	-0.409	0.135	-0.106	<b>0.740</b>
Zn	0.078	-0.338	-0.092	<b>0.590</b>
Variance	0.230	0.168	0.137	<b>0.281</b>

*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
Catchment locations

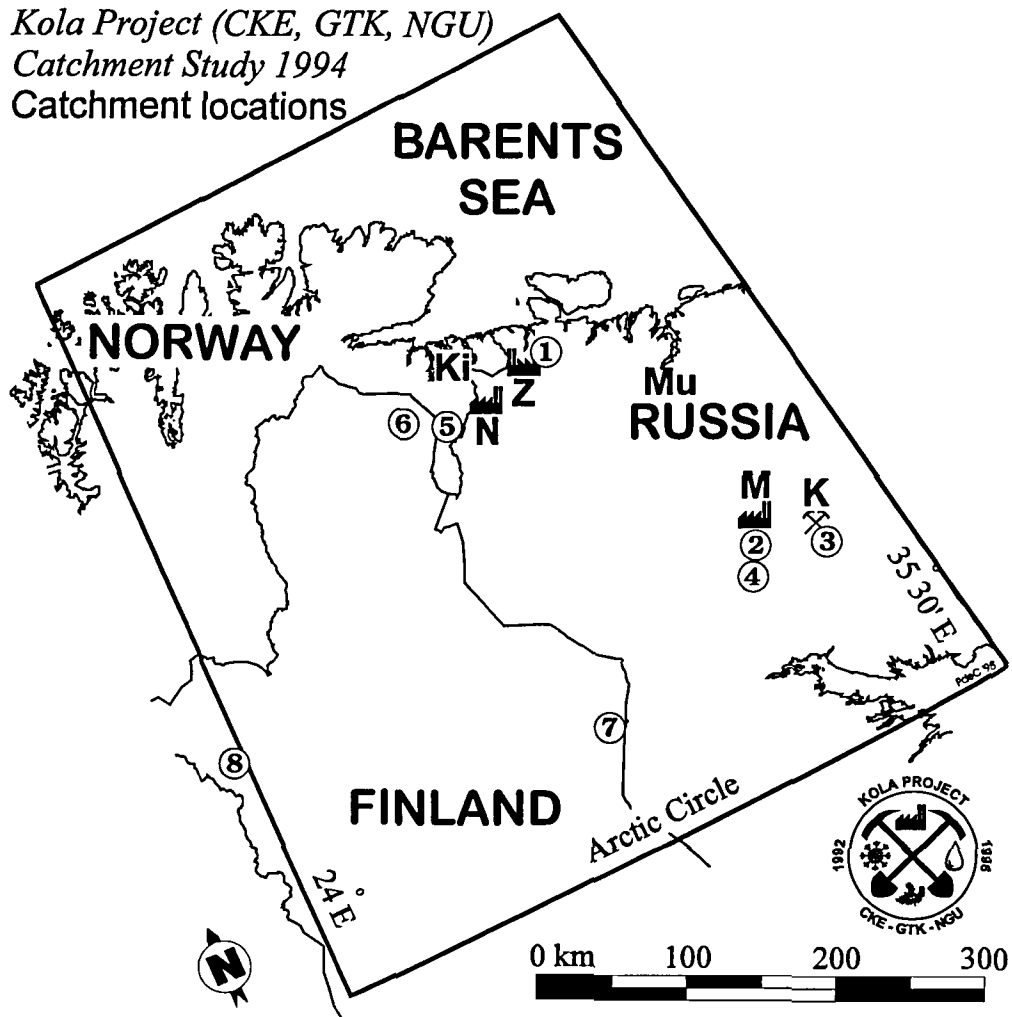
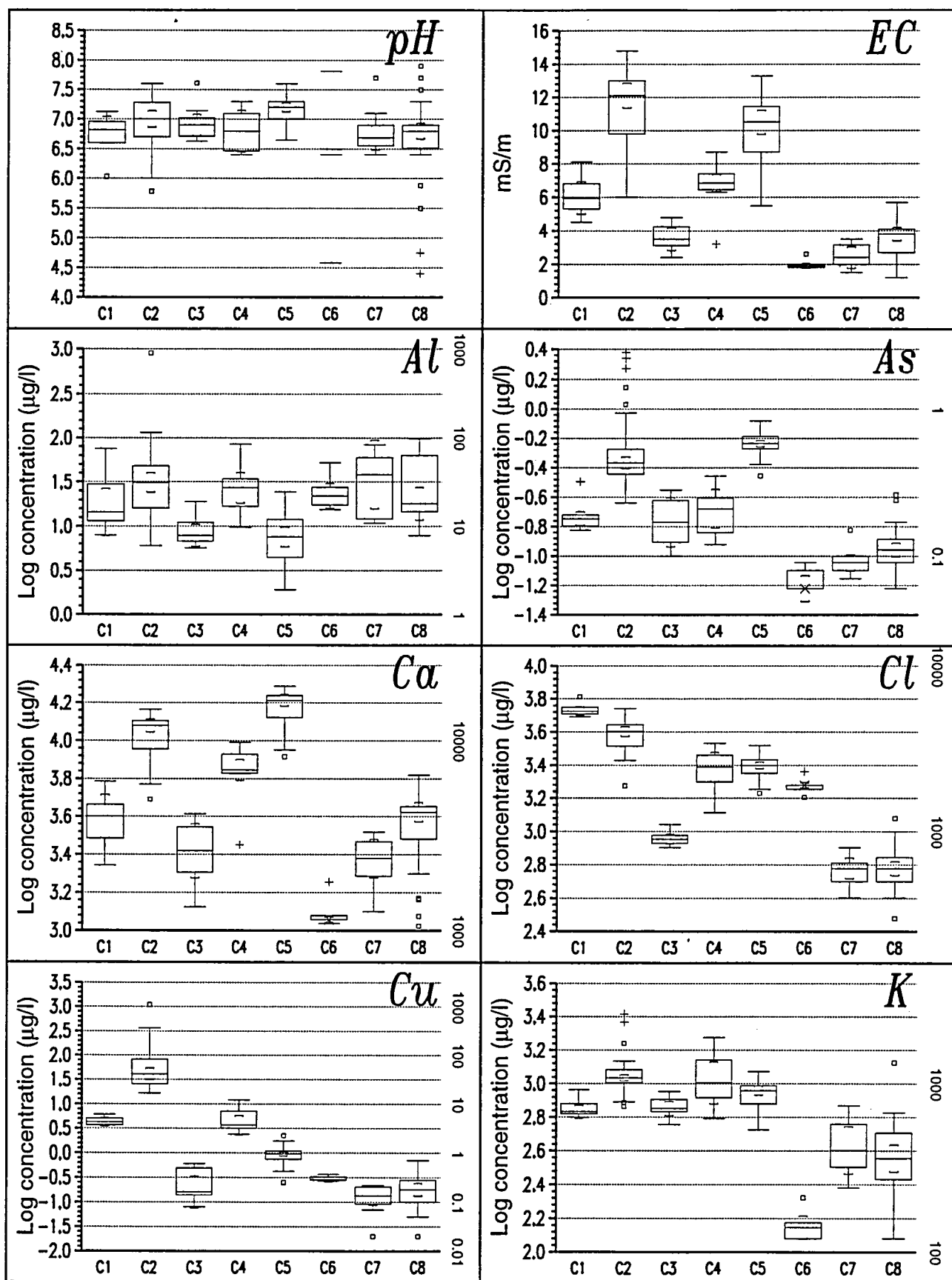


Figure 4.1.1. Location of study area for regional geochemical mapping (frame) and of the eight catchments discussed herein (1: Zapoljarniy, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, and 8: Pallas), and selected townships/industrial centres (K: Kirovsk, Ki: Kirkenes, M: Monchegorsk, Mu: Murmansk, N: Nickel, Z: Zapoljarniy).



Figure 4.1.2. Boxplot diagrams showing the variation in pH, EC (mS/m) and 14 selected elements ( $\mu\text{g/l}$ ) in stream water from the eight catchments (C1 to C8 on X-axis). Please note that the concentrations are log-transformed (base 10) to accommodate the large concentration differences reported. The values on the right are the actual concentrations in  $\mu\text{g/l}$ . For an explanation of the boxplot representation, see text.



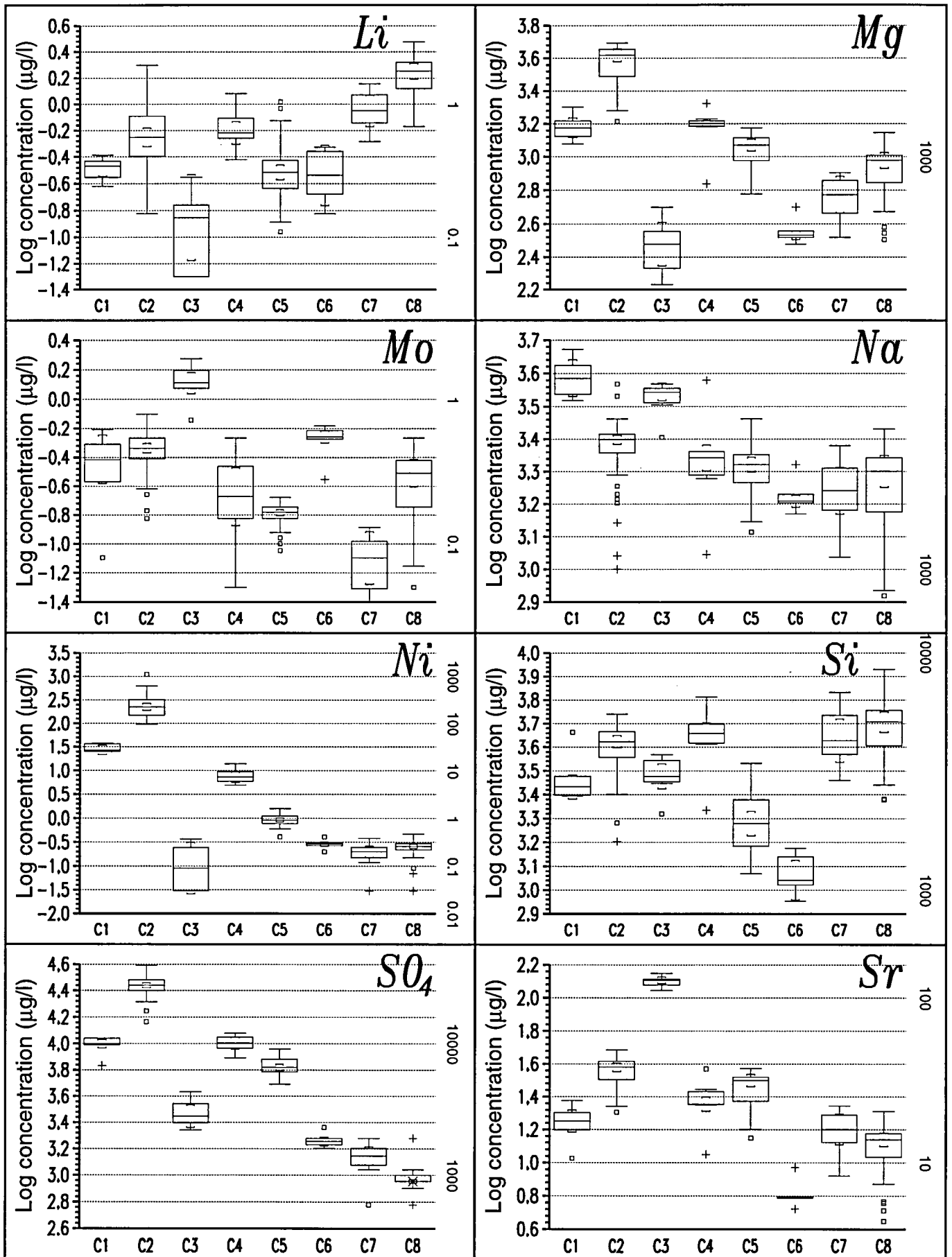


Figure 4.1.3. Diagrams showing the correlations between Na and Cl, Ca and SO<sub>4</sub>, S and Ni, Cu and Ni, and Si and EC in all catchments (all concentrations in µg/l, EC in mS/m). The sea water dilution line (SW) is shown for the first two diagrams.

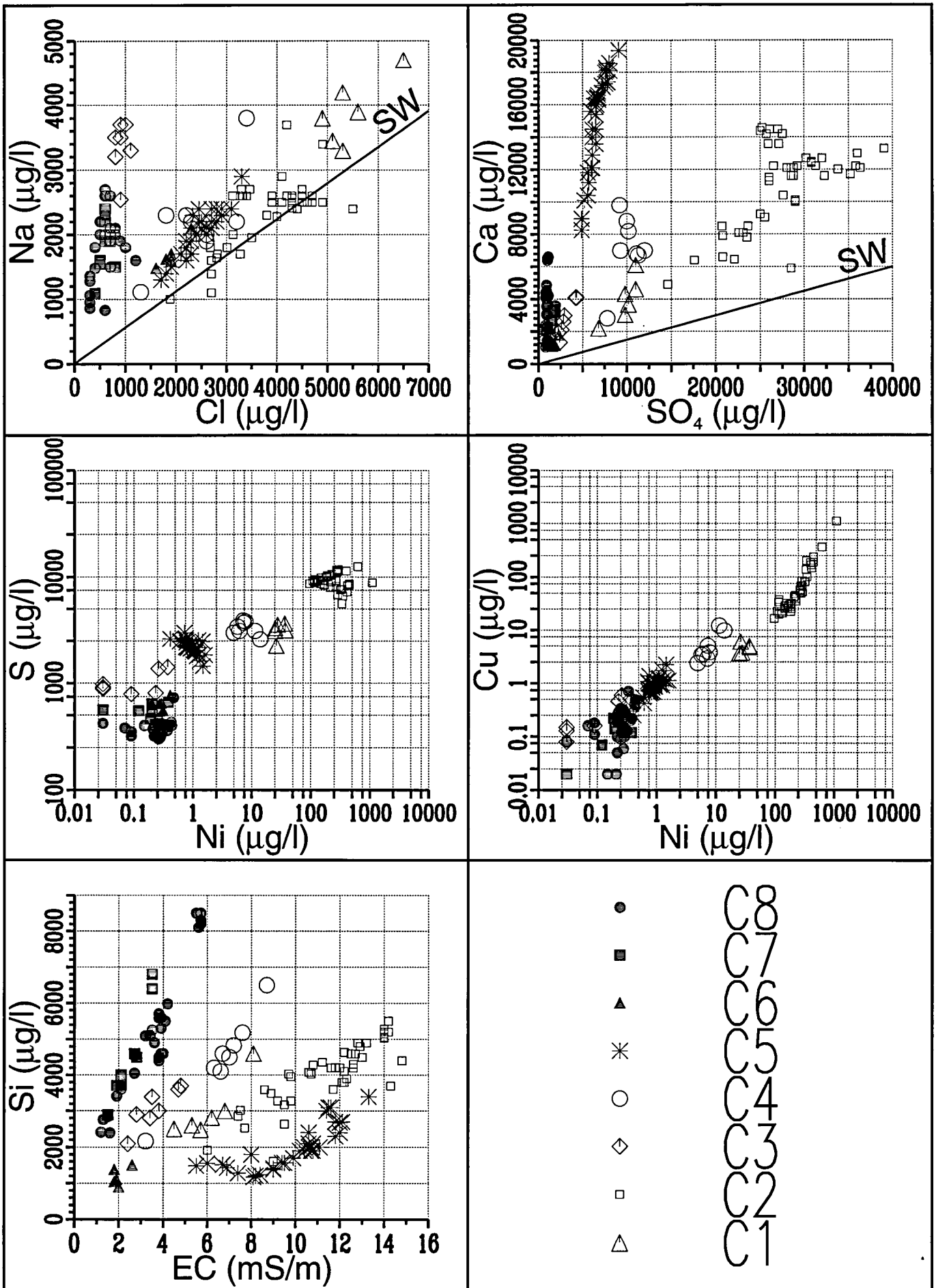
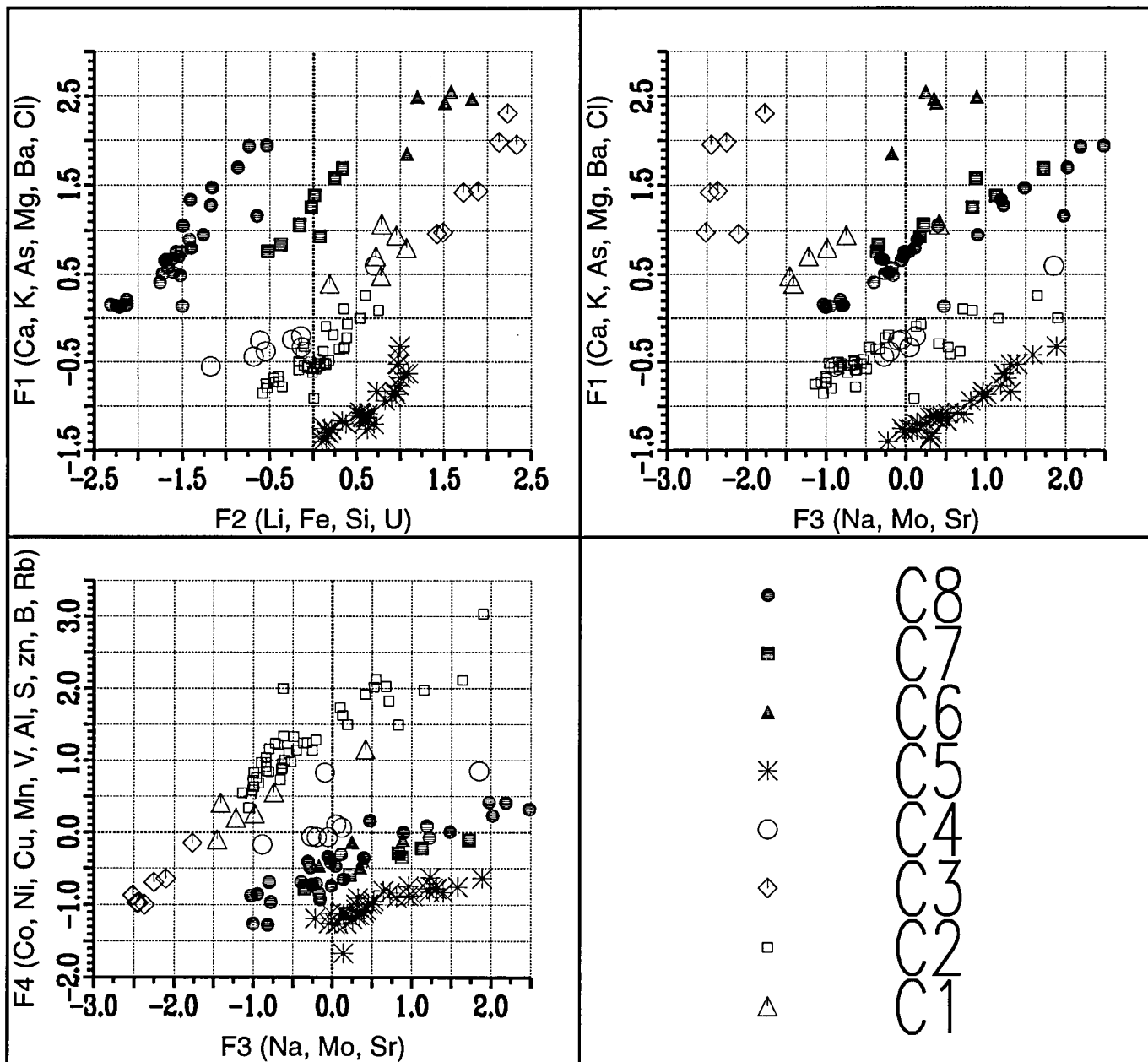


Figure 4.1.4. Results of factor analysis of the stream water data: F1 vs. F2 and F3, F4 vs. F3, for all catchments.



## 4.2 Stream water geochemistry from selected catchments on the Kola Peninsula (NW Russia) and in neighbouring areas of Finland and Norway: 2. Time-series

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### ABSTRACT

The pH, electrical conductance (EC) and concentration of six anions and 33 cations were measured weekly for 8-9 months in 1994 in stream waters from three arctic catchments on and around the Kola Peninsula (Russia, Finland and Norway). The catchments are located at various distances from the major industrial centres of Monchegorsk, Nickel and Zapoljarniy (Russia), at various distances from the sea, and within different geological provinces. Flood caused by melting of the snow cover in the springtime is the most outstanding hydrologic event in these watersheds, and profoundly affects stream chemistry. In the catchment (C2) most contaminated by industrial emissions of SO<sub>2</sub> and trace elements, the response to this event includes a sharp decrease in pH, a marked drop in EC and Ca, and distinct peaks in Ni, Cu, As, S, Al, Rb, Cl and Cu:Ni, for instance. Some elements (Cl, S) are mobilised out of the snowpack before its complete thawing. The substrate (soil, overburden and bedrock) of a catchment controls to a large extent its ability to buffer acid inputs.

### INTRODUCTION

The western half of the Kola Peninsula (NW Russia) is a heavily industrialised area, and has common borders with northernmost Finland and Norway. Among other industries, nickel-copper ore smelters are found at Monchegorsk and Nickel, and an ore roasting plant at Zapoljarniy (Fig. 4.2.1). These industrial activities have released large amounts of various pollutants (SO<sub>2</sub>, NO<sub>x</sub>, CO, Ni, Cu, Co, Mn, As, Cr, Zn, etc.) (Baklanov et al., 1994) into the atmosphere during the last few decades, leading to locally severe environmental degradation (e.g. Kozlov et al., 1993). In 1994, the year relevant for the present investigation, ca. 98,000 tonnes SO<sub>2</sub> were emitted from the Monchegorsk smelters, ca. 129,000 tonnes from Nickel, and ca. 69,000 tonnes from Zapoljarniy; Cu and Ni emissions were of the order of 934 and 1619 tonnes, respectively, from Monchegorsk (or a value of US \$ 13 million for Ni alone!), and 82 and 136 tonnes from Nickel, and 81 and 161 tonnes from Zapoljarniy (MRCENR, 1995).

The Monchegorsk smelter complex started production in 1938 using only local, low-sulphur (ca. 6.5% S) ore, and from 1968 onward, sulphur-rich (up to 30% S) ore from Norilsk has been smelted there also (see Alexeyev (1993) and Pozniakov (1993)). In a cumulative perspective, historical emissions to air from Monchegorsk amount to a staggering 6.5 million tonnes of SO<sub>2</sub> from 1960 to 1994 (data collated from Alexeyev (1993), Pozniakov (1993), Baklanov et al. (1994), and MRCENR (1995)). Emissions have varied over the production years and, in addition, available estimates differ somewhat (compare data in Alexeyev (1993), Pozniakov (1993), Sivertsen (1994), Baklanov et al. (1994), Jaffe et al. (1995) and the official MRCENR (1995) release). In any case, atmospheric pollution by industry in the area is one of the worst in the world. It is not surprising, therefore, that trace metal and sulphur contamination of ecosystems in arctic western Russia and neighbouring countries is a problem currently receiving much attention. This is particularly true when it comes to surface waters

(e.g. Lahermo et al., 1994, Moiseenko et al., 1995, Rodushkin et al., 1995), which are used as the main drinking water source in the region.

Following the completion of a pilot project (1992-1993) focusing on environmental pollution around Nikel (Niskavaara et al., 1996, Väisänen et al., in prep.), the Central Kola Expedition (CKE) and the Geological Surveys of Finland (GTK) and Norway (NGU) are carrying out (1994-1996) a major, co-operative regional geochemical mapping project of a 188,000 km<sup>2</sup> area north of the Arctic Circle on the Kola Peninsula (NW Russia) and in neighbouring areas of Finland and Norway (Fig. 4.2.1). More information about the «Kola Ecogeochemistry Project» is posted on the world wide web (<http://www.ngu.no/Kola>). The aims of the main project are: (1) to establish regional geochemical maps of the distribution of about 40 elements and radionuclides in an ecologically vulnerable area, (2) to build up a soil sample data bank for future study, and (3) to assess the environmental impact of the nickel smelting and other heavy industry in the survey area. Here and in the companion paper (Caritat et al., in prep. a), part of the results of a detailed study and monitoring of eight different areas (catchments) within the regional geochemical mapping area is presented. The catchment study was undertaken to investigate in greater detail the source, residence and fate of elements cycling through catchments that differ greatly in degree of contamination, in proximity to the sea, and nature of underlying bedrock. In this paper, we focus on the time-dependent behaviour of stream water chemistry in three monitored catchments in arctic Europe (see below), paying particular attention to the relationship between contamination, buffering capacity and stream water response.

## STUDY SITES AND SAMPLING

As part of the main geochemical project, element content in various sampling media (rain water, snow, moss, organic stream sediments, stream water, ground water, overbank sediments, different soil horizons, Quaternary deposits and bedrock) was determined and/or monitored during 1994 in eight catchments (watersheds or drainage basins) located at various distances from the smelters (5-300 km), and in different geological settings. Four of these catchments are situated in Russia (*C1*: Zapoljarniy, *C2*: Monchegorsk, *C3*: Kirovsk, *C4*: Kurka), one in Norway (*C5*: Skjellbekken) and three in Finland (*C6*: Kirakka, *C7*: Naruska, *C8*: Pallas) (Fig. 4.2.1). An overview of the major characteristics of the catchments is given in Table 4.2.1. In Russia, prevalent wind directions are from the north and south (Mäkinen, 1994), spreading most contaminated air primarily in these directions.

The stream water samples discussed here were collected weekly near the outlet of each watershed in *C2*, *C5* and *C8* between March and November 1994, and water levels were reported. The main hydrological event of the year in these arctic catchments is the flooding induced by snow melt each spring. In 1994, snowmelt flooding commenced in late April in *C2*, early May in *C5* and mid-May in *C8*, and lasted approximately 2-3 weeks. Snowmelt events can lead to acid and trace metal surges in streams, as elements deposited over the watershed area and stored (immobilised) in the snowpack during winter suddenly are released (for a discussion of snowpack chemistry in the same catchments, see Äyräs et al., (1995) and Äyräs et al., (in prep.)). The topsoil layer having not completely thawed by then and thus being partly impervious, melt water often runs off directly to the stream, carrying along its load of soluble and particular contaminants (though some interaction with topsoil does occur). The snowmelt event can lead to an increase in element concentration in stream water, or to a decrease by dilution, depending on the concentrations in snowmelt water relative to baseflow water. A second flooding event related to summer rainfall took place between late July and mid-August

primarily in C2. High water levels were again reported in autumn, toward the end of the monitoring period (October-November).

In terms of location relative to major element sources other than geogenic, the three monitored catchments represent:

(1) proximity to industry: C2 is 5-10 km SSW from the major Ni-Cu ore smelting centre and point-source emitter of S and trace metals of Monchegorsk, and is remoteness from the sea,

(2) proximity to sea and intermediate distance to industry: C5 is 65 km S (mostly upwind) of the Barents Sea, 30 km WSW (mostly off wind) from the Nickel smelter, 48 km SSW from the Kirkenes iron open-cut mine and mill (average yearly emission: 800 tonnes SO<sub>2</sub> in 1985-1990, Sivertsen et al., 1993), and 52 km WSW from the Zapoljarniy Ni-Cu ore roasting and open-cut mine, and

(3) background: C8 has a remote location both in terms of anthropogenic emissions and marine influence.

Discussion about comparison of element content in stream water from the eight catchments, and about element sources is given by Caritat et al. (in prep. a).

## METHODS

Two subsamples of stream water were taken in the field at each site, one (500 ml) unacidified and unfiltered, the other (60 ml) filtered (0.45 µm) and acidified with suprapure nitric acid. At the (accredited) GTK laboratory, the unacidified subsamples were used for the determination of pH, electrical conductance (EC), five anions (Br, Cl, F, NO<sub>3</sub> and SO<sub>4</sub>) by ion chromatography (IC) and PO<sub>4</sub> by spectrophotometry. The acidified subsamples were used for the determination of six major cations (Ca, Mg, Na, P, S and Si) by inductively coupled plasma-atomic emission spectrometry (ICP-AES) and of 27 trace cations (Ag, Al, As, B, Ba, Be, Bi, Cd, Co, Cr, Cu, Fe, K, Li, Mn, Mo, Ni, Pb, Rb, Sb, Se, Sr, Th, Tl, U, V and Zn) by inductively coupled plasma-mass spectrometry (ICP-MS). In addition, the sulphur isotopic composition of stream water sulphate was determined once in each catchment (Caritat et al., in prep. b). For more information about the methods, accreditation, detection limits, accuracy and precision, see Äyräs and Reimann (1995), Niskavaara (1995) and Caritat et al. (in prep. a).

## RESULTS

Results from all the eight catchments were summarised by Caritat et al. (in prep. a), and are illustrated here in the form of time-series for a selection of 'raw' parameters/elements from C2, C5 and C8 in Figures 4.2.2-4.2.4. The parameters pH and EC are fundamental water properties depicting the dramatic effect of springtime snow melt on stream water chemistry, whilst the selected elements are meant to represent mostly anthropogenic emissions (Cu, Ni, As, S, but also in part Al and Cl), geogenic input (Al, Rb, but also in part Ca) or sea spray (Cl, but also in part Ca), as interpreted elsewhere (Caritat et al., in prep. a).

## DISCUSSION

### *Ph time-series*

Figure 4.2.2a shows the time-series obtained for pH from the three monitored catchments. All stream waters overall have circum-neutral pH values (medians  $C2 = 7.0$ ,  $C5 = 7.2$  and  $C8 = 6.8$ ), a major deviation from this being a 6 week-long dip down to  $\text{pH} = 4.4$  in  $C8$ . In this latter watershed, pH is above 6.5 until mid-May, and again from early July onward. As snowmelt flooding starts there in mid-May, we can confidently attribute this pH dip to the classical «acid surge» mentioned above. Given that the snowpack at  $C8$  is relatively unaffected by anthropogenic acids (see Äyräs et al., 1995), this highlights the importance of the substrate in buffering even moderately acid melted snow ( $C8$  is on quartzite bedrock with no buffering capacity). Further illustrating this point,  $C2$  receives massive anthropogenic acid and trace element fallout, yet its more mafic (basic) lithology (Tab. 4.2.1) efficiently maintains stream water pH above 5.8, which is also reached at snow melt (late April). After this event, pH increases until July-August ( $\text{pH} \cong 7.5$ ), then decreases to a stable level (ca. 7) during the autumn. Stream water pH in  $C5$  (with mixed mafic, silicic and minor carbonate bedrock) is not negatively affected at all by the flooding event in early May, and increases progressively from early spring to mid-summer ( $\text{pH} \cong 7.5$ ), when it stabilises or decreases slightly (7.2).

### *Electrical conductance time-series*

Figure 4.2.2b shows the time-series obtained for EC from the three monitored catchments. Quite different EC levels typify the catchments (medians  $C2 = 12.2$ ,  $C5 = 10.4$  and  $C8 = 3.8$  mS/m), but all three show the same curve shape: a high plateau in late winter (ground water fed baseflow), abrupt fall synchronous with snow melt (runoff surge causing dilution), and progressive increase in salinity throughout the remainder of the year (ground water contribution to stream flow regains importance, and evapotranspiration reaches a maximum in summer). This progressive EC increase is interrupted by subdued dilution due to the minor summer floods. The fact that the general EC levels in the three streams are so different is caused by: (1) high technogenic deposition contributing major ions (S) and causing plant death, soil erosion and perhaps increased weathering rates in  $C2$ , (2) naturally high weathering rates of mafic and carbonate rocks in  $C2$  and  $C5$ , and (3) naturally low weathering rates of quartzitic rocks (extremely limited base cation release) in  $C8$ .

### *Copper time-series*

Figure 4.2.3a shows the time-series obtained for Cu from the three monitored catchments. Immediately noticeable is the significant difference in Cu content of the three streams, with  $C2$  having Cu levels >1 order of magnitude higher than  $C5$  (median  $C2/\text{median } C5 = 43$ ), which in turn generally has more of this metal than  $C8$  (median  $C5/\text{median } C8 = 5$ ). Next, the presence of a sharp peak for  $C2$  corresponding to the main snowmelt event in the catchment (end of April 1994) is observed. This is a typical example of heavy metal surge (and acid surge, as discussed above). No significant Cu peaks occur in  $C5$  or  $C8$  at snow melt. A much more subdued Cu spike in  $C2$  is associated with the second high water event of July.

### *Nickel time-series*

Figure 4.2.3b shows the time-series obtained for Ni from the three monitored catchments. Here, the difference in general level between  $C2$  and  $C5$  (median  $C2/\text{median } C5 = 237$ ), is even greater than for Cu, but the difference between  $C5$  and  $C8$  (median  $C5/\text{median } C8 = 4$ ) is smaller. The Ni peak in late April in  $C2$  coincides again with snowmelt flooding. A similar, though much more attenuated, Ni hump in early May in  $C5$  can be detected. The second flooding period in  $C2$  (July) is contemporaneous with a small Ni increase in the stream water. For both Cu and Ni,  $C8$  shows some variability throughout the summer months (enhanced at



this low end of the ordinate by the log scale), which could be caused by a combination of real fluctuation and some sampling/analysis noise due to the low metal levels.

#### *Arsenic time-series*

Figure 4.2.3c shows the time-series obtained for As from the three monitored catchments. The general level in C5 (median = 0.6 µg/l) is higher than in C2 (median = 0.4 µg/l), as a result of the anomalous As content in the local bedrock and glacial deposits at C5 (Caritat et al., in prep. a). Both catchments, however, have higher As in the stream water than C8 (median C2/median C8 = 3.9, median C5/median C8 = 5.6). At snow melt, As in C2 has a well defined peak (up to 2.2 µg/l), whereas in C5 it only has a modest spike that is only a local maximum. This different behaviour in these two catchments supports a dominantly technogenic source for As in C2 (As in snow), contrasting with a dominantly geogenic source in C5 (As in ground /baseflow water), (Caritat et al., in prep. a). In July-August, As increases again in C2 and C5 (where it reaches the highest concentration recorded for this catchment).

#### *Sulphur time-series*

Figure 4.2.3d shows the time-series obtained for S from the three monitored catchments. The difference between watershed is large (medians C2 = 9.3, C5 = 2.2 and C8 = 0.4 mg/l) and greater than the time-dependent variations. A clear S peak in C2 due to snow melt is recorded in April, apparently preceding the heavy metal (e.g. Cu, Ni, As) surge by ca. 2 weeks, as caused presumably by the greater mobility of S (as sulphate) in the snowpack. Once S is washed out of the snowpack, further melting leads to S dilution in the stream, giving a marked dip in early May. Another smaller S peak occurred in late July, associated with the second flooding event. In C5, snow melt (early May) coincides with a low S content in stream water, as caused by the thawing of S-poor snow (<0.2 mg/l, see Äyräs et al., 1995). In both C2 and C5, the period May-November is characterised by a more or less regular S increase in stream water. S levels in C8 are relatively stable throughout the period recorded.

#### *Aluminium time-series*

Figure 4.2.4a shows the time-series obtained for (total) Al from the three monitored catchments. C2 and C8 have similar Al levels (medians of 31 and 20 µg/l, respectively), higher than those in C5 (8 µg/l). However, whereas C8 only shows a broad, plateau-like Al increase between April and July, C5 and especially C2 have a sharp peak in early May and late April, respectively, clearly related to the melting of the snow cover. This is followed, in these two watersheds, by a progressive decline in Al content, with some short-lived and less important peaks (notably one related to the July-August high water event). Rodushkin et al. (1995) reported similar results from the Kola Peninsula for the snowmelt month in 1993, and concluded after a speciation study that the Al surge in surface water was due mainly to increasing ionic Al concentration. They also showed that there was a clear correlation between total Al in water and Al concentration in various fish organs (esp. gills, skeleton and kidney). This suggests that monitoring total Al in stream water is valuable from the point of view of ecological and bio-toxicological implications. The low level of Al in stream water at baseflow in C5 compared to C8 illustrates the pH-dependency of Al content in surface water (see Fig. 4.2.2a).

#### *Calcium time-series*

Figure 4.2.4b shows the time-series obtained for Ca from the three monitored catchments. Overall levels and concentrations at baseflow decrease in the order C5, C2, C8 (medians = 16, 12 and 4 mg/l, respectively), reflecting the geological influence, the fact that Ca is not a dominant technogenic contaminant (though some Ca-bearing dust from mining and related

activities is deposited in the proximity of open-cast mines (Reimann et al., 1996)), and perhaps also the effect of sea spray. In all three catchments, snow melt is reflected by an abrupt decline in Ca content from a winter plateau, indicating that the snow contains much less dissolved Ca than the streams at baseflow. During the remainder of the monitoring period, Ca levels progressively increase to regain their winter (pre-snowmelt) maxima. Ca is thus supplied to the streams mainly from ground water, after being mobilised by subsurface water-rock interaction in the aquifers. This seems to be true whether the area contains minor carbonates (C5), calc- and alumino-silicates (C2), or is devoid of significant Ca-bearing minerals (C8). Absolute Ca levels, however, show these lithological differences very well.

#### *Rubidium time-series*

Figure 4.2.4c shows the time-series obtained for Rb from the three monitored catchments. Here, the levels are similar everywhere (medians C2 = 0.96, C5 = 0.66 and C8 = 0.54 µg/l), but the springtime behaviour is unusual and distinctive. C8 clearly displays the highest and sharpest peak (3.5 µg/l) in early May, i.e. slightly preceding spring flood. C2 has a broader, but distinct, peak in mid- to late April (i.e. also 1-2 weeks ahead of the snowmelt flood), and C5 has an even broader and smaller bulge at about the same time. The origin of Rb is most likely to be geogenic (bedrock and glacial overburden). Its low abundance in snow (Äyräs et al., 1995; Äyräs et al., in prep.) and its appearance here as a pulse at the streams just ahead of the snowmelt flooding, rather than as a drop from a winter plateau as for Ca, suggests that it is leached out of the thawing topsoil layer at snow melt. The higher levels in stream water and the sharper peak in Rb during snowmelt at C8 correspond well with the fact that this is the watershed with the poorest acid buffering characteristics.

#### *Chlorine time-series*

Figure 4.2.4d shows the time-series obtained for Cl from the three monitored catchments. Levels of Cl are quite different between the three watershed (medians C2 = 4.3, C5 = 2.6 and C8 = 0.6 mg/l), but not in the order expected from the distance to the sea (C5 < C2 < C8). In C2, a sharp Cl peak occurs in mid-April, i.e. ca. 1-2 weeks ahead of the flood, and is followed by a trough during the flood (dilution). Smaller peaks are visible also for C5 and C8, where they also precede the snowmelt flooding event. This behaviour again suggest that Cl is washed out of the snow before complete thawing occurs. The fact that C2 has greater levels of Cl than the more coastal watershed C5, is a result of industrial Cl emissions from Monchegorsk (340 tonnes Cl emitted in 1994; MRCENR (1995)).

#### *Copper:nickel time-series*

Figure 4.2.5 shows the time-series obtained for Cu:Ni from the three monitored catchments. Levels are quite similar (mostly 0-2), but trends differ. C2 has the lowest Cu:Ni values, which are very stable around 0.2 for most of the year, except for a marked peak (up to 1) at snow melt (late April). This is caused by the melting of snow, which has a median Cu:Ni of 2.5 (snowmelt water) in C2 (Äyräs et al., in prep.) and which flows to the stream without interacting significantly with the soil. Summer precipitation is characterised by Cu:Ni values of 2-8 (Reimann et al., in prep. a), depending on which ore (Pechenga or Norilsk ore) is fed into the smelter at Monchegorsk. In any case, the very much lower Cu:Ni of stream water compared to winter and summer precipitation indicates that Cu is bound much more strongly in the soil than Ni. In fact, a result of the pilot project was that only about 1% of the total Cu present in the O-horizon is easily extractable (ammonium acetate), while that figure is around 10% for Ni (Reimann et al., 1995). In C5, Cu:Ni in stream water varies considerably between 0.5 and 1.5, with no obvious trend; the springtime flood does not appear to affect this ratio here. Even more variability in Cu:Ni occurs in C8 (0-3), and this corresponds well with the

findings that the abundance of these elements is rather limited, and that this ratio is extremely variable both in snowmelt water (Äyräs et al., in prep.) and rain (Reimann et al., in prep. a) in this background catchment.

#### *Factor analysis*

Caritat et al. (in prep. a) carried out a factor analysis of the stream water data. Here, this analysis is briefly summarised for the purpose of discussing the time-series of two selected factors. Elements that did not have important detection limit problems in either of the eight catchments were selected, log-transformed, and submitted to principal factor analysis (SMC) with varimax rotation. Resulting factor loadings are: F1 (Ca, K, As, Mg, Ba, Cl), F2 (Li, Fe, Si, U), F3 (Na, Mo, Sr) and F4 (Co, Ni, Cu, Mn, V, Al, S, Zn, B, Rb), where elements are ranked in order of decreasing importance. These four factors explain 81% of the total variance, and are interpreted to represent a mixed geogenic-marine source typical of C5 (F1), a geogenic factor characteristic of C8 (F2), another geogenic group representing the alkaline rocks of C3 (F3), and a technogenic factor depicting contamination (F4).

#### *Factor 2 time-series*

Figure 4.2.6a shows the time-series obtained for F2 from the three monitored catchments. The negative loadings of this geogenic factor representing acid rock sources separates well C8 from C2 and C5. The effect of springtime flooding is to sharply increase the value of F2 in all three watersheds, i.e. to reduce the relative influence of the natural geogenic source by dilution and/or increase of technogenic factor F4. The following and progressive decrease in F2 during the remainder of the growth season reflects the regained importance of the geogenic influence upon stream water chemistry after the most dramatic hydrologic perturbation of the year (snowmelt flood).

#### *Factor 4 time-series*

Figure 4.2.6b shows the time-series obtained for F4 from the three monitored catchments. This technogenic factor, with positive loadings, shows a much greater value for C2 than for C5 or C8. In addition, an enhanced technogenic signal is very well defined in late April for C2, coincident with snowmelt flooding, suggesting massive transfer of a whole spectrum of major and trace elements to the stream at that time. A broad and low hump in F4 is identifiable for C8, with a maximum in mid-May (springtime flood), and this is attributed to acidification in this catchment (effect of Mn, Al, Zn and Rb in the loading of F4). In contrast, C5 does not seem to incur a significant increase at the time of snowmelt (early May). This suggests that where the acid buffering capacity of the substrate (soil, overburden and bedrock) is very low (as C8), the low pH of the snowmelt water can result in mobilisation of base cations from the topsoil (acidification). In contrast, more substantial deposition in areas subject to moderate airborne contamination (as C5) can have no or little impact on the topsoil or surface water if the substrate has sufficient buffering and binding capacity. This is also illustrated quite dramatically comparing the pH time-series for these two watersheds (see Fig. 4.2.2a). Of course, at some point contamination can exceed even the high buffering capacity of a substrate (as C2).

## CONCLUSIONS

Water from streams draining eight catchments (*C1-C8*) on the western Kola Peninsula (Russia) and in neighbouring areas of Finland and Norway (see Fig. 4.2.1, Tab. 4.2.1) were collected in 1994 and analysed for major, minor and trace constituents to determine the levels of element content (from heavily contaminated to pristine drainage basins), their origin (discussed in Caritat et al., in prep. a), and their time-dependent behaviour. From this study, focusing on the time-trends obtained from three closely monitored streams, a number of conclusions can be drawn.

(1) The major hydrological event of the year in these arctic catchments, namely the flooding caused by the snow cover thawing at springtime, has a major influence upon the chemistry of the streams: overall this perturbation dilutes the baseflow waters considerably (as shown by the EC time-series, Fig. 4.2.2b), but for several elements a short-lived peak in concentration is observed (see below, and Figs. 4.2.2-4.2.6).

(2) The time-dependent behaviour of different stream water parameters/elements is different, and the patterns at the springtime snow melt are diagnostic of processes in the catchment:

- some parameters/elements show a clear, short-lived, more or less sharp peak: this is indicative of a lower value in baseflow (winter) and summer stream water than in snowmelt water, and is typical either of the major contaminants/technogenic parameters (Cu, Ni, As, Cu:Ni, F4) or of acidification products (Al, Rb);
- some parameters/elements show a marked, short-lived trough, or negative peak: this suggests a significant dilution of the otherwise high baseflow and summer levels by the melted snow, and is characteristic again of elements/parameters with a low value in the snow (pH);
- some parameters/elements show a peak just prior to snow melt followed by a trough during snow melt: this indicates that these elements are abundant in the snow (technogenic, geogenic or sea spray origin) and are mobilised out of the snowpack before the latter has completely melted, and this is followed by dilution when the flood occurs (Cl, S);
- some parameters/elements show simply an abrupt drop followed by a slow and progressive recovery: this is symptomatic of parameters/elements that have a high value in the baseflow water and thus likely are controlled by the geogenic environment (EC, Ca).

All of these trends generally are most obvious in the more contaminated areas (e.g. *C2*).

(3) The substrate (soil, overburden, bedrock) of any catchment controls to a large extent how snow melt influences stream chemistry: even very low, background-level deposition of trace elements will show up in the stream if the pH is low as a result of poor buffering (*C8*); conversely, moderately high deposition may not affect stream composition too much in a well buffered catchment (compare the pattern of pH or F4 in *C8* and *C5*, Figs. 4.2.2a or 4.2.6b).

(4) Some elements are flushed to the stream with the snow melt (e.g. Ni, Cu, As), while others precede the flood in the stream (e.g. S, Cl) and thus are selectively mobilised out of the thawing snow. The different behaviour of different elements in a dynamically freezing/thawing environment like a snowpack in the springtime is poorly understood. This, and the interaction of snowmelt water with the partially frozen topsoil are areas that need further investigation in order to gain better understanding of element cycling in catchments.

(5) The regional variation in element content in stream water is much bigger than any local, short-lived fluctuation in time, and is governed by (a) lithology in the catchments, and (b) local anthropogenic emission sources.

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## TABELS

Table 4.2.1 Overview of the main characteristics of the catchments

## FIGURES

Figure 4.2.1. Location of study area for regional geochemical mapping (frame) and of the eight catchments discussed herein (1: Zapoljarniy, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, and 8: Pallas), and selected townships/industrial centres (K: Kirovsk, Ki: Kirkenes, M: Monchegorsk, Mu: Murmansk, N: Nickel, Z: Zapoljarniy).

Figure 4.2.2. Time-series for (a) pH, and (b) electrical conductance in the three catchments (C2, C5 and C8) monitored in 1994.

Figure 4.2.3. Time-series for (a) Cu (log scale), (b) Ni (log scale), (c) As, and (d) S in the three catchments (C2, C5 and C8) monitored in 1994.

Figure 4.2.4. Time-series for (a) Al (log scale), (b) Ca, (c) Rb, and (d) Cl in the three catchments (C2, C5 and C8) monitored in 1994.

Figure 4.2.5. Time-series for the Cu:Ni ratio in the three catchments (C2, C5 and C8) monitored in 1994.

Figure 4.2.6. Time-series for factor-analysis factor (a) F2, and (b) F4 in the three catchments (C2, C5 and C8) monitored in 1994.



Table 4.2.1 Overview of the main characteristics of the catchments

No.	Name	Coordinates of catchment outlet	Size (km <sup>2</sup> )	Elevation (m a.s.l.)	Annual precip. (mm)*	Vegetation	Bedrock	Surface cover, peculiarities
RUSSIA								
C1	Zapoljarniy	69°27'01"N 31°03'49"E	19.02	25-373	454	birch forest tundra	gneiss	till, fluvioglacial, outcrop
C2	Monchegorsk	67°50'30"N 32°54'48"E	22.38	128-507	391	technogenic desert, birch shrubs	dacite & andesite & tuffs, gabbro/norite	till, prone to erosion
C3	Kirovsk	67°32'50"N 33°48'55"E	20.01	240-1075	502	spruce forest, mountain tundra birch forest	nephelinite	till, diluvial/eluvial
C4	Kurka	67°41'25"N 32°50'14"E	20.49	152-466	502	north taiga spruce forest, birch; incipient deterioration	amphibolite, gneiss	till, fluvio-glacial
NORWAY								
C5	Skjellbekken	69°21'25"N 29°27'25"E	34.56	80-297	422	north taiga pine forest, birch	andesite, basalt & tuffs, 'black shale'	till, esker
FINLAND								
C6	Kirakka	69°35'12"N 28°51'46"E	11.86	110-200	386	north taiga pine forest	granite	outcrop, till, moraine ridge
C7	Naruska	67°21'44"N 29°22'05"E	20.16	263-490	513	north taiga spruce forest	gneiss	till, peat, outcrop
C8	Pallas	68°09'14"N 23°52'50"E	24.42	303-500	405	north taiga spruce forest	quartzite	till, peat

\* from the closest meteorological station (data from 1994)

Kola Project (CKE, GTK, NGU)  
 Catchment Study 1994  
 Catchment locations

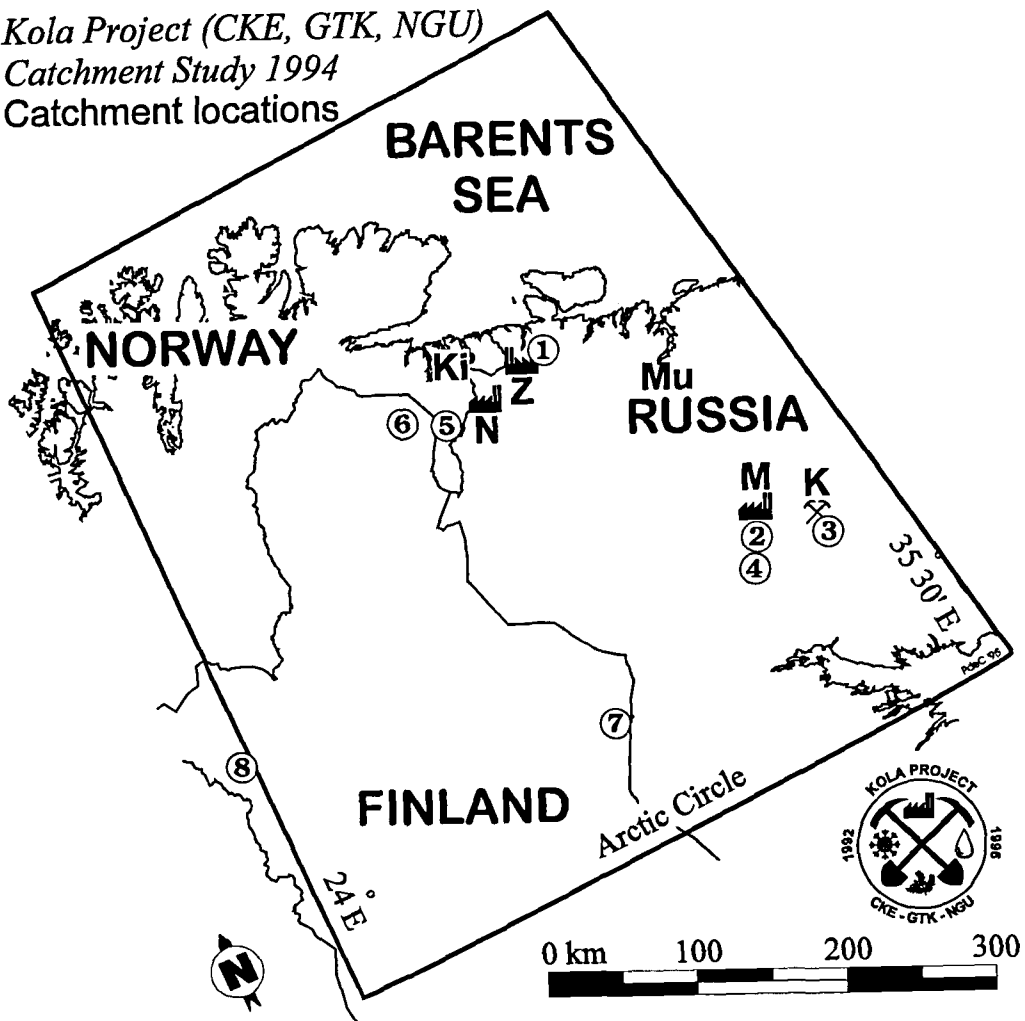


Figure 4.2.1. Location of study area for regional geochemical mapping (frame) and of the eight catchments discussed herein (1: Zapoljarniy, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, and 8: Pallas), and selected townships/industrial centres (K: Kirovsk, Ki: Kirkenes, M: Monchegorsk, Mu: Murmansk, N: Nickel, Z: Zapoljarniy).

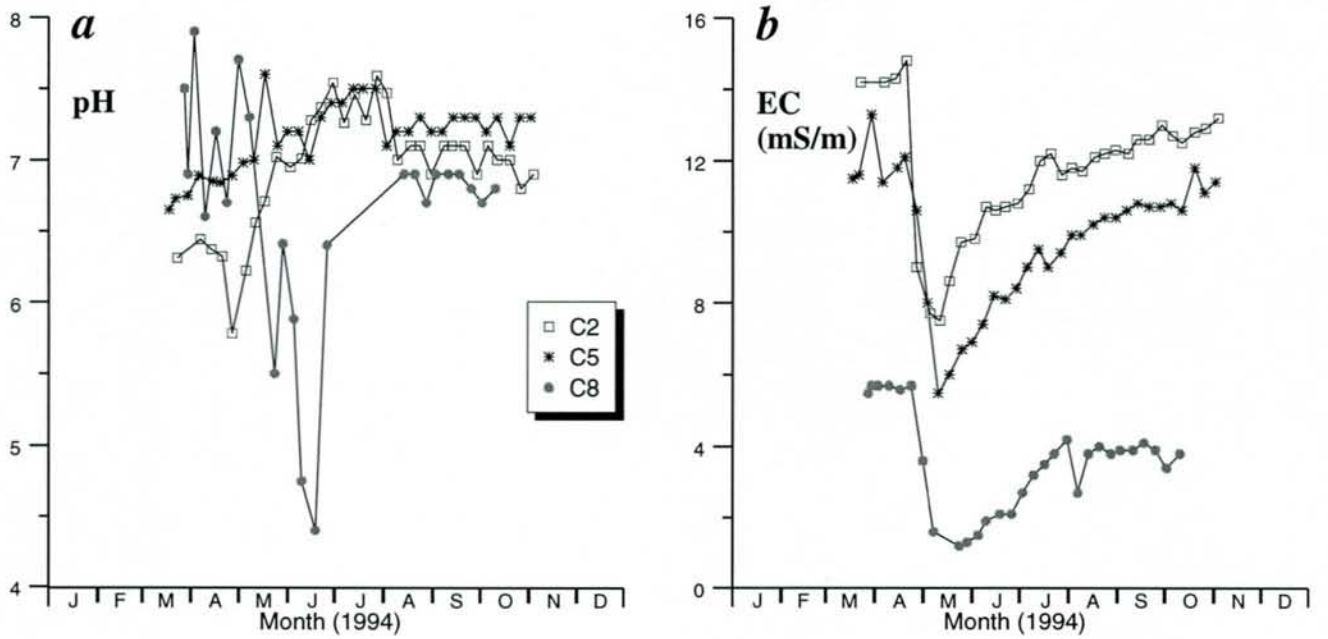


Figure 4.2.2. Time-series for (a) pH, and (b) electrical conductance in the three catchments (C2, C5 and C8) monitored in 1994.

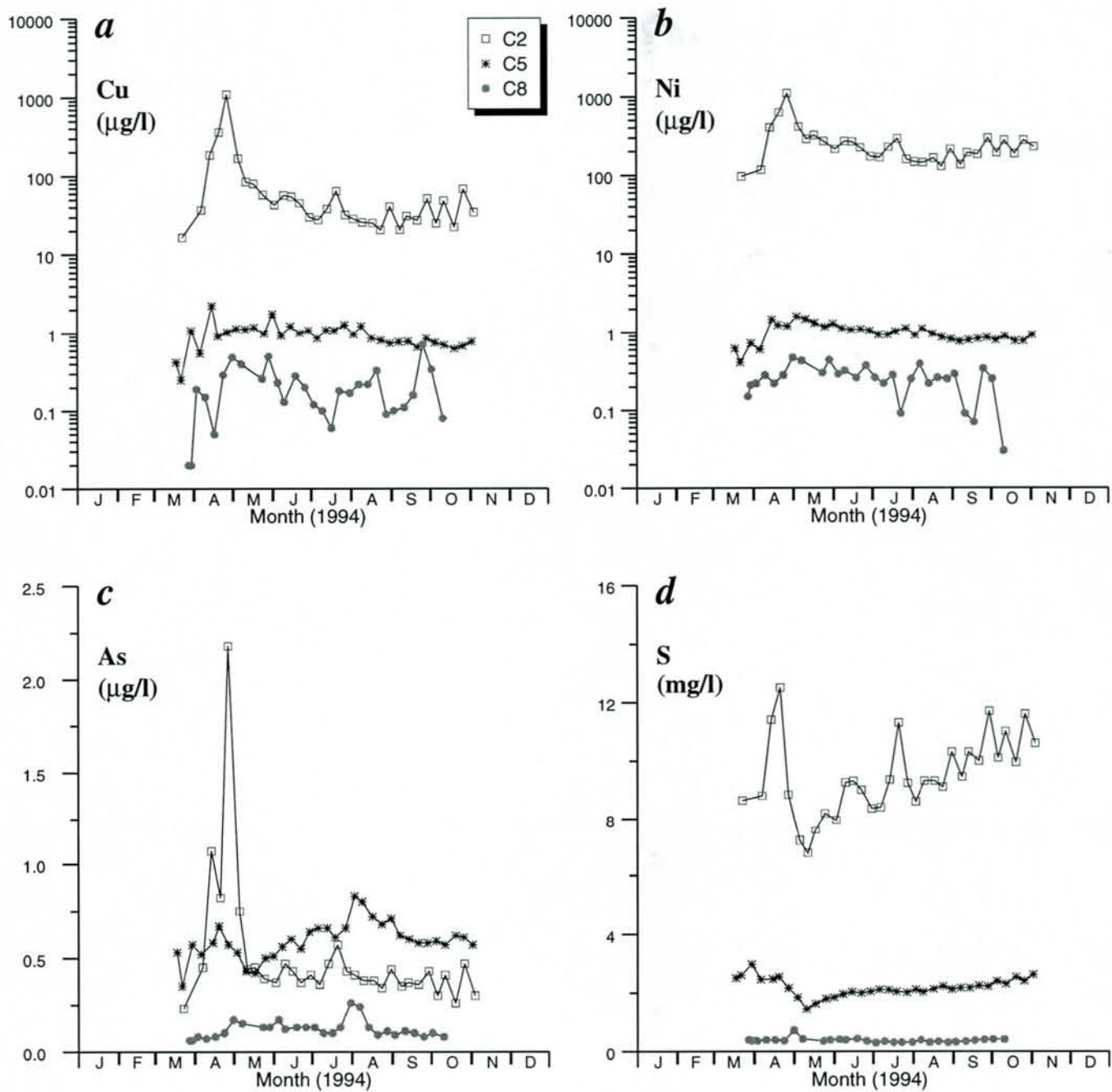


Figure 4.2.3. Time-series for (a) Cu (log scale), (b) Ni (log scale), (c) As, and (d) S in the three catchments (C2, C5 and C8) monitored in 1994.

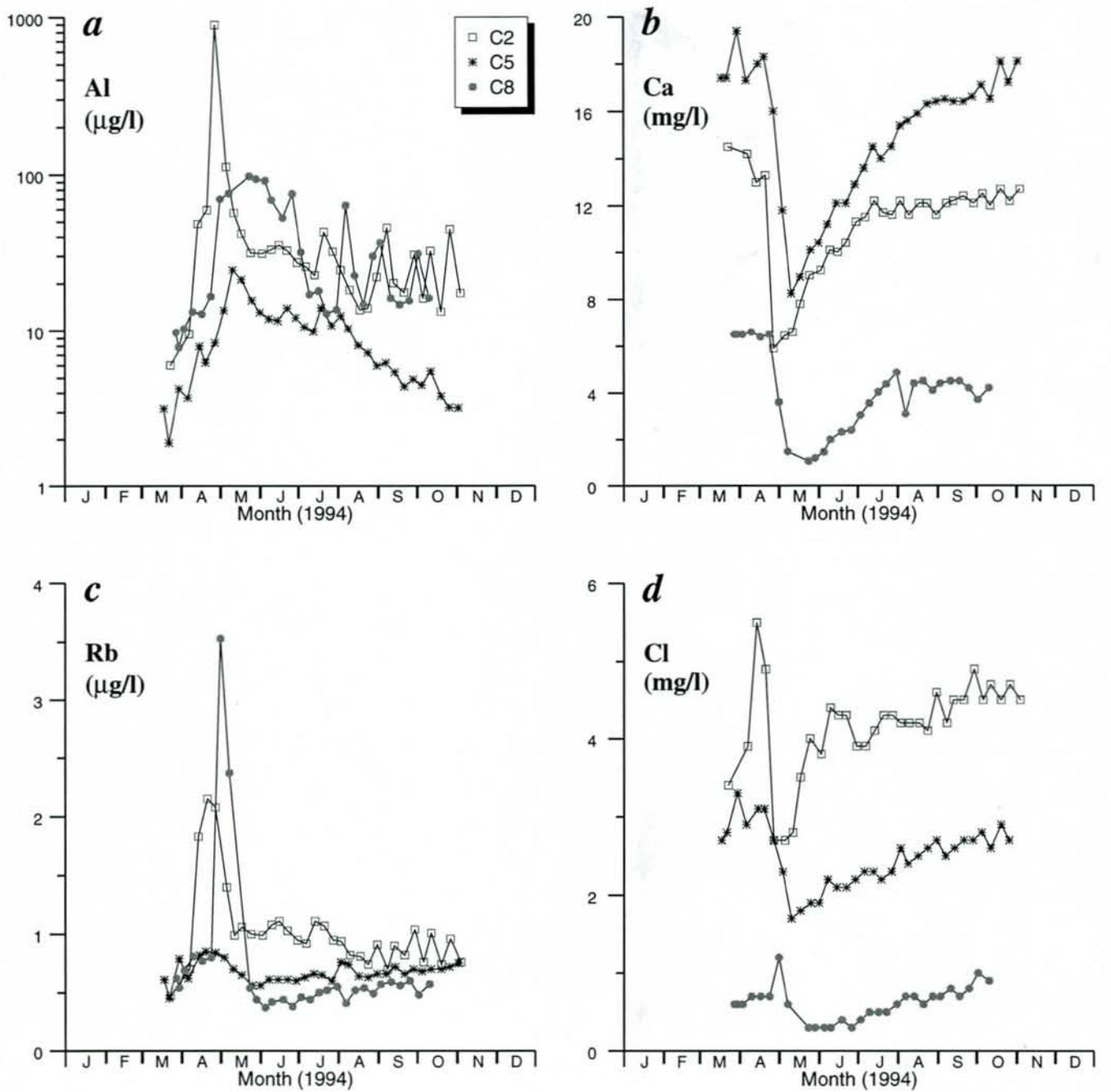


Figure 4.2.4. Time-series for (a) Al (log scale), (b) Ca, (c) Rb, and (d) Cl in the three catchments (C2, C5 and C8) monitored in 1994.

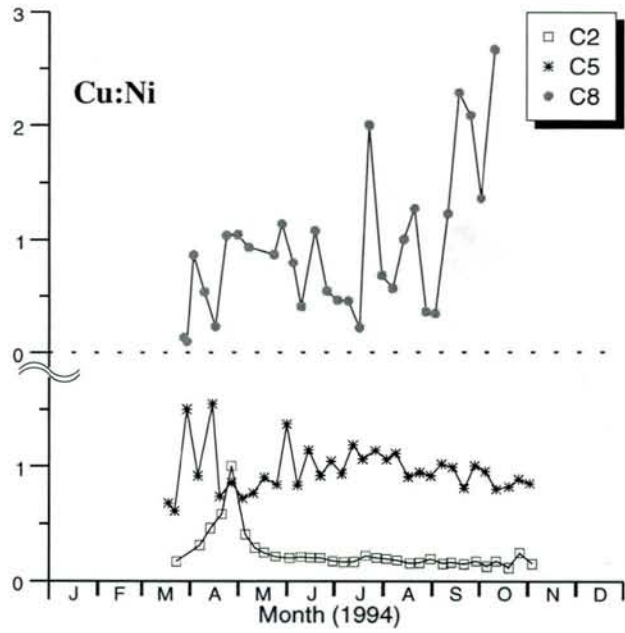


Figure 4.2.5. Time-series for the Cu:Ni ratio in the three catchments (C2, C5 and C8) monitored in 1994.

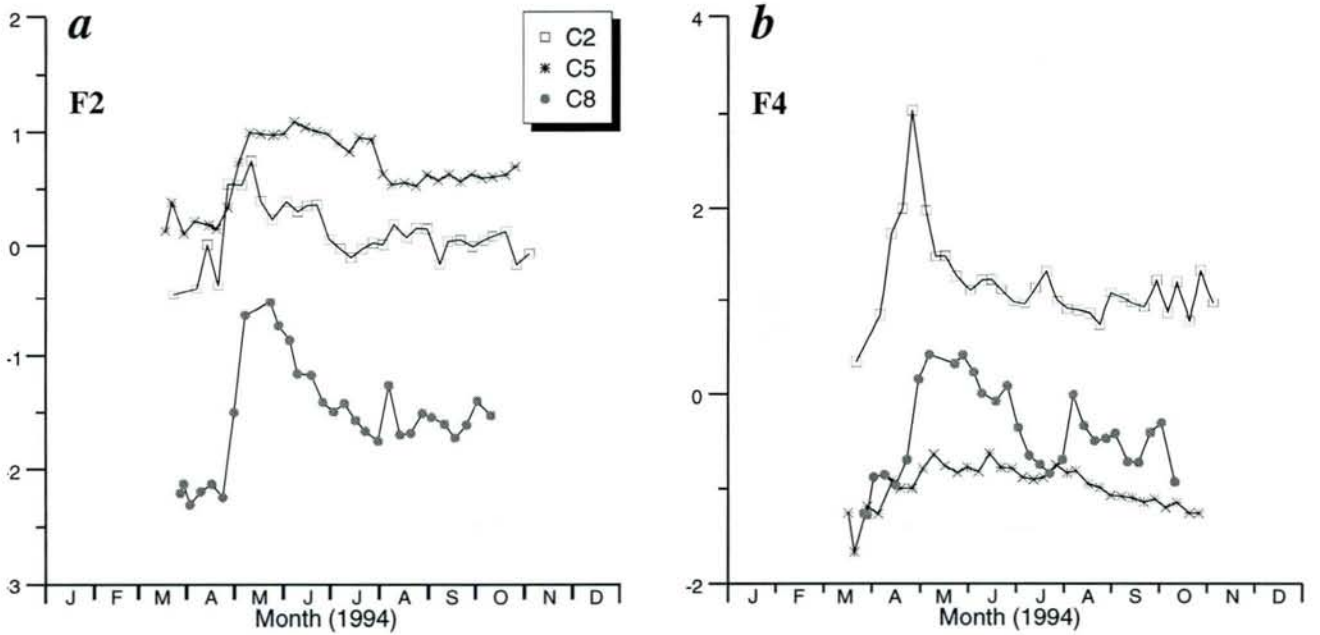


Figure 4.2.6. Time-series for factor-analysis factor (a) F2, and (b) F4 in the three catchments (C2, C5 and C8) monitored in 1994.

### 4.3 Sulphur isotope composition of stream water, moss and humus from eight arctic catchments in the Kola Peninsula region (NW Russia, N Finland, NE Norway)

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#### ABSTRACT

In summer 1994, stream water, moss and humus samples were collected for sulphur isotopic analysis from eight catchments located in the western Kola Peninsula region, where several industrial centres emit high loads of SO<sub>2</sub> and other elements to the atmosphere. Three potential sources of sulphur and their isotopic signature were identified: (1) marine ( $\delta^{34}\text{S}$  +20 to +21‰ CDT), (2) anthropogenic emissions (<+10‰), and (3) geogenic (variable  $\delta^{34}\text{S}$ , mostly <+10‰). Averaged per catchment, the sulphur isotopic composition varies between +6.0 and +16.3‰ for stream water sulphate, +6.0 and +8.4‰ for moss sulphur, and +5.2 and +12.2‰ for humus sulphur. The  $\delta^{34}\text{S}$  composition of stream water from the more remote catchments is quite variable, reflecting several natural (geogenic) sources, but it becomes restricted to the range +8 to +10‰ near the pollution sources. A plot of  $\delta^{34}\text{S}$  vs. 1:SO<sub>4</sub> in stream water suggests that sulphate originating from the smelters has a  $\delta^{34}\text{S}$  value  $\approx$  +9.5‰, and is a dominant source. Sulphur isotope values for moss and humus are consistent with the deduced composition for the emitted sulphur, though for humus a component of geogenic sulphur incorporated via vegetation uptake may play a role. Further isotopic characterisation of atmospheric emissions, together with environmental samples, is needed to better understand sulphur sources and sinks in the area.

#### INTRODUCTION

The western half of the Kola Peninsula in northwestern Russia is the site of several heavy industry and mining centres (the 'Pechenga' complex in Nikel-Zapoljarniy; the 'Severonikel' complex in Monchegorsk; Kirovsk and Apatity; Kandalaksha; among others). These have been polluting the local environment for several decades with impressive emissions of sulphur and trace elements (e.g. Kryuchkov, 1993, Tuovinen et al., 1993, Sivertsen et al., 1994). Of particular interest here are the main sulphur emitting point-sources of Monchegorsk, Nikel and Zapoljarniy (Fig. 4.3.1), where copper-nickel sulphidic ores are being processed and smelted. These ores are in part locally derived, sulphur-poor (~5% S) ores, and, since 1969, in part ores with a particularly high sulphur content (up to 30% S) from the Norilsk region (Alexeyev, 1993, Pozniakov, 1993). Amounts and proportions of these ores are different for the two complexes and have changed through time (e.g. Pozniakov, 1993).

In 1994, the year relevant for the present investigation, ca. 98,000 tonnes SO<sub>2</sub> were emitted from the Monchegorsk smelters, ca. 129,000 tonnes from Nikel, and ca. 69,000 tonnes from Zapoljarniy (MRCENR, 1995, in Reimann et al., 1996). In a cumulative perspective, historical

emissions to air from Monchegorsk alone amount to a staggering 6.5 million tonnes of SO<sub>2</sub> from 1960 to 1994 (data collated from Alexeyev, 1993, Pozniakov, 1993, Baklanov et al., 1994, and MRCENR, 1995, in Reimann et al., 1996)! The close proximity of these world-class point sources of SO<sub>2</sub> (Gunn et al., 1995) with some of the more pristine and remote tracts that can still be found in Europe (i.e. Finnish Lapland), makes this area particularly interesting to study from the viewpoint of environmental sulphur isotopy.

The primary aim of the present investigation is to test whether these massive sulphur emissions have left behind a recognisable, and thus traceable, sulphur isotopic signal in the environment, a task which, to our knowledge, has not been accomplished in this region heretofore. Eight catchments (watersheds or drainage basins) were selected for geochemical investigation by the Geological Surveys of Finland and Norway and the Central Kola Expedition (Russia) (e.g. Reimann et al., 1995, Äyräs et al., 1995, Caritat et al., in prep. a,b), as part of the cooperative 'Kola Ecogeochemistry Project' (see World Wide Web site <http://www.ngu.no/Kola>). The main characteristics of the catchments are summarised in Table 4.3.1. Figure 4.3.1 illustrates the location of the catchments together with the major hydrographic attributes, the location of major industry and roads, and a wind rose for the Monchegorsk area. Northerly and southerly winds are dominant over most of the area where the point sources of sulphur emission are found (Tuovinen et al., 1993, Mäkinen, 1994). Main drainage direction on the Kola Peninsula is also north-south (to the Barents Sea or the White Sea). The principal lithological features of the area are shown in Figure 4.3.2.

#### SAMPLING, METHODS AND NOTATION

Samples of stream water, moss (*Hylocomium splendens* and *Pleurozium schreberi*) and humus were collected from the eight catchments (Fig. 4.3.1) in July-August 1994 for sulphur isotope analysis. Duplicate stream water samples (2 × 5 litre) were collected near the outlet of each catchment, whereas several locations within each catchment were sampled for moss and humus.

Field procedures are described in detail in Äyräs & Reimann (1995). Stream water was sampled, filtered through 0.45 µm Millipore™ membrane filter and acidified (pH 4.5 to 5), before addition of barium chloride in the field to precipitate BaSO<sub>4</sub>. A separate stream water sample was collected simultaneously for the determination of pH, electrical conductance and chemical composition. The sulphate content (ion chromatography) of this water sample is quoted in Table 4.3.2. Additional stream water samples for chemical analysis were collected with variable frequency from all catchments between March and November 1994 (Caritat et al., in prep. a,b).

Moss and humus samples were carefully collected and air-dried. The median sulphur content of moss and humus (see Äyräs et al., in prep.) is also reported in Table 4.3.2.

Analytical methods for stream water consisted in washing and separation of the BaSO<sub>4</sub> filtrate, which was subsequently dried and mixed with SiO<sub>2</sub> and V<sub>2</sub>O<sub>5</sub> for high-temperature conversion to SO<sub>2</sub>, following the method of Yanagisawa & Sakai (1983) and Ueda & Krouse (1986). The sulphur isotope composition of the SO<sub>2</sub> was determined with a stable isotope mass spectrometer built around basic VG Micromass 602 components.

For moss and humus, total sulphur was extracted from up to 1 g of air-dried material by combustion in a Parr Instrument Co. oxygen bomb using 22 atm O<sub>2</sub>. Water added prior to



combustion was combined with bomb washings, and a few drops of 50% H<sub>2</sub>O<sub>2</sub> were added to assure complete oxidation of sulphur compounds to sulphate. After filtering, BaCl<sub>2</sub> solution was added to precipitate BaSO<sub>4</sub>, which was treated as detailed above for the stream water samples.

The sulphur isotope abundance of a specimen, expressed in permil (‰) on a δ<sup>34</sup>S scale, reflects the deviation in parts per thousands of its <sup>34</sup>S/<sup>32</sup>S abundance ratio from that of the defined standard V-CDT:

$$\delta^{34}\text{S (in ‰ CDT)} = \left[ \frac{(^{34}\text{S}/^{32}\text{S})_{\text{specimen}}}{(^{34}\text{S}/^{32}\text{S})_{\text{standard}}} - 1 \right] \times 1000. \quad (1)$$

## RESULTS

In total, 82 sulphur isotope determinations were made, and results are given in Table 4.3.2 and illustrated in Figure 4.3.3. No sulphur isotope data are reported for moss from Catchment 2 (Monchegorsk), as no moss exists there as a result of massive airborne pollution ('technogenic desert'). Duplicate analyses of stream water samples indicate reproducibility of δ<sup>34</sup>S values better than 0.3‰ in all cases but one, where it was 0.6‰ (Catchment 3).

Averaged per catchment, δ<sup>34</sup>S values vary between +6.0 and +16.3‰ for stream water, +6.0 and +8.4‰ for moss, and +5.2 and +12.2‰ for humus. Maximum measurement frequency classes for each sampling medium are: +9 to +10‰ (40% of the data) for stream water, +6 to +7‰ (34%) for moss, and at +6 to +7‰ (24%) for humus (Fig. 4.3.3).

## INTERPRETATION

### *Rationale*

The isotopic composition of stream water sulphate and of moss and humus sulphur can be used as a sulphur tracer, as it is thought to vary only slightly during sulphur transformations under aerobic conditions (Krouse et al., 1991). Sulphate assimilation (conversion of SO<sub>4</sub><sup>2-</sup> to S-amino acids, etc.) by plants and bacteria is accompanied by quite small shifts in δ<sup>34</sup>S values (Krouse et al., 1991). This fact will be particularly true in Arctic regions because of the lower level of bacterial activity under cool climates. It has been shown that the sulphur isotopic composition of epiphytic lichens and of many mosses tends to be close to that of atmospheric S compounds, implying rather direct uptake mechanisms of gaseous (and perhaps dissolved) sulphur compounds with little attending isotopic selectivity (Krouse, 1977, Winner et al., 1978, Case and Krouse, 1980, Krouse, 1989). Therefore, it will be attempted to use the observed sulphur isotope compositions in the eight catchments to differentiate sulphur sources of distinct sulphur isotopic character.

### *Sulphur sources*

The most important sources contributing atmospheric sulphur to the area are: (1) natural seaspray from the Barents sea, and (2) anthropogenic SO<sub>2</sub> emissions from local industry (primarily at Monchegorsk, Nickel and Zapoljarniy). Average δ<sup>34</sup>S of sea water sulphate is remarkably uniform with values in the range +20 to +21‰ (e.g. Thode et al., 1961, Rees et al., 1978, Longinelli, 1989). The second source, industrial emissions (for which no δ<sup>34</sup>S data exist), results from processing of sulphide ore, which is partly local ore and partly sulphur-richer Norilsk ore. Average δ<sup>34</sup>S values of local ores are lower than +4.2‰, whereas Norilsk ore has

$\delta^{34}\text{S}$  values between +4.9 and +10.6‰, with an average of +8.2‰ (Grinenko & Grinenko, 1974). Given the high amounts and high sulphur contents (up to 30%) of the Norilsk ore smelted on Kola, resulting  $\text{SO}_2$  emissions are likely to have a  $\delta^{34}\text{S}$  compositions in the range +5 to +10‰. Therefore, in simplified terms, the two most significant sources of atmospheric S in the area (Fig. 4.3.3) have  $\delta^{34}\text{S}$  values either  $\geq +20\text{‰}$  (sea water sulphate) or  $\leq +10\text{‰}$  (industrial emissions).

A third source of sulphur that is not atmospheric but found within the terrestrial ecosystems themselves, is the (3) geogenic sulphur pool. In inland areas devoid of important local atmospheric pollution, a strong lithological control on the  $\delta^{34}\text{S}$  of stream water sulphate has been demonstrated, indicating a subordinate role of bacterial fractionation (e.g. Hitchon & Krouse, 1972, Cameron et al., 1995). In very general terms, sedimentary sulphate minerals mostly have positive  $\delta^{34}\text{S}$  values, sedimentary sulphides have negative  $\delta^{34}\text{S}$  values, and igneous sulphides most often have  $\delta^{34}\text{S}$  values near 0‰. The four catchments in Russia are located in Proterozoic greenstone belts with mafic/ultramafic intrusions, thus giving anticipated near-zero  $\delta^{34}\text{S}$  values for geogenic sulphur. Melezhic et al. (1994) reported  $\delta^{34}\text{S}$  values mainly in the range -2 to +6‰ for sulphide ores from the Pechenga region (Zapoljarniy). They also analysed local sedimentary sulphides, which fall mostly within the ranges -4 to +4‰ for synsedimentary and early diagenetic sulphides, or +6 to +16‰ for late stage diagenetic sulphides. The igneous and early diagenetic sulphide sources are expected to be isotopically indistinguishable from the local atmospheric pollution source discussed above. It should also be noted that the  $\delta^{34}\text{S}$  composition of till deposits and groundwater may be highly variable (e.g. Hendry et al., 1986, 1989, Fennell et al., 1994, Van Donkelaar et al., 1995, Grasby et al., in press).

In addition to the above three sources, (4) biogenic sulphur and bacterial fractionation of S may play an important role. Here, we are specifically concerned about sulphur mineralization and biogenic sulphate reduction (reoxidized organic sulphur from plants and animals, fungal conversion in soil, etc.).

#### *Stream water*

Stream water sulphate can contain geogenic, marine, anthropogenic, and biogenic sulphur. As such, it is a complex parameter to interpret without detailed knowledge of the contribution from each of these four sources.

A map of the distribution of stream water  $\delta^{34}\text{S}$  values averaged per catchment (Figure 4.3.4) shows that  $\delta^{34}\text{S}$  values recorded at Catchments 1-5, i.e. those located closest to the major industrial pollution centres in the area (primarily Nickel, Zapoljarniy and Monchegorsk, and to a much lesser extent Kirovsk and Kirkenes), are in the range +6.0 to +9.7‰. This range is coincident with that expected either from atmospheric emission of nickel ore-derived sulphur or from indigenous igneous sulphides, with no or very minor mixing with sulphur of marine origin and with little modification by bacterially mediated sulphur transformation in the catchments. Indeed, the most ecologically damaged catchment (Catchment 2) has a  $\delta^{34}\text{S}$  values of +9.1‰. Since this area is in large parts devoid of vegetation and soil as a result of industrial pollution ('technogenic desert'), the isotopic composition of the stream water sulphate there reflects most closely atmospheric and geogenic sources, without the complicating effects of biological fractionation. Indeed, the  $\delta^{34}\text{S}$  value there is close to the average Norilsk ore value.

Catchments 6-8 in Finland have  $\delta^{34}\text{S}$  values between +10.1 and +16.3‰, i.e. clearly higher than for the other catchments. The high values in Catchments 7 and 8 can not be explained by

mixing with significant amounts of marine sulphur, as these catchments are more continental than, say, Catchments 1 or 5. A contribution from isotopically heavier, geogenic sulphur currently appears to be the best explanation of the higher values obtained. This would also be consistent with the very similar  $\delta^{34}\text{S}$  values of stream water in Catchments 7 and 8.

The diagram of  $\delta^{34}\text{S}$  vs.  $\text{SO}_4$  concentration (Figure 4.3.5a) shows a trumpet-shaped distribution resulting from the coexistence of several subordinate sulphur sources of diverse isotopic composition with one dominant sulphur source of fixed isotopic composition but of variable influence ('Case 8' in Fig. 11-4 of Krouse, 1980). Catchments 3, 5, 4 and 2 define well the lower curve of the domain, and when plotted in the classic (e.g. Krouse, 1980)  $\delta^{34}\text{S}$  vs.  $1:\text{SO}_4$  space (Fig. 4.3.5a, inset), they yield a straight line, from which the  $\delta^{34}\text{S}$  composition of the dominant source (the industrial emissions from the smelters) can be deduced ( $\approx +9.5\text{‰}$ ). Note that this isotopic composition is also consistent with expected geogenic sulphur values for igneous sulphides within the local rocks, but the very high loads of sulphur emission and deposition versus the slow rate of sulphide weathering have to be kept in mind.

It is possible to separate the individual catchments in Figure 4.3.5a into two groups. The first group, consisting of the least polluted stream waters (Catchments 3, 6, 7 and 8), has low  $\text{SO}_4$  content ( $<4\text{ mg/l}$ ) and a wide range of  $\delta^{34}\text{S}$  values ( $+6.0$  to  $+16.3\text{‰}$ ). Catchment 3 ( $\delta^{34}\text{S} = +6.0\text{‰}$ ) is located in an area of apatite quarrying and handling; the till-covered bedrock is dominantly apatite-nephelinite ('hibinite'). It is not known what  $\delta^{34}\text{S}$  signature any S-bearing minerals present in the bedrock or Quaternary deposits might have. But, clearly, the stream water in this catchment is not significantly influenced by sulphur emissions from nearby Monchegorsk, as a result of the dominant wind directions. Catchment 6 ( $\delta^{34}\text{S} = +10.1\text{‰}$ ) is close to the sea, and the  $\delta^{34}\text{S}$  composition of its stream water might reflect a mix of geogenic and marine S-sources. Stream water there has  $\text{Cl}:\text{SO}_4 \approx 1$  (Caritat et al., in prep. a), evidently closer to the sea water ratio ( $\approx 7$ ) than any of the other catchments studied here (0.2 to 0.6), supporting a marine influence. According to equation (2) of Wadleigh et al. (1994):

$$\text{Marine sulphate (\%)} = \left\{ \frac{0.14}{(\text{SO}_4/\text{Cl})_{\text{sample}}} \right\} \times 100, \quad (2)$$

up to 14% of the stream water sulphate in Catchment 6 may have a marine origin, assuming Cl to be conservative and sourced solely from the sea. In Catchments 7 and 8 ( $\delta^{34}\text{S} = +16.1$  and  $+16.3\text{‰}$ ), the isotopically heavy sulphur would seem to be geogenic, although the high  $\delta^{34}\text{S}$  values appear inconsistent with the bedrock type (gneiss and quartzite).

The second group, representing the more polluted stream waters (Catchments 5, 1, 4 and 2), is characterised by higher  $\text{SO}_4$  contents (6 to 27 mg/l) and a narrower range of intermediate  $\delta^{34}\text{S}$  values ( $+7.8$  to  $+9.7\text{‰}$ ). The simplest explanation for this trend is that, as pollution and atmospheric S deposition increase, the stream water  $\delta^{34}\text{S}$  becomes more influenced by the isotopic composition of the sulphide ores (ca.  $+9\text{‰}$ ).

Plotting the  $\delta^{34}\text{S}$  composition vs. another pollution indicator, for instance the Ni content (Caritat et al., in prep. a) of the stream waters (Fig. 4.3.5b), yields essentially the same picture, with a group of least polluted stream waters with Ni content  $< 0.5\text{ }\mu\text{g/l}$  and widely variable  $\delta^{34}\text{S}$  (shaded envelope in Fig. 4.3.5b), and a group of progressively more polluted stream

waters with Ni content up to 100 µg/l or more and a narrower range of  $\delta^{34}\text{S}$  compositions (dark grey arrow in Fig. 4.3.5b).

The use of geogenic indicators like the Se content in stream water is hampered by the fact that the majority of the results are below detection limit (0.5 µg/l), except for a few values in Catchment 2 (up to 1.4 µg/l).

#### *Moss*

Since mosses receive the bulk of their nutrients directly from wet and dry deposition, their sulphur isotopic composition should more faithfully and more straightforwardly reflect atmospheric sulphur isotopic composition (Krouse, 1989) than either stream water or humus do. In this respect, it is interesting to note that the recorded variation in moss  $\delta^{34}\text{S}$  values is very small (+6.0 to +8.4‰) in all catchments where moss was found (Figure 4.3.6). These values are all consistent with the range of values from Norilsk ore. This indicates that, as the air becomes cleaner with distance away from the pollution sources (from  $>50 \mu\text{g SO}_2/\text{m}^3$  near Nikel to  $<10 \mu\text{g SO}_2/\text{m}^3$  20 km to the south and south-west, e.g. Sivertsen et al., 1994), airborne sulphur retains the same isotopic composition, suggesting that there is no mixing with S from other sources, at least in these directions. Thus, atmospheric sulphur concentration decreases purely by dilution as it is progressively deposited by wet or dry processes with distance from source.

#### *Humus*

The map of catchment-averaged  $\delta^{34}\text{S}$  composition of humus (Figure 4.3.7) shows, with the exception of Catchment 6 (+12.2‰), a narrow range of values (+5.2 to +8.3‰) with no evident trend with distance away from the industrial centres. This supports the interpretation given above for moss, namely that no other sulphur source of importance mixes into the air away from the smelters. The only exception to this is Catchment 6, where the higher value is interpreted to indicate minor influence of marine sulphur. This is also consistent with the above discussion based on stream water isotopic composition.

## CONCLUSIONS

This preliminary investigation of the sulphur isotopic composition of stream water, moss and humus from the Kola Peninsula has yielded the following results:

(1) Three main sources of sulphur input into the ecosystems were identified: industrial emissions ( $\delta^{34}\text{S}$  between +5 and +10‰ CDT), marine sulphur ( $\geq +20$ ‰), and geogenic sulphur (variable  $\delta^{34}\text{S}$  as a function of sulphur-bearing rock types; most important will be igneous sulphides,  $\delta^{34}\text{S}$  between 0 and +10‰).

(2) Averaged per catchment, recorded  $\delta^{34}\text{S}$  values vary between +6.0 and +16.3‰ for stream water, +6.0 and +8.4‰ for moss, and +5.2 and +12.2‰ for humus.

(3) Stream water sulphate  $\delta^{34}\text{S}$  values are lower near the pollution centres (+6.0 to +9.7‰, Catchments 1-5), reflecting industrial sulphur emissions (and/or local geogenic sulphur), and increase further away from these (+10.1 to +16.3‰, Catchments 6-8), reflecting relatively less technogenic influence. A diagram of  $\delta^{34}\text{S}$  vs.  $\text{SO}_4$  concentration shows a distribution typical of one dominant source of constant isotopic composition ( $\approx +9.5$ ‰) coexisting with several minor sources of inhomogenous isotopic composition (+6 to +16‰). Plotted against Ni

content of stream water,  $\delta^{34}\text{S}$  shows clearly a tendency toward values in the range +9 to +10‰ with increasing pollution.

(4) Moss and humus  $\delta^{34}\text{S}$  results are mostly restricted to a narrow range (+6 to +10‰) consistent with the anthropogenic emissions but, for humus, indigenous sulphur from bedrock or Quaternary deposits may play a role via vegetation uptake. Moss results indicate that no mixing of other sulphur sources occurs in the atmosphere with increasing distance from the smelters. Humus from Catchment 6 (+12.2‰) could contain a proportion of heavy, marine sulphur.

(5) There is a need for gathering more information on potential sulphur sources, sampling more locations considering the area covered, and investigating directly the sulphur isotopic composition of emissions and of the atmosphere, if we are to better understand the processes controlling the sulphur dynamics in this area.

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## TABLES

Table 4.3.1. Main characteristics of the eight catchments

Table 4.3.2. Sulphur isotope analysis results. Stream water:  $\delta^{34}\text{S}$  ( $\text{SO}_4$ ) and  $\text{SO}_4$  concentration in samples collected at the same time; Moss and humus:  $\delta^{34}\text{S}$  (total S) and median S concentration.

## FIGURES

Figure 4.3.1. Location map showing the eight catchments discussed here (circles numbered 1: Zapoljarniy, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, and 8: Pallas), Kola Project area (frame), major lakes and rivers, selected townships, industry (smelters, mines, power plant), roads (dashed lines), and a wind rose for the Monchegorsk area (from Mäkinen, 1994).

Figure 4.3.2. Main geological (lithological) features of the study area.

Figure 4.3.3. Histograms of sulphur isotopic compositions recorded for stream water, moss and humus. The number within each box refers to the catchment number. Typical  $\delta^{34}\text{S}$  values for Kola ores, Norilsk ores and sea water are also indicated.

Figure 4.3.4. Map of catchment-averaged sulphur isotopic composition of stream water.

Figure 4.3.5.  $\delta^{34}\text{S}$  composition vs.  $\text{SO}_4$  content (a) and vs. Ni content (b) for stream water. Numbers 1 to 8 refer to catchment numbers. The horizontal, double-arrowheaded lines represent the range of  $\text{SO}_4$  or Ni concentrations recorded for each catchment during the whole monitoring period (March-November 1994, Caritat et al., in prep. a,b). The errors of each measurement are equal to or smaller than the symbol size.

Figure 4.3.6. Map of catchment-averaged sulphur isotopic composition of moss. No moss was found in Catchment 2 (n/a: not applicable).

Figure 4.3.7. Map of catchment-averaged sulphur isotopic composition of humus.

Table 4.3.1. Main characteristics of the eight catchments

No.	Name	Coordinates of catchment outlet	Size (km <sup>2</sup> )	Elevation (m a.s.l.)	Annual precip. (mm)*	Vegetation	Bedrock	Surface cover, peculiarities
RUSSIA								
C1	Zapoljarniy	69°27'01"N 31°03'49"E	19.02	25-373	454	birch forest tundra	gneiss	till, fluvio-glacial, outcrop
C2	Monchegorsk	67°50'30"N 32°54'48"E	22.38	128-507	391	technogenic desert, birch shrubs	dacite & andesite & tuffs, gabbro/norite	till, prone to erosion
C3	Kirovsk	67°32'50"N 33°48'55"E	20.01	240-1075	502	spruce forest, mountain tundra birch forest	nephelinite	till, diluvial/eluvial
C4	Kurka	67°41'25"N 32°50'14"E	20.49	152-466	502	north taiga spruce forest, birch; incipient deterioration	amphibolite, gneiss	till, fluvio-glacial
NORWAY								
C5	Skjellbekken	69°21'25"N 29°27'25"E	34.56	80-297	422	north taiga pine forest, birch	andesite, basalt & tuffs, 'black shale'	till, esker
FINLAND								
C6	Kirakka	69°35'12"N 28°51'46"E	11.86	110-200	386	north taiga pine forest	granite	outcrop, till, moraine ridge
C7	Naruska	67°21'44"N 29°22'05"E	20.16	263-490	513	north taiga spruce forest	gneiss	till, peat, outcrop
C8	Pallas	68°09'14"N 23°52'50"E	24.42	303-500	405	north taiga spruce forest	quartzite	till, peat

\* from the closest meteorological station (data from 1994)

Table 4.3.2. Sulphur isotope analysis results. Stream water:  $\delta^{34}\text{S}$  ( $\text{SO}_4$ ) and  $\text{SO}_4$  concentration in samples collected at the same time; Moss and humus:  $\delta^{34}\text{S}$  (total S) and median S concentration.

Catch.	Sample	$\delta^{34}\text{S}$ (‰ CDT)	Catch. av. $\delta^{34}\text{S}$ (‰ CDT)	$\text{SO}_4$ (mg/l)	Catch.	Sample	$\delta^{34}\text{S}$ (‰ CDT)	Catch. av. $\delta^{34}\text{S}$ (‰ CDT)	S (mg/kg)
<b>Stream water</b>					<b>Humus</b>				
C1	SW1	9.6			C1	HU1	7.4		
C1	SW2	9.8	9.7	9.8	C1	HU2	8.1		
C2	SW3	9.1			C1	HU3	9.7		
C2	SW4	9.3			C1	HU4	13.3	9.6	1435
C2	SW5	8.9			C2	HU5	6.6		
C2	SW6	9.0	9.1	26.5	C2	HU6	6.6		
C3	SW7	6.6			C2	HU7	7.1		
C3	SW8	5.4	6.0	2.8	C2	HU8	9.7	7.5	1188
C4	SW9	9.1			C3	HU9	4.7		
C4	SW10	8.5	8.8	10	C3	HU10	5.2		
C5	SW11	7.9			C3	HU11	6.2	5.3	1400
C5	SW12	7.7	7.8	6.1	C4	HU12	3.6		
C6	SW13	10.1	10.1	1.8	C4	HU13	4.2		
C7	SW14	16.1	16.1	1.3	C4	HU14	4.7		
C8	SW15	16.3	16.3	1	C4	HU15	5.5		
					C4	HU16	6.2		
					C4	HU17	7.1	5.2	1400
					C5	HU18	7.1		
					C5	HU19	7.1		
					C5	HU20	7.3		
					C5	HU21	8.9		
					C5	HU22	9.6		
					C5	HU23	9.7	8.3	1325
					C6	HU24	10.4		
					C6	HU25	11.2		
					C6	HU26	11.9		
					C6	HU27	12.8		
					C6	HU28	14.6	12.2	1360
					C7	HU29	5.7		
					C7	HU30	6.1		
					C7	HU31	6.6		
					C7	HU32	7.2		
					C7	HU33	10.0		
					C7	HU34	10.4	7.7	1300
					C8	HU35	6.3		
					C8	HU36	6.4		
					C8	HU37	6.7		
					C8	HU38	9.0	7.1	1470
Catch.	Sample	$\delta^{34}\text{S}$ (‰ CDT)	Catch. av. $\delta^{34}\text{S}$ (‰ CDT)	S (mg/kg)					
<b>Moss</b>									
C1	MO1	6.6	6.6	1470					
C3	MO2	5.6							
C3	MO3	5.6							
C3	MO4	4.3							
C3	MO5	6.9							
C3	MO6	7.6	6.0	947					
C4	MO7	8.4	8.4	1190					
C5	MO8	5.6							
C5	MO9	6.3							
C5	MO10	6.4							
C5	MO11	6.4							
C5	MO12	6.6							
C5	MO13	6.9	6.4	884					
C6	MO14	5.6							
C6	MO15	7.7							
C6	MO16	8.4							
C6	MO17	8.4							
C6	MO18	8.7	7.7	757					
C7	MO19	5.6							
C7	MO20	6.4							
C7	MO21	6.0							
C7	MO22	7.0							
C7	MO23	7.1							
C7	MO24	7.8	6.6	714					
C8	MO25	6.3							
C8	MO26	7.3							
C8	MO27	7.4							
C8	MO28	7.8							
C8	MO29	8.1	7.4	760					

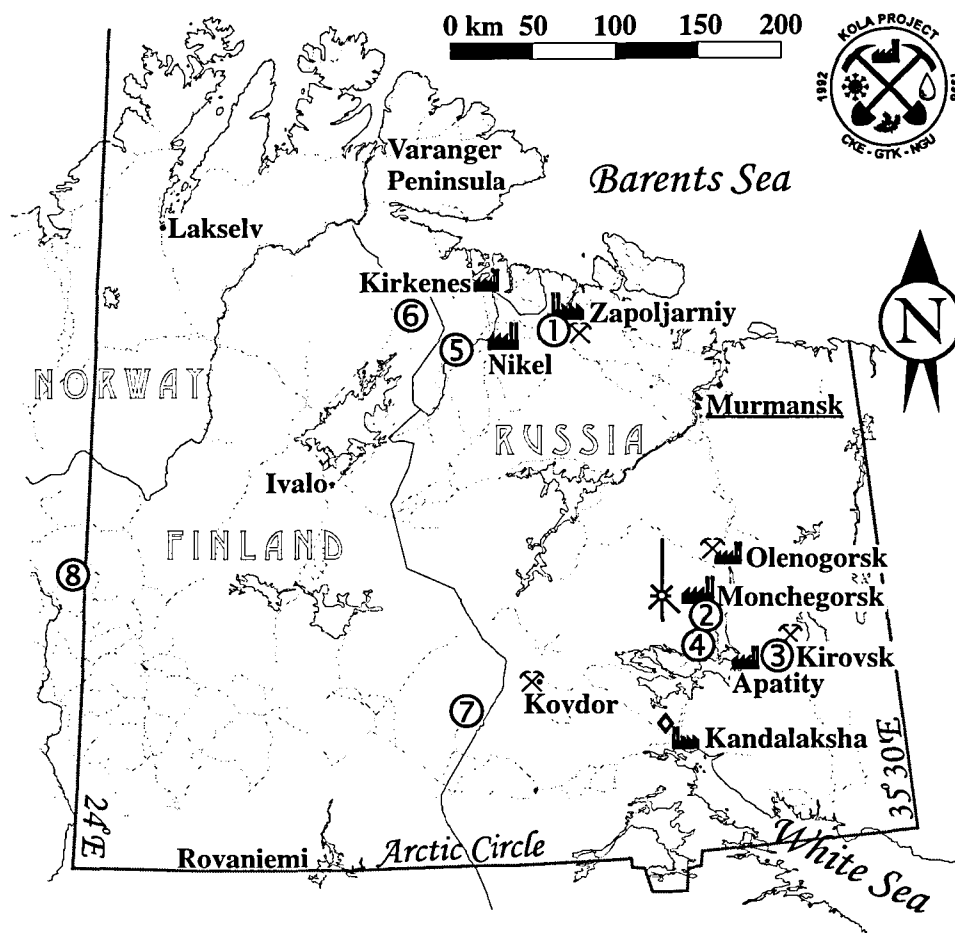


Figure 4.3.1. Location map showing the eight catchments discussed here (circles numbered 1: Zapoljarniy, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, and 8: Pallas), Kola Project area (frame), major lakes and rivers, selected townships, industry (smelters, mines, power plant), roads (dashed lines), and a wind rose for the Monchegorsk area (from Mäkinen, 1994).

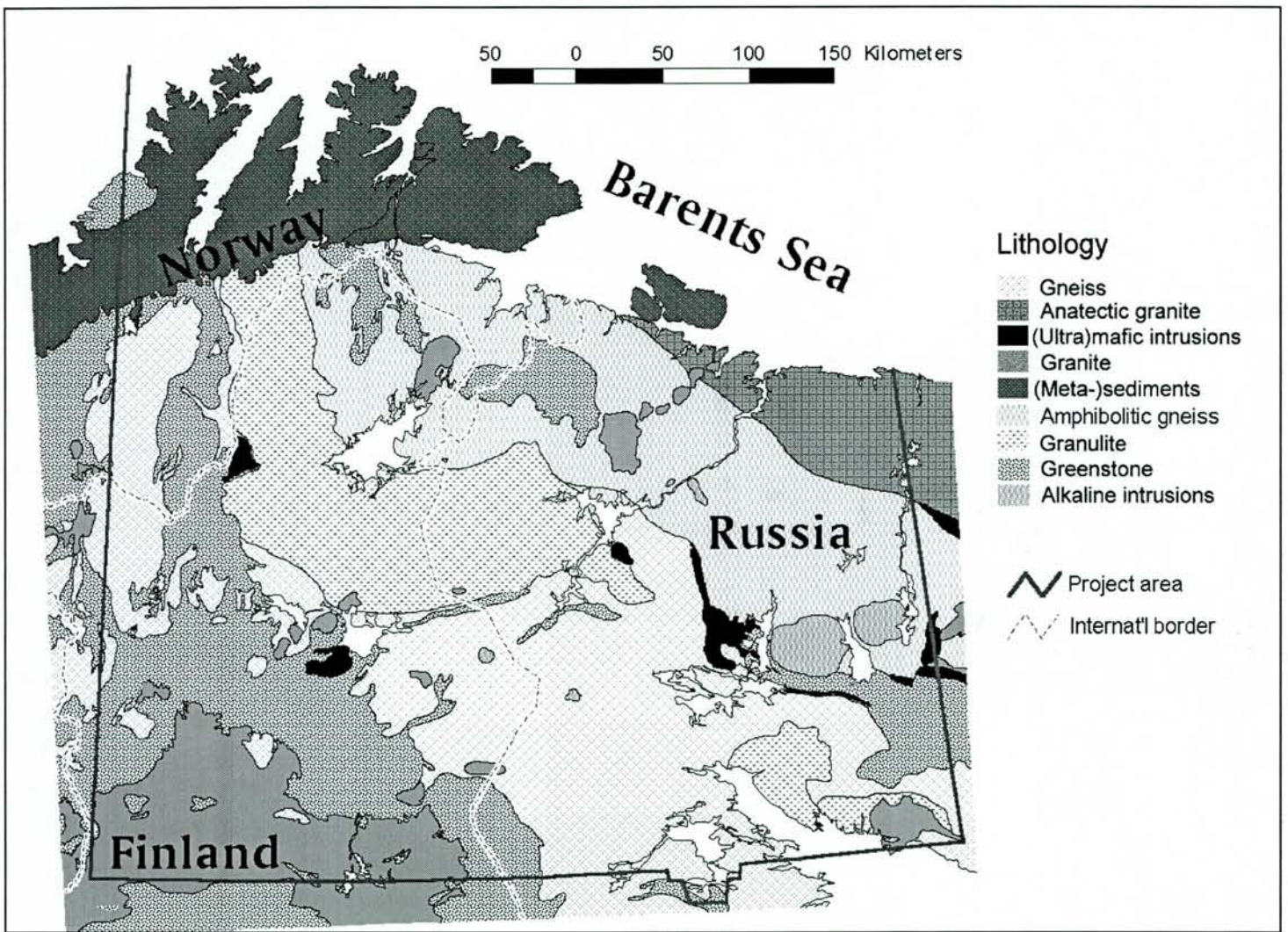
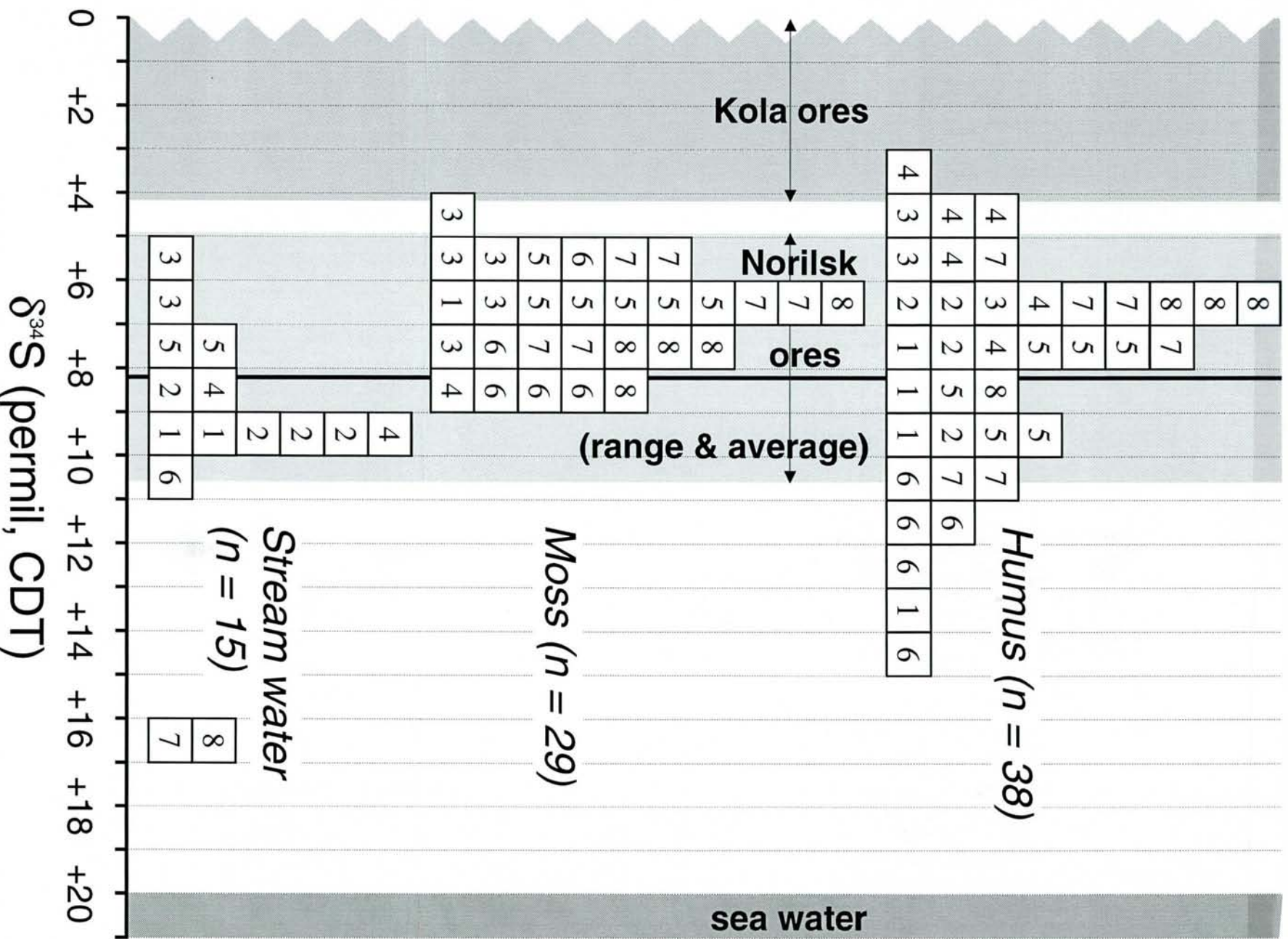


Figure 4.3.2. Main geological (lithological) features of the study area.



Figure 4.3.3. Histograms of sulphur isotopic compositions recorded for stream water, moss and humus. The number within each box refers to the catchment number. Typical  $\delta^{34}\text{S}$  values for Kola ores, Norilsk ores and sea water are also indicated.



# Stream water $\delta^{34}\text{S}$ (‰, CDT)

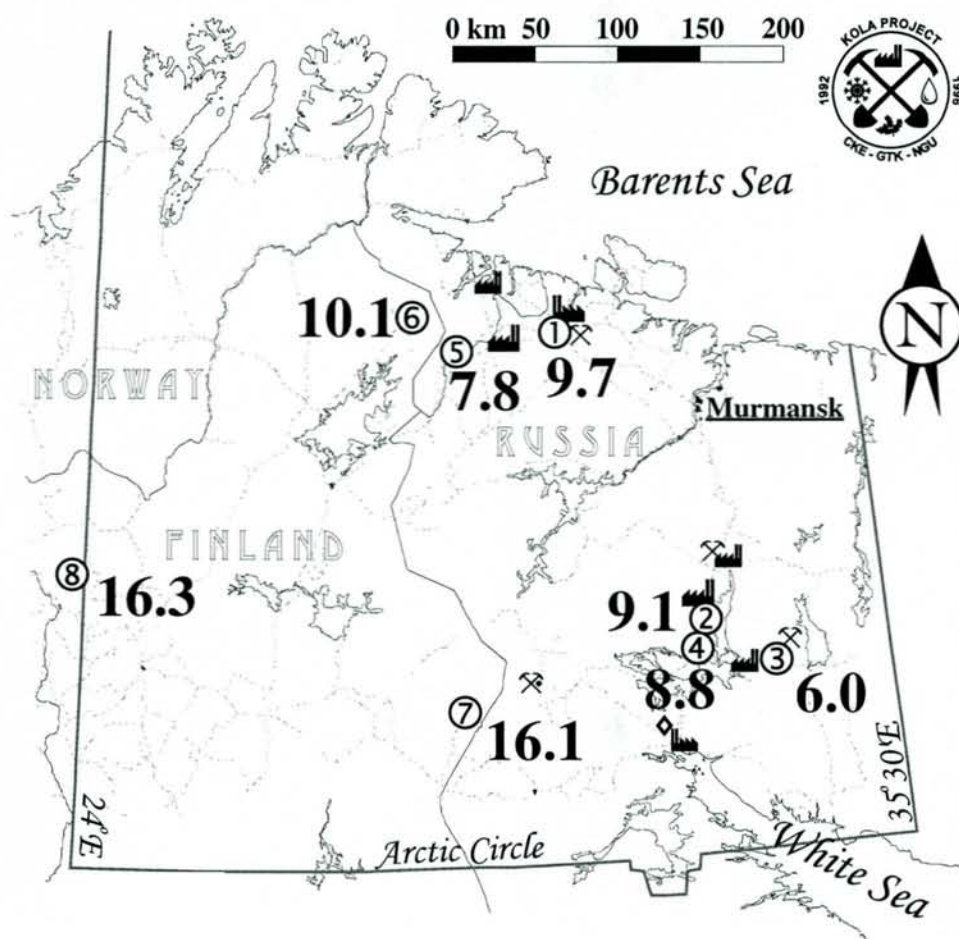


Figure 4.3.4. Map of catchment-averaged sulphur isotopic composition of stream water.

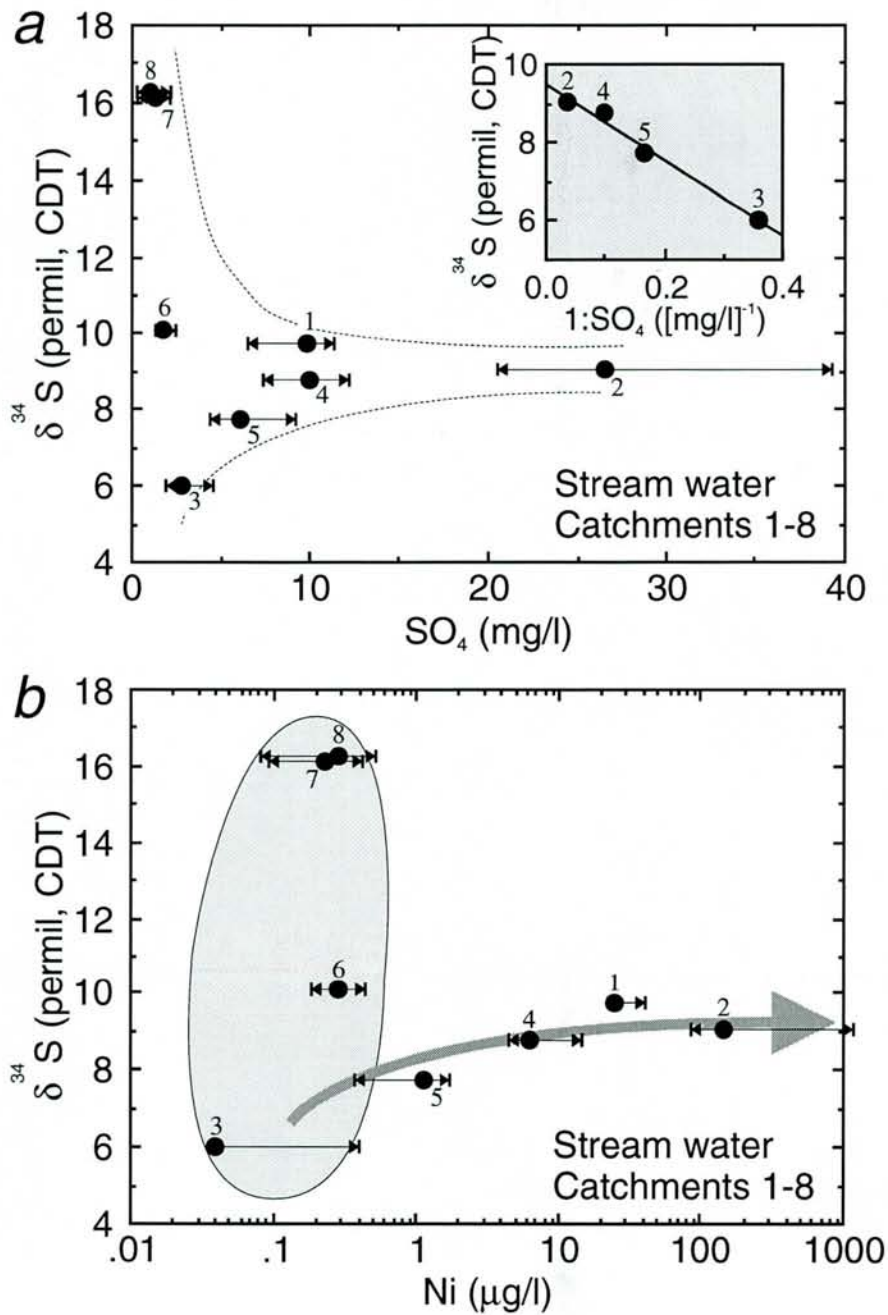


Figure 4.3.5.  $\delta^{34}\text{S}$  composition vs.  $\text{SO}_4$  content (a) and vs. Ni content (b) for stream water. Numbers 1 to 8 refer to catchment numbers. The horizontal, double-arrowheaded lines represent the range of  $\text{SO}_4$  or Ni concentrations recorded for each catchment during the whole monitoring period (March-November 1994, Caritat et al., in prep. a,b). The errors of each measurement are equal to or smaller than the symbol size.



# Moss $\delta^{34}\text{S}$ (‰, CDT)

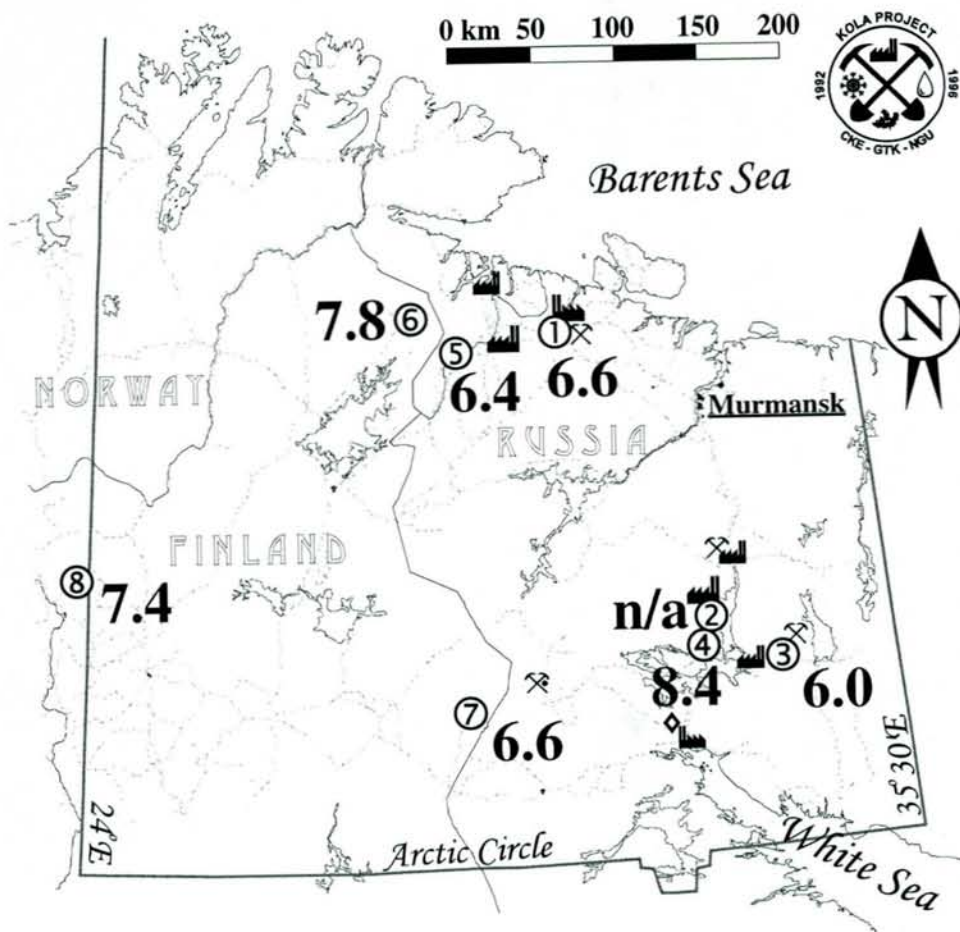


Figure 4.3.6. Map of catchment-averaged sulphur isotopic composition of moss. No moss was found in Catchment 2 (n/a: not applicable).

# Humus $\delta^{34}\text{S}$ (‰, CDT)

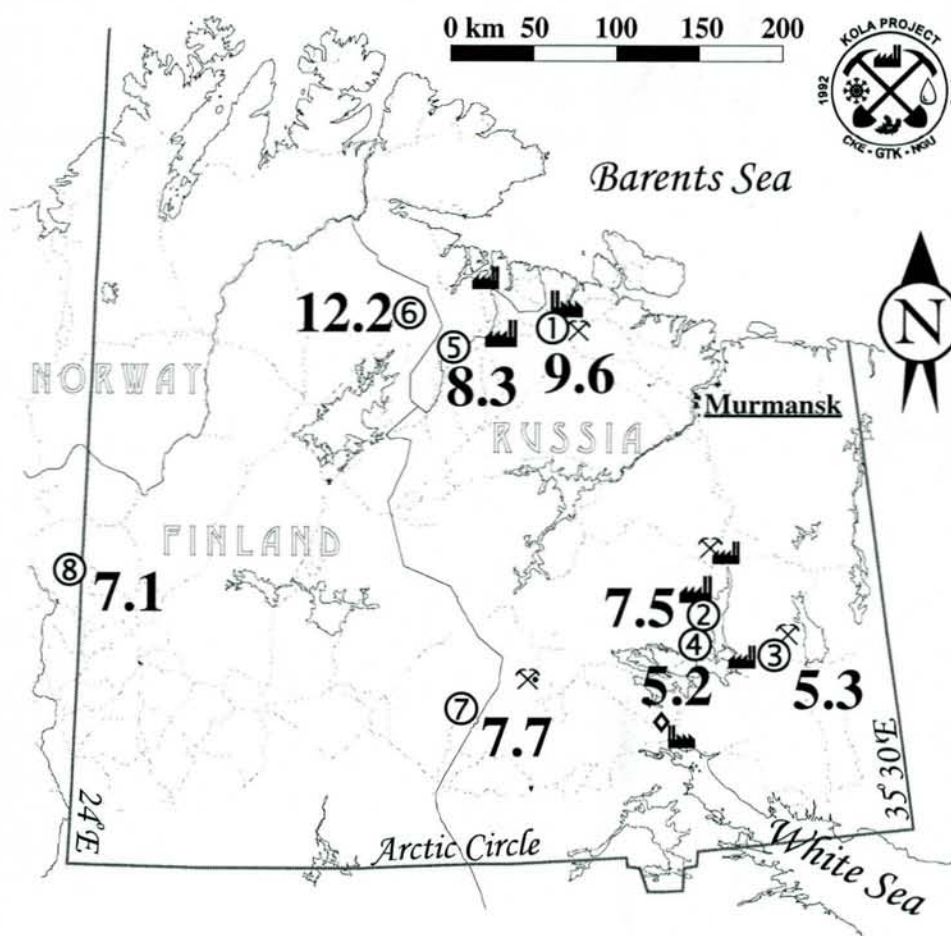


Figure 4.3.7. Map of catchment-averaged sulphur isotopic composition of humus.

## 5 STREAM SEDIMENTS

### 5. 1. Organic stream sediments as environmental indicators in eight intensively studied catchments in the Barents region.

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#### ABSTRACT

The chemical composition of organic stream sediments reflects both industrial pollution and the geological features in the areas under investigation in the Barents region. This is a drawback of this sampling medium in areas with low or intermediate levels of contamination. The opposite situation occurs in areas of heavy atmospheric deposition of heavy metals, where they are useless for geochemical prospecting. Organic stream sediments are highly representative, even though samples were taken three times during the season from the same location within each catchment. A documented significant variation in the stream water chemistry, sampled simultaneously from the same catchments, shows that organic stream sediments are suitable as an environmental indicator, giving an integrated picture of the chemical status for a sampled part of the river. Organic stream sediments ("mud") possess moderately high absorption capacity, and reliable results are found even for trace elements like Ag, Be, Bi, Se, Th, Tl and U, that are barely detected in other sampling media collected simultaneously from the same catchments. This paragraph is very hard to understand!

#### INTRODUCTION

The Geological Surveys of Finland (GSF) and Norway (NGU) and the Central Kola Expedition (CKE) in Russia are carrying out a major ecogeochemical project in the West Murmansk region and contiguous areas in Northern Norway and Finland (Fig. 5.1.1). As a preparatory phase of this mapping project, eight small catchments (hereafter called C1-C8), situated at different distances from major contamination sources in the Murmansk region, were studied in detail during 1994. The following media were sampled, at least once and some several times, within each catchment: snow cover, rainfall, streamwater, organic stream sediments, moss, topsoil, podzol profiles, Quaternary deposits and bedrock.

The use of aquatic sampling media rich in organic matter has gained popularity in environmental investigations. Regional sampling of Northern Fennoscandia was done in the collaborative Nordkalott project in 1980 - 1986, based on, among other media, stream organic matter (humic material, plant roots, leaf litter etc. from the stream bank) and stream moss samples (mixture of various species), which illustrated the regional patterns of 25 and 14 elements respectively throughout the area (Bølviken et al., 1986; Steenfelt, 1993). In 1990, the GSF conducted a nation-wide reconnaissance scale geochemical mapping project by using a new sampling method for organic stream sediments. Stream sediments were sampled with a scoop net at a mean density of one composite sample per 300 km<sup>2</sup> (Lahermo et al., 1995). Preliminary results of the regional data have been published by Lahermo & Väänänen (1993) and Lahermo et al. (1995). An atlas including data from the sediments and stream water is in preparation.

Organic stream sediments, when sampled with a scoop net, contain mineral material and varying amounts of decomposed organic particles, deposited recently (during the last 1-5 years) on the river bed (Lahermo et al. 1995), and can popularly be called "mud". The samples in this study were collected by stirring the bottom sediments with a scoop net. This method of sampling gives samples richer in organic and fine-grained minerogenic materials than a sample collected in the hand or with a spade or cup, and this is true even in catchments with a low rate of sedimentation of organic matter. These organic rich sampling medium make it possible to analyse even low concentrations of rare elements (Lahermo et al. 1995). In Finland, the total carbon concentration of the stream sediments varies from 1 to 30%, on average 10 % (Lahermo et al. 1995). According to Lahermo & Väänänen (1993), sediments rich in organic matter commonly exist in those catchments where the proportion of peat lands to surface area is greatest.

The composition of stream sediments reflects both local bedrock geochemistry and the distribution of elements in overburden (Lahermo et al. 1995). Since the organic-rich sediments are influenced by a combination of sources like the composition of groundwater, viz. water filtered through the soil, stream water and materials dispersed in rain water (atmospheric contamination), they reflect changes in the state of the terrestrial environment as suggested by Sipilä and Salminen (1995). Sipilä and Salminen (1995) report of contaminated organic sediments downstream from tailings areas, even several kilometres away from mining areas.

In this paper, we will focus on the results of organic stream sediments sampled from eight selected catchments. The aim of this study is to evaluate the use of this sampling medium for regional environmental investigations, as an indicator of anthropogenic impact versus natural geochemical background levels (prospecting). In addition, this study will evaluate seasonal variation, and the importance of varying proportions of organic content in the samples.

## MATERIALS AND METHODS

### *Sampling*

The sampling method used in our study is almost the same as used in the nation-wide mapping of Finland by use of organic stream sediments (Lahermo et al. 1995). After suspension of the sediments from the river bed, samples were collected by using a scoop net with mesh diameter of 0.06 mm. The coarser material (>1 mm) was excluded by sieving the sample through a screen cup with 1 mm holes into a pail. In order to avoid collecting the heavy metal detritus, the light organic sample was carefully swirled back through the sampling net (Räisänen et al., 1992). The extra water from the net was gently wrung out. According to Räisänen et al. (1992), the loss of clay-size particles is <0.001%, and the final grain size of the sample is then <1 mm.

Organic stream sediments were sampled three times during 1994 (in spring, summer, and autumn) at five stations per catchment in C1-C5, and at three stations per catchment in the Finnish areas (C6-C8). The sampling stations were located in a regular pattern within each catchment, i.e. one in the main stream near the outlet and two to four in tributaries. The composite sample was made from 3-5 subsamples taken at 20-50m distances from each other. A more comprehensive description of the field procedures is given by Äyräs and Reimann (1995).

### *Analysis*

All the organic stream sediment samples were analysed by GSF in Rovaniemi. The following method of preparation was used for stream sediment samples: drying at temperatures below 40°C, reduction with a Moulinex household food processor, and dissolution with nitric acid in microwave oven. Multielement determination was made by ICP-MS (20 elements), ICP-AES (16 elements) and HAAS techniques (determination of Hg) (Niskavaara, 1995). The loss of organic ignition (LOI) was determined by NGU, by heating ca. 1.5 gram of dry sample at 430 °C for 20 hours.

### CHARACTERISTICS OF THE CATCHMENTS

Tables 5.1.1a and 5.1.1b give an overview of various features of the studied catchments.

Bedrock, Quaternary deposits and topography are described in detail by Pavlov et al. (1996).

C3 represents a special environment due to its alkaline bedrock. The rest of the catchments can be grouped, on the basis of the main bedrock type, as follows:

Ultra mafic            - mafic            - acid bedrock

C3 >> C4 = C2 > C5 > C7 > C1 > C8 = C6.

It is worth noting that the overburden in C1 contains a markedly high concentration of Mg and Fe, and to some extent Ca, in stark contrast to what is expected due to the predominantly granitic and granodioritic rocks (Pavlov et al. 1996). Similar contrasts also exist in C6-C8 which are dominated by acidic rocks, where the till contains moderately high concentrations of Ca, Mg, Co or Ni, which commonly characterise tills on mafic bedrock (Table 5.1.1b).

Furthermore, high contributions of S and several heavy metals like Fe, Zn, Ni, Cu, Co and Cd occur in C5 due to the occurrence of black schists (20% of the bedrock).

Juve (1995) reports of uneconomical Ni-Cu sulphide mineralization in gabbro-wherlites in C5. In C2, the Cu-Ni mineralization occurs in pyroxenites (V. Pavlov pers. comm. 1996). There are no reports on mineralization in the rest of the catchments.

The variable nature (climate, topography, vegetation etc.) of the catchments causes differences in the production of organic materials entering the river systems. On the basis of field observations, we can presume that catchments C1, C2, C6 represent areas of low organic carbon productivity. C2 is mostly barren due to erosion and high levels of pollution. Parts of C2 and C6 are open tundra with arid vegetation species such as lichen. C6 in particular is influenced by erosion caused by overgrazing by reindeer.

### RESULTS AND DISCUSSION

#### *Element distribution*

The mean concentrations of elements in organic stream sediments are presented in Table 5.1.2. Box plot comparisons in Fig. 5.1.2 a-b present distributions of selected elements in the catchments. Very few detection limit problems occur in the dataset, and only B and Sb were under the detection limit for more than 50% of the analyses, while Ag had 3 such samples, and Se one, and all the other elements were above the detection limits. No other sampling media in our study have accumulation potential as high as the organic stream sediments, and detection limits occurred more frequently for the other media.

Table 5.1.2 shows a distinct contrast in heavy metal concentrations between the studied areas. High levels of As, Se and Hg were found in the Russian catchments (C1, C2 and C4), which are situated between 5 and 25 km from the Cu-Ni smelters, compared to the other catchments.

The sediment samples contain 100 times more Cu and Ni in C1 and C2 than in the background catchment (C8). In addition, sediments in C2 were characterised by abnormally high concentrations of Ag, As, Bi, Co, Cr and Se in particular (Table 5.1.2 and Figs. 5.1.2a-b for Ag, As and Se). Concentrations of Cd (Fig. 5.1.2a) and Zn were highest in C4. The highest concentrations of Hg and Tl occurred in sediments from C1, but sediments in C4 also revealed elevated concentrations of Tl (Fig. 5.1.2b). Most of these elements are known to belong to the emission spectrum of the Russian mining and smelting industry (Äyräs et al., 1995, Chekushin et al., 1996).

Elements such as Al, Be, K, La, Na, Sr and Rb are typical for alkaline rocks of the Hibiny massive, and characterise C3 (Figs. 5.1.2a-b for Be and Rb). As seen in Table 5.1.2, concentrations of Al and Be reached the second greatest level in the granitic environment of C6. Sediments in C6 revealed also high concentrations of Th, U and Mo (Figs. 5.1.2a-b). According to Kesola (1991), moderately high concentrations of Th, U and Mo are typical for the Vainospää granite. Similarly, moderate concentrations of Mo in C1 may have originate from granite intrusives (R. Kesola, pers. comm. 1996). Abnormally high concentrations of U also occurred in some of the organic sediments from C8. This indicates a mineralization of unknown origin in the bedrock in this area (E. Vanhanen, pers. comm. 1996, see also Inkinen 1979).

Amphibolite layers in gneisses from C7 led to slightly increased concentrations of Cr, Co and Fe in the organic stream sediments. Carbonaceous sedimentary rocks are an apparent source of Ca and Mg in sediments of C5. Moreover, sediments in C5 contain more Cu, Ni, As and S than the Finnish catchments. In this case, the source may be either black schists and/or emissions from the Ni-Cu smelter in Nikel. Similarly, the high concentrations of Mg and Ca in C1 could be related to the greenstone rocks derived from the overburden of the Pasvik-Pechenga greenstone belt. This suggestion is supported by the fact that greenstone boulders occur in tills in C1. However, contamination could possibly be part of the explanation here too.

#### *Element variability*

The influence of three different factors on the total variability of element concentrations in the organic stream sediments, are estimated by use of one-way ANOVA algorithm calculation (Krumbein & Graybill 1965) available in the STATGRAFICS program. ANOVA analysis is conducted to estimate the influence of the three main factors on the variability of the element content in the sediment samples, namely between catchments (R1), between sampling points within each catchment (R2) and finally seasonal variation (R3), as shown in Fig. 5.1.3.

Concentrations of many trace elements, such as Ni, Cr, Co, Cu, Ag, Sr, La, Y and Th, varied most from catchment to catchment (more than 80% of total variation explained by R1), whereas concentrations of Ca, K, Al, Mg, Ti and S varied somewhat less (about 60% of total variation explained by R1). The wide variation in Ca, K, Al, Mg and particularly Ti within each catchment (R2), can partly be explained by differences in the amount of minerogenic materials of sediments for several of the catchments, by comparing to element values to the LOI-values. Due to the weak extraction power of the concentrated nitric acid on the main silicates (quartz, feldspars, amphiboles), the amount of the major elements mainly reflects the abundance of trioctahedral micas, Mg-bearing clays and precipitates (M. L. Räisänen, unpublished stream sediment data, see also Dole et al., 1968).

The lower total variation in the concentrations of As, Zn, Cd, Se, Tl, Bi and Hg led to higher values for factor R2, which also indicates a strong variation in the sampling points within the catchment. Concentrations of Cu, Cr and Ni show the lowest variation within each catchment.

In general, seasonal variation has a negligible effect on chemical variations in the stream organic sediments. Similar low time-dependent concentrations were found in vegetation samples from the same areas and period (Bogatyrev et al, 1996). The highest seasonal variation in the three sampling sets occurred in concentrations of Ca, Tl, Zn, Ti, S and B, but at most 2.37 % of the variation is explained by the seasonal factor (see R3 in Figure 5.1.3). The similarly small seasonal variation in the trace element chemistry of organic stream sediments has also been reported by Chork (1976) and Lestinen (1979). We can conclude that, for most of the studied elements, the repeated seasonal sampling of the same localities within each catchment reveals an excellent representativity, actually as satisfying as a field duplicate should be. This indicates that this aquatic medium possesses superior accumulation properties, and reflects an integrated picture of the chemical status of the river. This organic-rich sediments do not respond to the episodic changes in the stream water chemistry, as revealed by Caritat et al. (1996)

The dual origin of the elements in organic stream sediments is clearly seen from our dataset. The variability of element concentrations can be linked to both industrial pollution and geological characteristics of the studied catchments. The main ore elements, Ni, Co, Cu and Cr, and the accompanying trace elements, Ag, As, Zn, Cd, Se, Tl, Bi and Hg, are typical for the catchments C1, C2 and C4, and any of these elements could be associated with the bedrock of C2 and C4, and overburden in C1 (Table 5.1.1b). The highest concentrations of these elements occurred in sediments in C2 and C1, closest to pollution sources, where the influence of the geogenic factor is hard to distinguish from the rate of contamination due to the heavy depositions of the main ore elements (Chekushin et al., 1996). Table 5.1.2 shows a very wide range of Ni, Cu, Co, Cr and As concentrations, which have a high positive relationship with distances from the smelters. This finding nevertheless indicates the strong contamination of these elements in stream sediments (Sipilä & Salminen 1995).

#### LOI

The loss of organic ignition (LOI) reveal a huge total variation, from 7.9 to 61.9 % (37.5 % in average) for all of the 102 collected samples (Tab. 5.1.2). C3 and C5 in particular have the lowest organic content, but also C8 is low, while the rest of the catchments do not show any significant differences. The highest correlation exists between LOI vs. S ( $r=0.76$ ) and vs. Hg ( $r=0.74$ ). Negative correlations exists between LOI and many other elements like Ti ( $r=-0.68$ ), K, Na, Rb and Sr ( $r=-0.44 - 0.54$ ). The last group of elements probably shows the importance of minerogenic input into the material. As mentioned earlier, the variation in LOI explain the high internal variation of certain elements within the catchments, e. g. Ca and Se in C5. However, for the most dominating pollutants (Cu, Ni), the variation between the catchments is so extreme that the differences in LOI do not have a significant effect.

#### *Anthropogenic contamination versus geogenic sources*

VARIMAX-rotated factor analysis was used here to distinguish different sources in the chemical composition of the organic stream sediments, and the main factors are presented in Table 5.1.3, while XY-comparisons of selected factors are shown in Figure 5.1.4. The factor analysis describes the geochemical peculiarities of each catchment, including influence of the three main features: contamination, local geology and conditions of sedimentation (presence of hydroxides of Fe and Mn). Factor 1 (contamination) explains most of the trace elements

known to originate from the emissions of the Russian mining and smelting industry in the Kola Peninsula (Äyräs et al., 1995; Chekushin et al., 1996, Reimann et al., 1996). Negative coefficients in F1 show exceptional behaviour of the trace elements in some of the subsets, and therefore the factor is called the contamination factor.

Factor 2 (positive), called the alkaline factor, consists of the elements typical of alkaline bedrocks. Factor 3 is called the hydromorphic factor (commonly also called the Mn factor), and contains trace elements which commonly are associated with secondary Mn- and/or Fe-precipitates. In this factor, however, the coefficient for Zn is negative, which suggests an unusual behaviour of Zn in the sediments. In the natural, undisturbed environment, most Zn occurs in Mn precipitates, and in polluted sediments the behaviour of Zn is independent of the occurrence of Mn (Salminen 1979a).

The low value of the contamination factor (F1) in C4 and particularly C1, which are situated close to the smelters (see Tab. 5.1.1a), is explained by the strong impact of other factors. In C4, the hydromorphic factor (F3) is strong, and in C1 both the Mo-Hg factor (F4) and the hydromorphic factor drag down the effects of the F1 factor in these contaminated catchments.

Factor 4 (Mo-Hg factor) and factor 5 (U-Th factor) have low loadings and are therefore less important. The element distribution in these factors reflects a specific granitic environment (see Table 5.1.1b). However, occurrence of Hg together with Mo, S and B with negative loadings may indicate that in some cases the source of these elements is rather anthropogenic than geogenic.

Fig. 5.1.4 shows the distribution of the organic stream sediments of eight catchments in the factors. Factor 1 characterises solely the behaviour of trace elements in stream sediments of C2 (Monchegorsk). In C4, but not in C1 (Zapoljarnij), the behaviour of trace elements possesses some similarities to those in C2 (see Fig. 5.1.4a). This indicates that F1 depicts not only the anthropogenic contamination of the sediments, but also the geogenic influence of mafic and ultramafic rocks. Evidently, the strong influence of mafic and ultramafic rocks in C2 and C4 explains why sediments in C1, regardless of the metal deposition, did not enter the contaminated factor (Chekushin et al. 1996).

Overall, it can be concluded that the substantial difference in loadings in F1 between sediments of C2 and the other catchments (Figs. 5.1.4a-b) suggest that the pollution in C2 is exceptionally strong compared with that in the other catchments. According to Äyräs et al. (1995), the element spectrum in the anthropogenic deposition and therefore also in contaminated sediments in C2 is wider than in C4 and C1. On the other hand, severe vegetation damage in C2 may cause differences in the way trace elements enter into stream sediments.

As expected, the geochemical properties of sediments in C3 and C6 (with lower loadings) entered the alkaline factor because of the geological finger print of this catchments (F2 in Fig. 5.1.4c). The hydromorphic factor (F3 in Fig. 5.1.4c) characterises the element behaviour in C4 and to some extent in C1. Here co-precipitation with Mn and Fe controls the behaviour of mobile traces such as Cd. Unlike the other traces, Zn had a negative coefficient, which indicates an anthropogenic source in C4 and C1 (Äyräs et al. 1995).

Factors 4 and 5 are associated with the geochemical properties of the sediments in C1 and C6, indicating the influence of a granitic environment (Table 5.1.1b). According to R. Kesola (pers. comm. 1996), Mo in Kirakka and evidently also in Zapoljarnij is associated with granite intru-



sives. However, F4 may also influence the contamination of Mo, Hg and S in sediments, particularly in C1 (Reimann et al. 1996).

#### *Comparisons to other sampled media*

By comparing the median values of selected other sampling media (moss, stream water, topsoil and Quaternary deposits) from the same catchments, several co-variations with the chemistry of the organic stream sediments can be seen. In general, concerning the main pollutants (Co, Cu and Ni) in media affected by the atmospheric environment, the variations between the catchments are much more extreme than the variation between the concentrations in bedrock and Quaternary deposits.

In the XY- diagram of Figure 5.1. 5, the concentrations of Cu in the organic stream sediments show the best relationship to the respective topsoil samples, but co-variations exist also with moss, stream water and to an extent Quaternary deposits. The order of the catchments (increasing median concentrations in the organic stream sediments) follows the distance to the main smelters.

## CONCLUSIONS

1. In less polluted catchments (C3, C6, C7 and C8), the element composition of organic stream sediments mostly reflects the geochemistry of tills and rocks. This conclusion is in agreement with earlier investigations (Salminen 1979b, Bølviken et al. 1986, Lahermo et al. 1995). It can be concluded that organic stream sediment is an appropriate sampling medium for prospecting in non-polluted areas.
2. Compared to the other catchments, abnormally high concentrations of Cu, Ni, Cr, As, Se, Ag and Bi in C2 suggest a high rate of contamination. In addition to Cu, Ni, Cr and Co, Hg, Tl and Mo were more concentrated in sediments in C1, and Mn, Zn, Ba, Cd and Tl in those in C4 than in the Norwegian and Finnish catchments, indicating contamination of sediments in C1 and C4. These suggestions are compatible with the element spectrum of the deposition in industrial areas of Monchegorsk, Nikel and Zapoljarnij.
3. Organic stream sediments possess very high absorption capacity, and reliable results are found for many trace elements e. g. Ag, Be, Bi, Se, Th, Tl and U, which are barely detectable in other sampling media collected simultaneously from the same catchments.
4. Negligible seasonal variations are observed, indicating that this aquatic sampling medium to be very representative for a given stretch of the river bed.
5. Finally, we conclude that the chemical composition of organic stream sediments reflects both anthropogenic contamination and geogenic factors of the catchment. By using factor analysis, it was possible to distinguish the strong pollution factor from the geogenic factors. However, moderate or low contamination levels were only weakly seen in the factor analysis.

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## TABLES

Table 5.1.1a. Description and location of the catchments studied, including I) distance to major industry, II) vegetation types, III) stream network and IV) topographical features.

Table 5.1.1b. Continued description of the catchments including V) bedrock, VI) Quaternary deposits and VII) geochemical characteristics of the local till and bedrock, after Pavlov et al. (1996).

Table 5.1.2. Mean concentrations of elements and the loss of organic ignition (LOI) in organic stream sediments from eight catchments in the Barents region (C1 - C8). Number of samples taken from each catchments shown in brackets.

Table 5.1.3. Principal factor analysis (SMC) with rotation VARIMAX of the organic stream sediments data (n=102) showing five factors. All 36 elements were included in this factor analysis.

## FIGURES

Figure 5.1.1. Location of the study area for the regional mapping project (frame) and the location of the catchments studied in 1994.

Figure 5.1.2. Box plot comparisons of a) Ag, As, Be, Cd, Hg, Mo and b) Rb, S, Se, Th, Tl and U in organic stream sediments. All concentrations in  $\mu\text{g}/\text{kg}$  (ppm).

Figure 5.1.3. Variability analysis (ANOVA) illustrated as estimates of the influence of three factors (R1-R3) for the analysed elements in organic stream sediments sampled in three periods in 1994 from eight catchments in the Barents region.

Figure 5.1.4. X-Y plots of selected factors from the principal factor analysis (SMC) with VARIMAX rotation; including the factors: Contamination factor (F1), Alkaline factor (F2), Hydromorphic factor (F3), Mo-Hg factor (F4) and U-Th factor (F5). C1 - C8 refer to the catchment numbers.

Figure 5.1.5. XY- diagram of the median concentrations of Cu in the Organic stream sediments (X-axis) from the eight catchments versus on the Y-axis the median concentration of Cu in stream water, moss and topsoil (aqua regia extractions) from the same catchments. All values in ppm.

No	Name	Coordinates - Size outlet (km <sup>2</sup> )	Altitudes (m.a.s.l.)	Annual precip (mm)*	Distance and position to nearest major industry (emission sources)	Vegetation	Stream network	Topography	
C1 (Ru)	Zapoljarnyi	69°27' N 31°03' E	19	25 - 373	454	10 km NE of Zapoljarnij (roasting), 30 km W of Nikel (Ni - Cu smelter)	Tundra forest (birch)	Dense	Undulating
C2 (Ru)	Monchegorsk	67°50' N 32°54' E	22.4	128 - 507	391	5 km S of Severonikel (Ni - Cu smelter)	"Mostly barren" (originally boreal conifer forest: spruce & birch)	Dense	Undulating
C3 (Ru)	Kirovsk	67°32' N 33°48' E	20	240 - 1075	502	2 km S of Kirovsk (ap-ne mines) 5 - 15 km N and W of several ap-ne	Mountain: birch>> spruce	Middle	Rough
C4 (Ru)	Kurka	67°41' N 32°50' E	20.5	152 - 466	502	25 km SW of Severonikel 20 km W of Apatity (ap-ne)	Boreal conifer forest: spruce>birch	Middle	Undulating
C5 (N)	Skjellbekken	69°21' N 29°27' E	34.6	80 - 297	422	30 km W of Nikel 50 km SW of Kirkenes (Fe-smelter)	Boreal conifer forest: Pine>> spruce & birch	Dense, many lakes	Smother
C6 (Fin)	Kirakka	69°35' N 28°52' E	11.9	110 - 200	386	60 km NW of Nikel 50 km W of Kirkenes	Tundra forest (pine & birch)	Dense, with some small lakes	Smother
C7 (Fin)	Naruska	67°21' N 29°22' E	20.2	263 - 490	513	65 km SW of Kovdor (Fe - ap-ne) 150 km W of Kandalaksha (Al-plant)	Boreal conifer forest: spruce & pine > birch	Middle	Rough
C8 (Fin)	Pallas	68°09' N 23°52' E	24.4	303 - 500	405	150 km W of Kiruna (Fe-mines) 290 km SW of Nikel	Boreal conifer forest: spruce & pine	Middle	Undulating

\* from the closest meteorological station (data from 1994)

Table 5.1.1a. Description and location of the catchments studied, including I) distance to major industry, II) vegetation types, III) stream network and IV) topographical features.

Catchment (No.)	Bedrock	Quaternary Deposits	Geochemical characteristics of rocks and tills (Pavlov et al. 1996)
Zapoljarnij (C1)	amphibole-biotite gneiss >> granites	basal till (60%) >> meltout till (26%) >> eluvial dep. (7%) >> rock terrains, water	Moderately high conc. of Mg, Ca, S, Cu, Cr and Ti in tills, low in rocks
Monchegorsk (C2)	andesitic effusives, tuffs > gabbros, gabbro-norite > pyroxenite	basal till (76%) >> glaciofluvial sed. (9%) > eluvial dep. (8%) >> glacio-lacustrine sed. (4%) > water, lacustrine sed., rock terrain	High conc. of Ni, Cr, S, Cu, Fe, Mg, Zn in rocks, moderately in tills
Kirovsk (C3)	Trachytoid>hibinite >> gabbro-norite > metabasalts & meta-andesites > black schists, mafic tuffs	basal till (57%) > deluvial - eluvial dep. (41.4%) >>rock terrain	High conc. of Al, Na, Sr, Y, K, Ba, As, Mo and S in rocks and lake sediments, less in tills
Kurka (C4)	amphibolite > two-mica gneiss > gabbro norite > gabbro-amphibolite	basal till (54%) > peat (30%) >> glaciofluvial sed. (6%) > lacustrine sed. (5%)> rock terrain, water	Moderately high conc. of Fe, Ni, Cr, Ba, Zn, V, S, Cu in rocks, less in tills
Skjellbekken (C5)	andesites, andesitic volcanoclastic schists (40%) > tholeiitic basalts & tuffs (30%) > black schists (20%) > others	basal till (59.2%) >> glaciofluvial sed. (17%) > meltout till (12.4%) >> rock terrain, water	Moderately high conc. of Mg, Ca, S, Fe, Cu, Ni and V in black schists; high conc. S, As and Cu in tills
Kirakka (C6)	granite (100%)	rock terrain, block fields (60%) > meltout till (34%) > water	High conc. of Th, La, Ba, Pb, Zn and Mo in granite (Kesola 1991), less in tills
Naruska (C7)	tonalitic gneiss (mica gneiss, amphibolite interlayers)	basal till (63%) >> peat (22%) > rock terrain, block fields (14%) > water	Moderately high conc. of Mg, Co and S in tills, less in rocks
Pallas (C8)	quartzite >> amphibolites	basal till (69%) >> peat (29%) >> rock terrain (2%) > water	Moderately high conc. of Mg and Ni in tills, low in quartzites

Table 5.1.1b. Continued description of the catchments including V) bedrock, VI) Quaternary deposits and VII) geochemical characteristics of the local till and bedrock, after Pavlov et al. (1996).

Element	Unit	C1 (15)	C2 (15)	C3 (15)	C4 (15)	C5 (15)	C6 (9)	C7 (9)	C8 (9)
LOI	%	45.5	46.5	23	46.1	22.5	48	38.1	29.5
Al	g/kg	10.4	22.5	34.5	16.7	12.4	25.4	12.3	8.65
Ca	g/kg	8.2	8.6	10.1	10	10.9	4.47	7.37	5.76
Fe	g/kg	62.6	32	12.2	81.1	26.9	28.8	44.4	70
K	g/kg	1	1.27	3.47	1.3	1.33	1.06	1.6	0.8
Mg	g/kg	7.02	4.04	2.9	3.47	6.07	1.47	2.74	2.35
Na	g/kg	0.21	0.29	3.46	0.2	0.24	0.16	0.18	0.15
Ag	mg/kg	0.31	1.18	0.45	0.49	0.1	0.1	0.1	0.1
As	mg/kg	22.7	45.4	6.73	18.1	17	4.19	3.22	6.93
B	mg/kg	7.53	1.67	2.8	4.53	3.17	4.17	2.56	1.5
Ba	mg/kg	109	106	283	416	129	48.3	96.7	246
Be	mg/kg	0.3	0.3	3.73	0.54	0.3	3.25	0.3	0.5
Bi	mg/kg	0.18	0.59	0.1	0.32	0.1	0.17	0.18	0.1
Cd	mg/kg	0.92	1.29	0.23	2.42	0.4	0.34	0.28	0.3
Co	mg/kg	195	228	7.43	250	15	10.6	27.1	20
Cr	mg/kg	84.6	149	21.5	70.4	32.5	14.4	56.4	23.5
Cu	mg/kg	698	4405	41.4	569	54.2	31.1	8.04	5.99
Hg	mg/kg	0.26	0.18	0.07	0.2	0.07	0.14	0.11	0.08
La	mg/kg	22.4	24	134	33.2	16.3	22.4	12.1	16.4
Mn	mg/kg	4635	1325	554	14597	1841	984	2337	2592
Mo	mg/kg	15.3	5.56	2.24	5.5	1.12	20.9	0.93	2.08
Ni	mg/kg	1170	2473	45.8	1016	39.6	18.5	16.1	10
P	mg/kg	1116	1646	2297	1401	823	1118	2183	2190
Pb	mg/kg	20.3	27.4	17	26.7	7.38	28.8	22	10.6
Rb	mg/kg	6.35	9.74	29.4	8.39	11.1	5.75	12.4	3.97
S	mg/kg	4635	3956	1209	2801	2432	3762	2340	1325
Sb	mg/kg	0.01	0.04	0.02	0.03	0.02	0.02	0.01	0.02
Sc	mg/kg	4.07	4.4	2.09	3.21	5.25	4.69	2.47	2.24
Se	mg/kg	5.89	9.51	3.08	5.59	3.04	3.76	2.77	1.17
Sr	mg/kg	46.9	38.8	789	46.7	33.6	29.1	67.1	29
Th	mg/kg	1.96	2.36	12.6	2.97	2.58	30.8	2.65	3.7
Ti	mg/kg	843	920	1393	966	1321	499	655	760
Tl	mg/kg	0.41	0.24	0.1	0.57	0.11	0.14	0.25	0.19
U	mg/kg	10.9	4.91	7.09	8.3	2.9	119	2.75	196
V	mg/kg	86	79.5	27.4	100	54.2	30.2	57.4	47.9
Y	mg/kg	11.1	10.7	32.9	15.1	10.2	26.6	3.97	7.97
Zn	mg/kg	107	93.8	97.1	292	94.7	65.4	40.7	56.9

Table 5.1.2. Mean concentrations of elements and the loss of organic ignition (LOI) in organic stream sediments from eight catchments in the Barents region (C1 - C8). Number of samples taken from each catchments shown in brackets.



Contamination factor (F1)		Alkaline factor (F2)		Hydromorphic factor (F3)		Mo-Hg factor (F4)		U-Th factor (F5)	
Cu	-0.967	Y	0.926	Mn	0.927	Mo	-0.792	U	0.568
Bi	-0.921	Al	0.916	Tl	0.844	Hg	-0.669	Th	0.547
Ni	-0.917	Be	0.91	Fe	0.794	S	-0.649	Mg	-0.693
Ag	-0.889	K	0.855	Cd	0.789	B	-0.539		
As	-0.839	Na	0.852	Ba	0.736	Li	0.67		
Se	-0.823	Rb	0.831	V	0.71	Ti	0.566		
Cr	-0.787	Sr	0.802	Co	0.681				
Sb	-0.704	La	0.794	Zn	-0.857				
Co	-0.625	Th	0.623						
Pb	-0.591	Ti	0.519						
(Cd)	(-0.466)								
(S)	(-0.444)								
Total variation	21.7 %		20.5 %		13.8 %		9.9 %		6.5 %

Table 5.1.3. Principal factor analysis (SMC) with rotation VARIMAX of the organic stream sediments data (n=102) showing five factors. All 36 elements were included in this factor analysis.

*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
Catchment locations

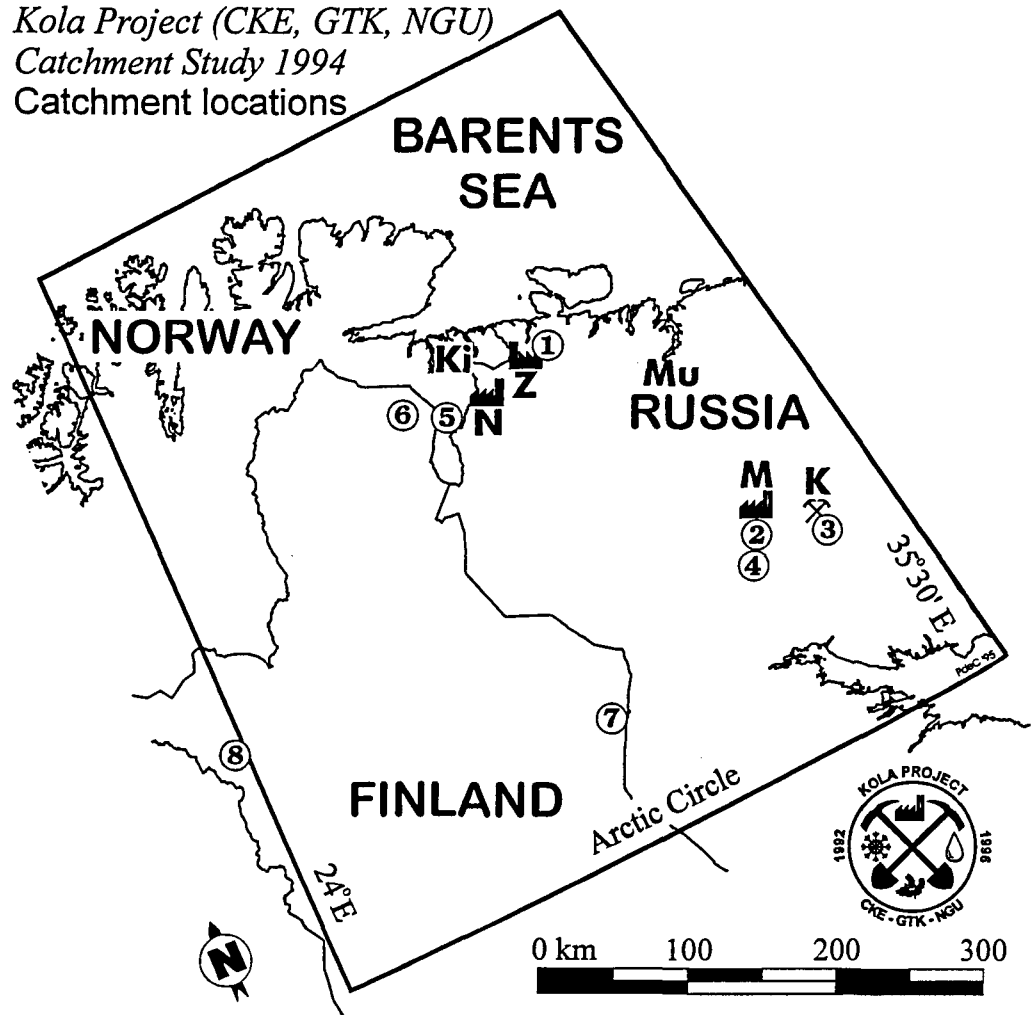
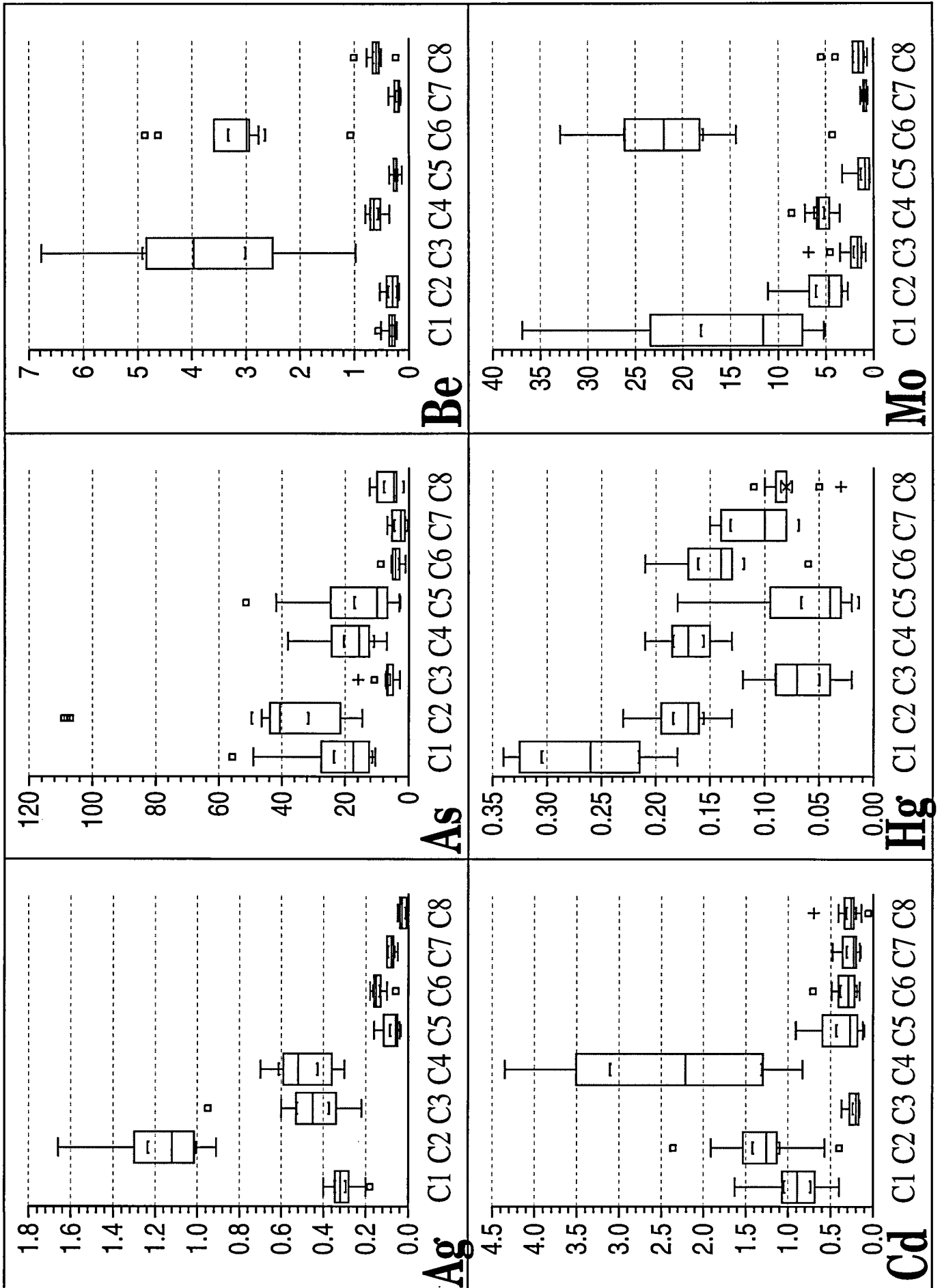
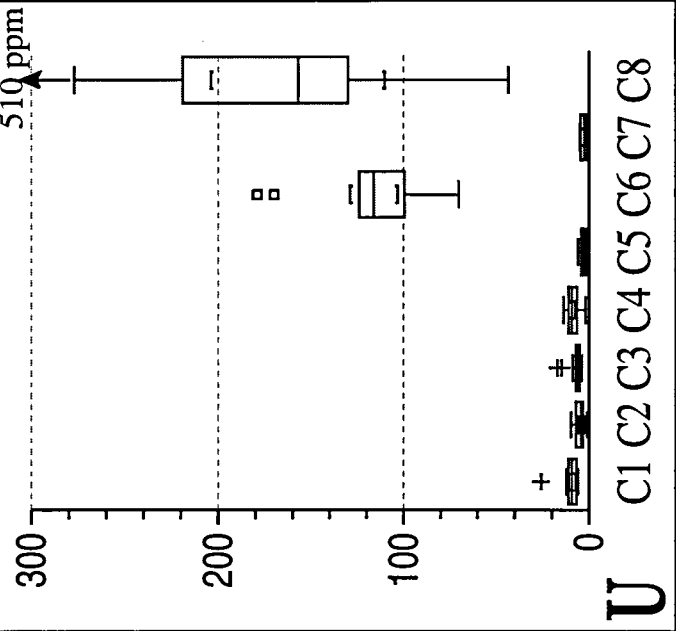
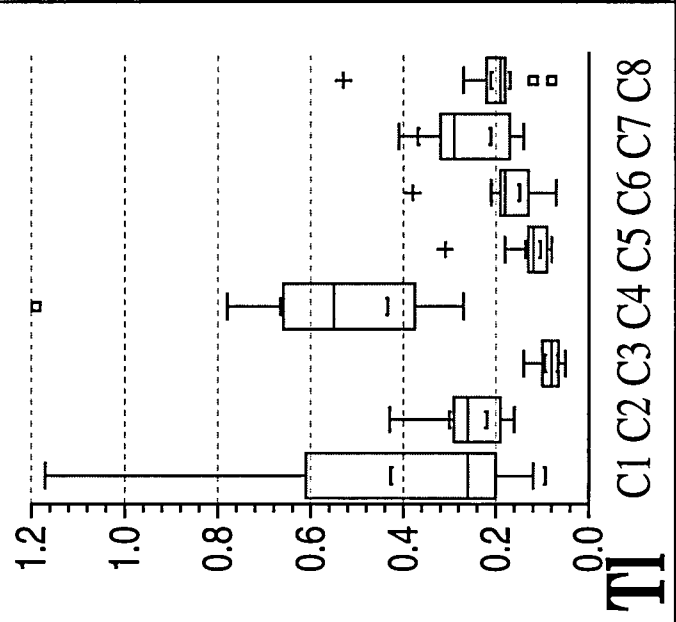
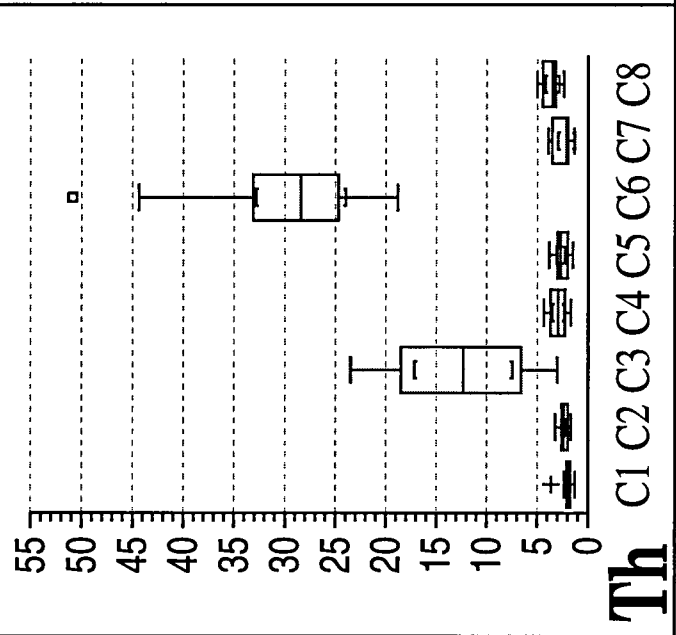
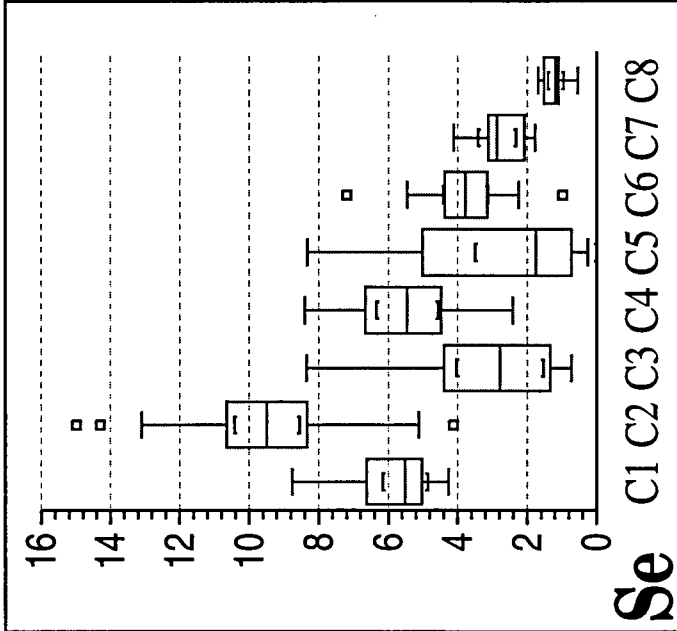
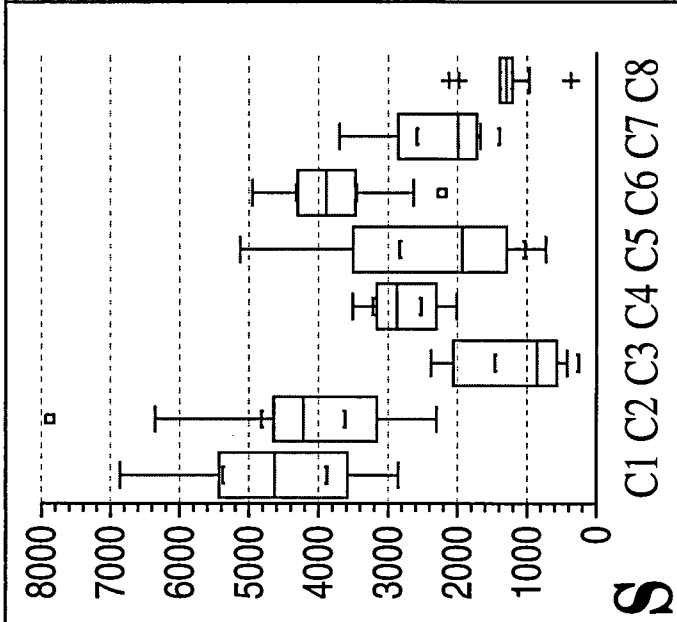
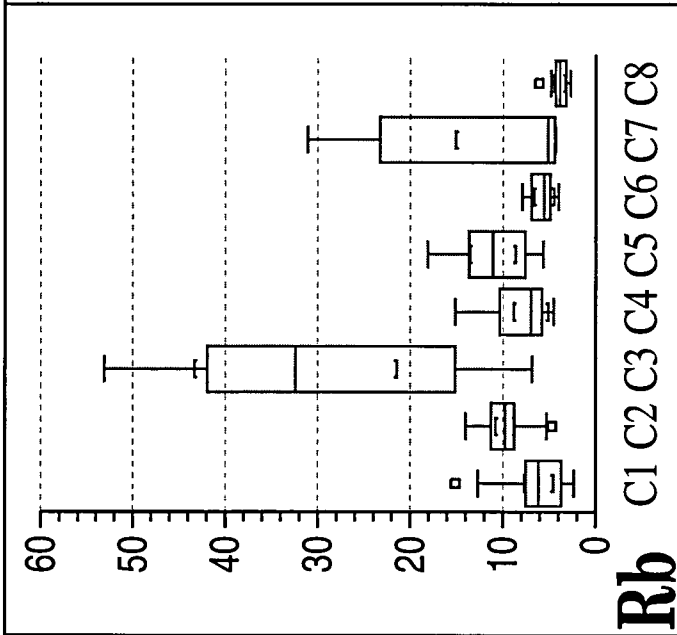


Figure 5.1.1. Location of the study area for the regional mapping project (frame) and the location of the catchments studied in 1994.

Figure 5.1.2. Box plot comparisons of a) Ag, As, Be, Cd, Hg, Mo and b) Rb, S, Se, Th, Tl and U in organic stream sediments. All concentrations in  $\mu\text{g}/\text{kg}$  (ppm).





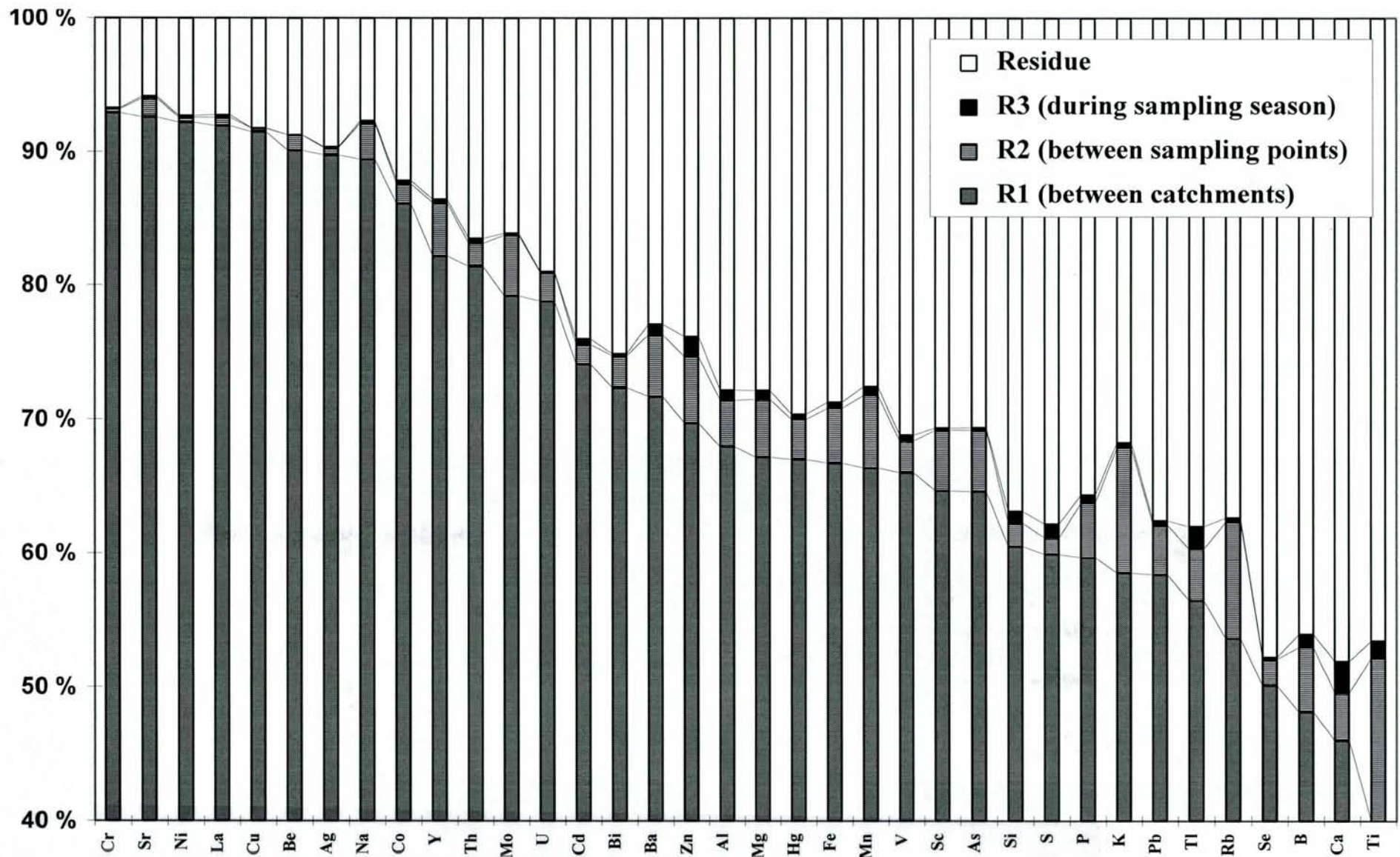


Figure 5.1.3. Variability analysis (ANOVA) illustrated as estimates of the influence of three factors (R1-R3) for the analysed elements in organic stream sediments sampled in three periods in 1994 from eight catchments in the Barents region.

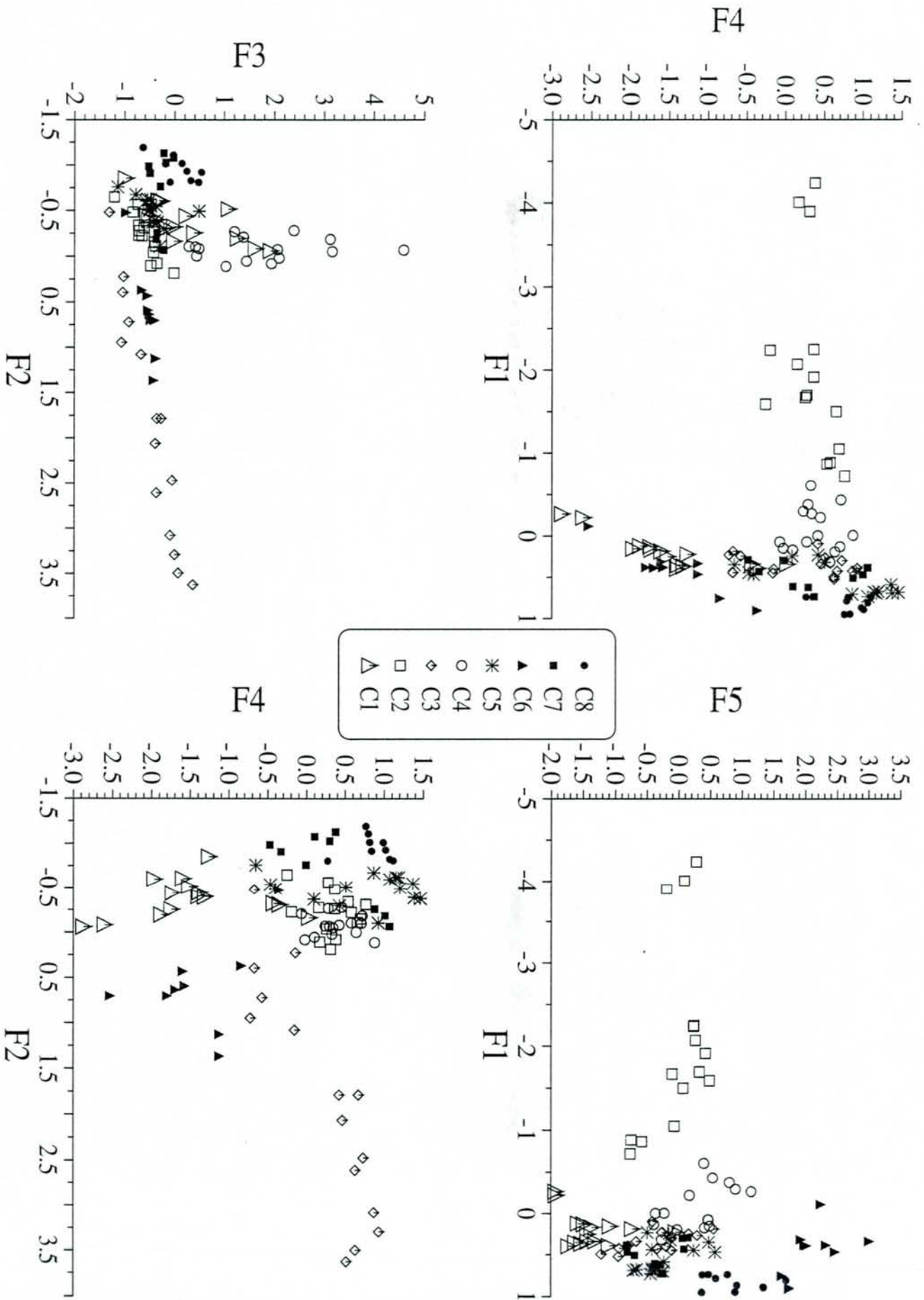


Figure 5.1.4. X-Y plots of selected factors from the principal factor analysis (SMC) with VARIMAX rotation; including the factors: Contamination factor (F1), Alkaline factor (F2), Hydromorphic factor (F3), Mo-Hg factor (F4) and U-Th factor (F5). C1 - C8 refer to the catchment numbers.

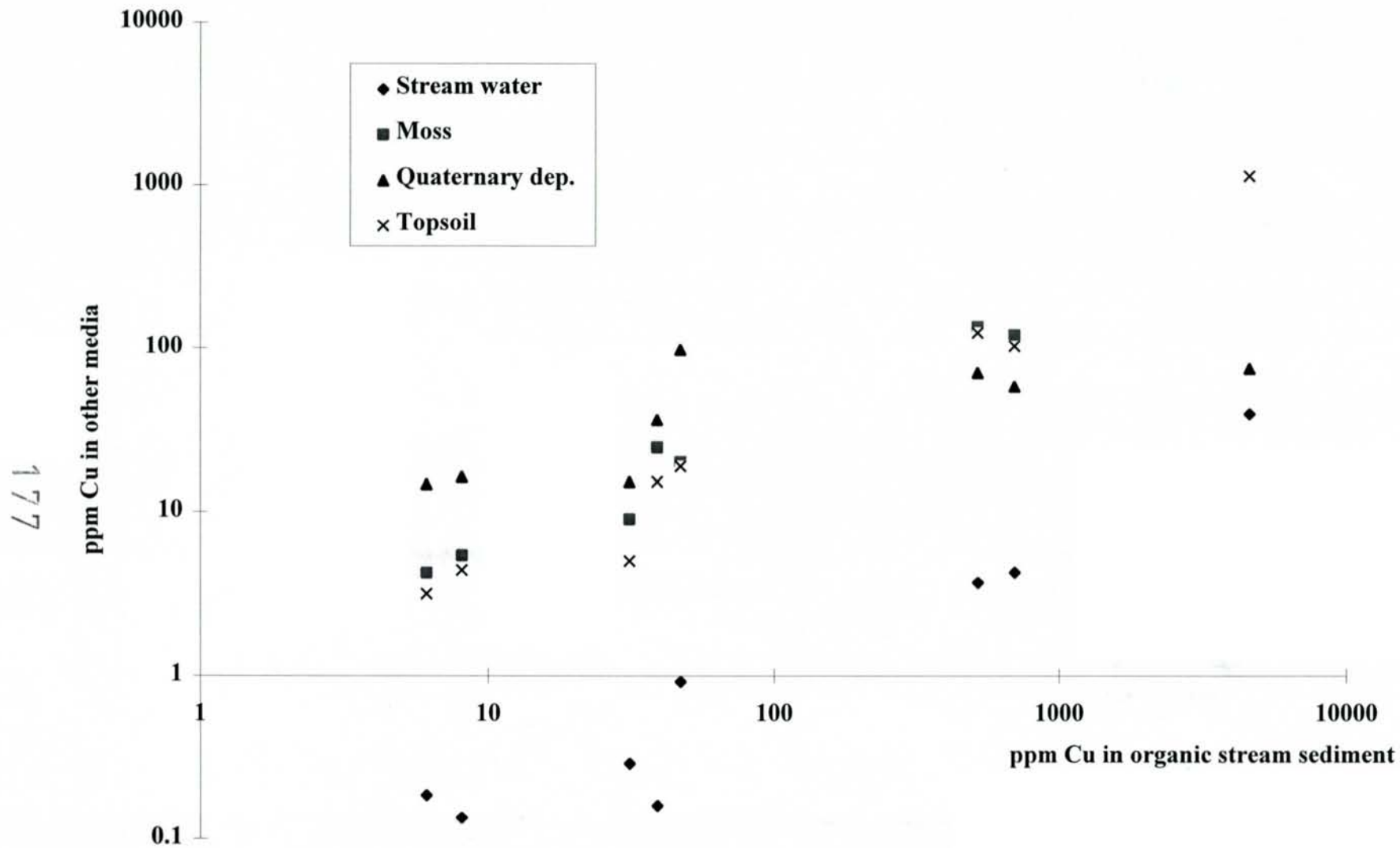


Figure 5.1.5. XY- diagram of the median concentrations of Cu in the Organic stream sediments (X-axis) from the eight catchments versus on the Y-axis the median concentration of Cu in stream water, moss and topsoil (aqua regia extractions) from the same catchments. All values in ppm.

## 5.2 Overbank sediments from the surroundings of the Russian nickel mining and smelting industry on the Kola Peninsula

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### ABSTRACT

At 43 sites top and bottom samples of overbank sediment were collected in a 12000 km<sup>2</sup> area in the surroundings of the nickel smelter in Nikel and the ore roasting plant at Zapoljarnij, Russia. In addition three detailed overbank profiles were sampled in 10 cm sections in 3 catchments representing different levels of pollution. All samples were analysed for more than 30 elements by ICP-AES following an aqua regia extraction.

Results obtained show that overbank sediments represent rather good natural (geogenic) background levels. Although the major pollutants (Co, Cu, Ni and S) are strongly enriched at some places close to industry, overbank sediment is not especially well suited to map the extent of airborne contamination in the area. Co, Cu Ni and S are mobile within the profile collected from the most polluted catchment.

### INTRODUCTION

Overbank sediments have been introduced relatively recently by Norwegian geochemists (Ottesen et al., 1989) as a sample medium for regional geochemical mapping. Overbank sediments are deposited on river floodplains outside the main drainage channel during large floods. Because the floods depositing overbank sediments transport a very high proportion of the total sediment load of a river, overbank sediments should be a more representative sample medium than normal stream sediments and thus give a better average geochemical characterisation of their catchment basin.

Depending on local sedimentation rates and age of the river, the age of overbank deposits can vary between tens and hundreds of years. Recent investigations in different parts of the world have proven that sampling of overbank sediments is well suited to provide a fast overview of the main geochemical characteristics of large areas (e.g. Ottesen et al. 1989, Edèn and Björklund 1994 and Xiachu and Mingkai, 1995). Overbank sediments have thus been proposed as one of the sample media for world wide low density geochemical mapping (Darnley et al., 1995).

A further interesting feature of overbank sediments is that they consist of nearly horizontal strata and thus offer, just like lake sediments, the opportunity to study the development of element contents over time. Many of the media currently used in pollution studies (e.g. precipitation, moss, soil) suffer under the shortcoming that they do not provide any information about historical, prepollution, element levels. Lake sediments are thus often used to establish the state of pollution (the ratio of the present element level to element levels measured at depth in layers predating industrialisation).

Advantages of overbank sediment profile samples when compared to lake sediments are (1) they dry out between major floods and are then easily accessible for sampling; (2) they are



more widespread than lake sediments and (3) sample collection is more controlled because overbank sediments can be visually inspected while sampling. However, in contrast to lake sediments they do not develop in clearly distinguishable yearly layers but layers as deposited with every major flood. It is thus necessary to age date different depth along the overbank profile to make best use of the inherent time information. Another difference is that lake sediments do normally develop under anoxic conditions, preventing any element mobility within the profile, while overbank sediments will experience steady changes of redox conditions, depending on the water level of the river.

Overbank sediments have been successfully used to map the state of pollution along streams draining mining or industrial areas (e.g. Ottesen et al, 1989, Demetriades et al., 1990, Bogen et al. 1992, Bølviken et al. 1993, Edèn and Björklund, 1994; and Langedahl et al., 1995). Even airborne pollution could be detected in an overbank sediment profile near the Norwegian/Russian border (Grense Jakobselv, Langedahl and Ottesen, 1995).

Overbank sediment profiles were also used for archaeological-oriented studies to identify ancient mining and smelting sites and to study and age date different stages of human activities within a catchment basin (e.g. Matschullat et al. 1996) for several hundred years into the past.

Bølviken et al. (1993) suggested that by sampling just two different layers (the uppermost 10 cm and a second sample at depth) of overbank sediment profiles it should become possible to map both, pollution and natural element levels fast and cheap over large areas.

Macklin et al.(1994) and Ridgway et al.(1995) demonstrated that in areas of historical mining and pollution only one overbank sample might not be very representative for geochemically characterising a rather large catchment basin. In addition they showed that rather detailed age dating is mandatory when planning to use overbank sediments for the assessment of natural versus man made element levels.

Studies using sediment cores to assess the human impact on the environment assume a constant natural element flux over time. This may not be the case as sediment sources or weathering conditions can change with time as well.

The Geological Surveys of Finland (GTK) and Norway (NGU) and the Central Kola Expedition (CKE) in Russia are carrying out a major geochemical mapping project (see World Wide Web site <http://www.ngu.no/Kola>) in a 188,000 km<sup>2</sup> area north of the Arctic Circle, comprising the entire area between 24° and 35.5° E north to the Barents Sea (Fig. 5.2.1). The area contains some of the world's largest point sources for S and heavy metal pollution: the nickel smelters in Monchegorsk and Nikel as well as the ore roasting plant in Zapoljarnij.

The project was started in 1992 in the form of a pilot project mapping a 12,000km<sup>2</sup> subarea around the nickel smelter in nikel and the ore roasting plant in Zapoljarnij (Fig.5.2.1). Several different sample media (snow, terrestrial moss, stream water, O-and C-horizon of soils, stream sediments, overbank sediments) were collected and analysed to decide on the best suited approach for the regional mapping project (Åyräs et al., in prep., Niskavaara et al. 1996, Reimann et al. 1995, 1996, in prep.a). Following the proposal of Bølviken et al. (1993) the top 10cm of overbank deposits were sampled to assess pollution and, in addition a second section was sampled at a depth of > 40 cm to represent the natural background level.

As second part of this project, eight catchments (hereafter abbreviated as C1 - C8) widely distributed in the area (Fig. 5.2.1, Tab. 5.2.1) were investigated in detail in 1994. Media sampled were: snow (meltwater and filter residue), rainwater, stream water, organic stream sediments, terrestrial moss, topsoil (0-5 cm), complete podzol profiles, Quaternary deposits and bedrock. In addition detailed overbank sediment profiles were sampled at the outlet of C2, C5 and C7, where no major sediment trap (lake, bog) existed upstream.

Here we present the results of regional mapping of two different layers of overbank sediment profiles in an 12,000 km<sup>2</sup> area that was pristine until 60 years ago and has since been so strongly polluted that the vegetation in large areas around industry is totally damaged. These results are compared with the results of soil samples taken from the same catchment basins (O- and C-horizon) and detailed overbank sediment profiles taken in 3 drainage basins with different distance to and levels of pollution to assess the applicability of overbank sediment sampling for regional mapping of the geochemical background and of anthropogenic pollution levels at the same time.

## STUDY AREA

The smelters in Nikel, Zapoljarnij and Monchegorsk (Fig. 5.2.1) have been active since 1932, 1955 and 1938 respectively. Two different types of ore are processed (1) local, Pechenga, ore with average contents of 1.5 to 2% Ni, 0.5 to 1.8% Cu and 3-5% S and (2) imported ore from the Norilsk-Talnakin deposits in northern Siberia which has considerably higher S-contents (see Boyd et al., in prep.). Prevailing south-westerly winds distribute the pollution plume mainly in a north-eastern direction, towards the Barents Sea. A steep gradient in deposition towards the west is reported in several studies (e.g. Sivertsen et al., 1992, Niskavaara et al., 1996, Reimann et al., 1995 a, in prep.d).

Studies of the precipitation chemistry in the eight catchments (Äyräs et al. 1995, Reimann et al., in prep.b) showed that of the three catchments investigated here C2 is strongly polluted, in C5 pollution is barely detectable and C7 can be considered as a pristine background catchment.

## SAMPLING

For the pilot project, at each sample site, representing a catchment basin of 30 to 100 km<sup>2</sup> in size, a vertical profile was dug to the base of the overbank sediment deposit (or the groundwater level). For the pilot project the recommendations given in Bølviken et al. (1993) were followed. From the pit a top sample was collected from the uppermost 10cm of the profile. The weight of the top samples was between 2 and 3 kg. To represent background or natural element levels, a bottom sample was taken from the base of the overbank deposit (or the groundwater level) to the middle of the profile. This sample had a weight of approximately 5 kg.

For the catchment study approximately 300g of samples were taken from every 10cm of overbank deposit from top to bottom on carefully selected floodplains in C2, C5 and C7.

## ANALYSIS

All samples were dried at temperature below 40°C at the Geological Survey of Norway (NGU) laboratory. The pilot project samples were sieved at <0.125 mm, the samples from the catchment study were sieved to < 0.063mm using nylon screens.

A 2 g subsample was then digested for 2 hours at 90°C with 12 ml aqua regia (1:3 HCl and HNO<sub>3</sub>) (modified ISO standard draft 11466, GTK-method 512) at the Geological Survey of Finland (GTK) laboratory. After digestion, samples were diluted with water to 60ml, mixed and centrifuged. The clear solution was analysed with ICP-AES for 31 elements (GTK-method 512P - for more details see Niskavaara, 1995). For the catchment study samples Ag, As, Cd and Pb were additionally determined by graphite furnace AAS, Hg with cold vapour AAS and Bi, S, Se, and Te with graphite furnace AAS after preconcentration with reductive co-precipitation (Niskavaara and Kontas, 1990).

All samples were in addition analysed at NGU's laboratory by ICP-AES following a 7N-HNO<sub>3</sub>-extraction. Results as obtained from both laboratories were in very close agreement. Here we use only the results of GTK's laboratory. As an additional quality control measure, field duplicates were collected at 13 sample sites. Analytical results received from these field duplicates were again in good agreement with the results of the original samples (with the exception of Ag, B and Cd). These results are documented in Chekushin et al. (1993).

The GTK laboratory is accredited according to EN45001 and ISO-Guide 25.

## RESULTS

### *Pilot Project*

Fig. 5.2.2 compares element levels and variation as observed in the top- and bottom layer of 43 overbank sediment sample sites with those obtained from O- and C-horizon (2 different grain size fractions) soil samples, taken within the same catchment basin, by use of the boxplot (Velleman and Hoaglin, 1981). Note that the analytical results for some elements (Cu, Ni, S) had to be log-transformed prior to plotting due to the very large variation of element contents observed in the survey area.

Generally, the results obtained from the overbank sediment samples are very comparable with the analytical results from the C-horizon soil samples, not, however with those from the O-horizon, the latter displaying considerably lower values for most of the elements. An exception are the main contaminants Cu and Ni, which are so strongly enriched in the O-horizon that these samples show similar median contents and the highest maximum values when compared to the C-horizon and the overbank sediments.

Sulphur is an element showing a strong enrichment in the overbank sediments (as in the O-horizon) when compared to the C-horizon samples. In the O-horizon, the observed high S-levels are associated with organic matter and not with contamination (Reimann et al., in prep.b). For the overbank sediments this relation is not so clear and it could actually be that S, not being accumulated in the soil, is at the moment accumulated in the overbank sediments (Niskavaara et al. 1996).

Lead is another element that shows a different behaviour. The highest median contents are observed in the O-horizon soil samples, where Pb is bound to organic matter and in the fine fraction of the C-horizon, where Pb is probably adsorbed on the surface of clay minerals. The overbank samples show here levels similar to the coarse fraction of the C-horizon.

These diagrams show clearly, that overbank sediments have a close relation to the geogenic, natural element levels in the area and that the bulk is not very much affected by atmospheric pollution. It is a rather interesting information that overbank sediment samples are so closely

related to the element contents as observed in the C-horizon soil samples (till). Regional results, e.g. those of the geochemical atlas of Norway (Ottesen et al. in prep.), can thus very well be taken as a first approximation of the element levels that have to be expected in the deeper soil layers throughout the country.

Top and bottom of the overbank sediment samples show very small differences in element levels in these boxplot comparisons.

Regional maps for the top and bottom part of overbank sediment profiles for the main pollutants in this area (Co, Cu, Ni and S - Fig. 5.2.3) demonstrate again that element levels are very comparable; the bottom samples, which should represent "natural" background values, actually displaying higher maximum contents for all four elements mapped here. The highest values are all observed in an overbank sediment profile sample close to the nickel smelter at Nikel. The Russian nickel industry is marked by high element levels in both layers of the overbank sediments at 3-5 (out of 43) sites. Maps from other media (e.g. snow, moss, O-horizon- soil samples or even stream water - Niskavaara et al. 1996, Reimann et al. 1995, 1996, in prep.a) outline the contaminated area considerably better. For the O-horizon samples between 10-12 sites (out of 45) show unusually high element contents in the surroundings of Nikel and Zapoljarnij.

The general lack of detectable higher element levels in the top samples throughout the survey area may be caused by the fact that a 10 cm slice of overbank sediment gives a too poor time resolution. Langedahl and Ottesen (1995) suggest that it is necessary to sample 1 cm intervals in order to avoid the combination of sediments from polluted and pre-industrial material in this area. Further on this is an area with known nickel occurrences, so that already the natural background levels can be expected to be rather high. The values observed in overbank samples from the surroundings of industry are, however, in both layers considerably above any reasonable natural level when compared with results from the C-horizon soils from the same area or with the levels reported for overbank sediments from Norway (Tab. 5.2.2).

The consistently higher maximum values at the bottom of the overbank sediment profiles for Co, Cu and Ni can be explained as a result of the severe damage to the ecosystem in the surroundings of the smelter. High levels of mobile Cu and Ni were detected in the soils around Nikel and Zapoljarnij (Niskavaara et al., 1996). At the same time humus formation and heavy metal binding by organic complexing are disturbed. Unusually high levels of mobile heavy metals thus reach the aquatic environment and are obviously accumulated in and moving through the overbank sediment profiles. Unusually high erosion rates due to the damaged vegetation in the surroundings of industry can further this process. Heavy metals are thus presently strongly accumulating in the aquatic ecosystem and will eventually reach their final (?) sink in the Barents Sea. Indications for the mobility of heavy metals in the overbank sediments are a rather serious threat to groundwater quality in the area.

Fig. 5.2.4 shows the correlation between top and bottom samples for eight selected elements. Generally the correlation between top and bottom is rather good. When studying the deviation from the 1:1 relationship between bottom and top samples as displayed by the lines in the diagrams a number of peculiarities can be detected. Most of the bottom samples show for example clearly higher Al-contents, arguing for a finer grain size distribution in the samples taken at the bottom of the profiles. This would almost automatically result in higher heavy metal values at the bottom and is especially prominent for the majority of the Russian samples. It can be speculated whether this is an indication of increased erosion, resulting in coarser

sediments being deposited at present. For Co, Cu and Ni a number of samples from Russia and Norway do actually display a slight enrichment in the top samples - this may be an indication that the top samples are indeed influenced by airborne pollution but that the differences are so small that they are hard to detect in regional mapping. Iron and V show a number of strongly enriched bottom samples in the Russian project area.

#### *Catchment study - detailed profiles*

Figures 5.2.5 and 5.2.6 compare the results of detailed overbank sediment profile sampling in three catchments (C2, C5 and C7 - see Fig. 5.2.1) situated at different distances to industry. C2, near Monchegorsk, represents a highly polluted catchment with a severely damaged ecosystem, C5 represents low level pollution with no obvious damage to the ecosystem while C7 represents a typical background catchment for the whole area. In C7 soil was developed on top of the overbank sediment profile. Sampling of the overbank sediment profile thus started here at a depth of 30 cm. Analytical results from the profiles are summarised in Tab. 5.2.3.

<sup>210</sup>Pb-age dating of six overbank sediment profiles from different parts of Norway (internal FORCE-report to NGU, 1992) resulted in quite different sedimentation rates for overbank sediments, ranging from 0.03 - 0.3 cm/y. For the project area, a value of 0.22 cm/y, measured for the Skjellbekken catchment (C5) can be taken as the most representative. 10 cm samples should thus represent about 40 years of sediment accumulation. Industry in the area started in the 1930ies, thus the uppermost one or two 10cm-composite samples of the profile should show the effects of contamination on the element levels measured.

In C2 the bottom of the overbank sediment profile was reached at 45cm. At above average sedimentation rate this should represent an age of over 200 years. When using sediment rates to model the age of overbank deposits one must, of course, keep in mind that overbank sediment accumulation is a discontinuous process, depending on the frequency of flooding events. These may be drastically increased when vegetation and humus layer are destroyed.

Fig. 5.2.5 shows the depth distribution of the major contaminants Co, Cu, Ni and S in overbank sediment profiles from the three catchments. The observed element levels are throughout the whole profile much higher in C2 than in C5 or C7. The highest Co, Cu and Ni values are observed in the topmost 2 samples in C2. These top values show about 10 times higher heavy metal values than the bottom samples from the same profile. Minimum element contents in C2 (still by about a factor of 10 higher than the element levels in the other two catchments) occur at a depth of 30 cm in the middle of the profile, the last two samples show increasing values again. No enrichment from bottom to top can be seen in C5 or C7 and levels measured can in both cases be taken to represent natural background levels.

Sulphur behaves differently. Again levels in C2 are generally higher than those observed in C5 or C7. Here the highest S-value occurs at 30 cm depth in C2, with decreasing levels to both, top and bottom. In C5 and C7 the highest S-values are actually occurring at the bottom of the profiles.

Fig. 5.2.6 shows the depth distribution of four further elements in these profiles - Al, Fe, Pb and Mn. Al shows strongly decreasing contents in the upper parts of the overbank sediment profile from C2 - probably indicating a change in the sediment source with increasing erosion following the increase in pollution and subsequent die-back of vegetation in the catchment. Fe, Pb and V - all minor contaminants - do surprisingly show in C2 the behaviour that should be expected for all elements having their main source in airborne pollution: strongly increasing

contents towards the top of the profile and levels that reach regional background as indicated by the samples from C5 and C7 at depth.

The most likely conclusion from the detailed study of the three profiles is thus that the major pollutants Co, Cu, Ni and S are mobile within the profile in C2.

Fe and Pb show actually a clear enrichment towards the top of the profile in C7 as well. Here we do not have an easy explanation of this feature. The Pb enrichment observed may, of course, be due to the general increase of lead in the environment observed world-wide. The higher Fe levels in the top two samples could be attributed to dust input from the large iron ore open pit mine near Kovdor in Russia. This shows one of the weaknesses of interpreting the historical records stored in sediment cores. It is generally only possible to find a likely explanation for observed patterns when a source is known.

## CONCLUSIONS

Multimedia regional geochemical sampling of a 12000 km<sup>2</sup> area in the European Arctic (Kola Project Pilot Project) showed that overbank sediment samples and C-horizon soil samples taken in the same catchment basins show quite comparable element levels. O-horizon soil samples show for most elements quite different concentrations. In regional geochemical mapping overbank sediments will thus best reflect the composition of the deeper soil levels, till and bedrock in any one area.

Close to major contamination sources (here the nickel smelter at Nikel and the ore roasting plant at Zapoljarnij), heavy metal values in the O-horizon samples increase drastically over rather large areas (10-12 out of 45 samples showing clearly enhanced heavy metal levels) (Niskavaara et al, 1996, Reimann et al. in prep.a). Using the uppermost 10 cm of overbank sediment this major antropogenic contamination was detected in only 2-5 out of 43 catchment basins sampled.

Element values observed in top and bottom samples of overbank sediment profiles throughout the pilot project area correlate rather well. No large differences in element concentrations between “young”(top) and “old”(bottom) samples are detected. This could be taken as an indication of rather low contamination levels over large parts of the survey area or rather high natural background levels in this area.

In some catchments, however, both, top and bottom samples show strongly enriched contents of the main contaminants Co, Cu and Ni. The bottom samples could thus not be used to give an indication or separation of “natural”, pre-industrialisation element levels from pollution. It rather appears that the main contaminants Co, Cu and Ni are be mobile within the profiles sampled in the immediate vicinity of industry.

A detailed study of 10 cm - slices of overbank sediment profiles in three catchments representing different levels of pollution gives strong evidence for the mobility of the main contaminants Co, Cu, Ni and S within the profile from the most polluted site. This result indicates that groundwater resources may be at risk in the surroundings of the Russian nickel smelters.

## ACKNOWLEDGEMENTS

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## TABLES

Tab. 5.2.1: Characteristics of the three catchments were detailed overbank sediment profiles were sampled.

Tab. 5.2.2: Element contents in top- and bottom parts of overbank sediment profiles (fraction <0.125mm) compared to C-horizon soils (fraction <2mm) and O-horizon soils (fraction <2mm) from the same area and overbank sediments (fraction <0.063mm, N= ) from the whole of Norway. Top- and bottom samples of overbank sediment aqua regia extraction, O-horizon soil samples conc.HNO<sub>3</sub> extraction, C-horizon soil samples aqua regia extraction and overbank sediments Norway 7N HNO<sub>3</sub> extraction.

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Fig. 5.2.2: Boxplot comparison (Velleman and Hoaglin, 1989) of element contents and variations in O-horizon soils ("a0t")(conc. HNO<sub>3</sub>-extraction), the C-horizon sieved at <2mm ("c2"), the same sieved at <0.063mm ("C06")(both aqua regia extraction) and top- ("OBT") and bottom- ("OBB") part of overbank sediment profiles (fraction <0.125mm, aqua regia extraction) from the pilot project area. Note that data for some elements were log-transformed prior to plotting.

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Fig. 5.2.5: Depth profiles of Co, Cu, Ni and S-content in C2 (strongly polluted) compared to C5 (low level pollution) and C7 (no pollution).

Fig. 5.2.6: Depth profiles of Al, Fe, Pb and V-content in C2 (strongly polluted) compared to C5 (low level pollution) and C7 (no pollution).

No name	Size (km <sup>2</sup> )	Elevation (masl)	Annual precipitation (mm)	Vegetation	Bedrock	Surface cover
C2 Monchegorsk	22	128 - 507	391	Technogenic desert, birch shrubs	Dacite & andesite & tuffs, gabbro/norite	Till, prone to erosion
C5 Skjellebekken	34	80-297	422	North taiga pine forest, birch	Andesite, basalt & tuffs, black shales	Till, esker
C7 Naruska	20	263-490	513	North taiga spruce forest	Gneiss	Till, peat

Tab. 5.2.1: Characteristics of the three catchments were detailed overbank sediment profiles were sampled.

Element mg/kg	Overbank sediment -top				Overbank sediment - bottom				Overb.sed. Norway median	C-horizon <2mm median	O-horizon <2mm median
	25 %	median	75 %	max.	25 %	median	75 %	max.			
Ag	<0.5	<0.5	<0.5	0.6	<0.5	<0.5	<0.5	0.8		0.12	na
Al	5500	7400	10300	21600	6300	10300	13800	24800	16000	7680	1410
As	<3	<3	<3	20	<3	<3	<3	21		0.9	1.2
B	<3	<3	<3	<3	<3	<3	<3	4.5		na	2.1
Ba	30	43	89	224	37	48	71	239	63	26	70
Ca	2500	3300	4000	10700	2500	3200	4200	25500	4600	2160	3250
Cd	<0.3	<0.3	<0.3	0.9	<0.3	<0.3	<0.3	0.5	<1	0.04	0.41
Co	5	7	14	60	5	8	12	94	12	5	2
Cr	7	12	15	172	9	14	21	447	27	19	2
Cu	10	16	27	595	14	20	27	849	22	16	13
Fe	10600	15300	25200	59800	11600	16100	24500	116000	23200	12000	1710
K	200	400	900	4300	300	600	1000	4400	1800	672	691
La	11	16	22	50	16	19	26	42	61	13	2
Li	5	7	10	24	6	9	13	24	16	6	0.3
Mg	1250	1550	2350	9500	1400	2000	2750	20500	6200	2950	932
Mn	112	174	386	13100	103	155	294	5060	300	105	178
Mo	<0.5	0.6	1.7	25.3	<0.5	0.5	1.6	12.9		0.25	0.6
Na	<400	<400	<400	800	<400	<400	<400	1400	200	141	98
Ni	9	15	22	645	10	17	25	938	18	11	14
P	578	704	889	1980	529	747	900	1830	800	490	775
Pb	<3	5	6	24	4	5	8	13	16	5	12
S	119	209	611	2220	84	144	447	7260		48	1460
Sb	<5	<5	<5	<5	<5	<5	<5	<5		0.6	1.3
Sc	0.6	0.9	1.1	3.1	0.9	1.2	1.7	3.9	3.9	2	0.5
Si	<800	<800	<800	2000	<800	<800	<800	1800	100	282	279
Sr	7	11	19	61	8	12	17	168	28	7	26
Th	3	5	8	15	3	5	9	41		4	0.6
Ti	493	801	1060	2250	676	1000	1300	2700	1200	788	193
V	24	37	53	85	25	40	52	150	39	28	5
Y	2	3	4	7	3	4	5	8		5	0.7
Zn	18	25	34	118	22	29	41	131	45	20	43

Tab. 5.2.2: Element contents in top- and bottom parts of overbank sediment profiles (fraction <0.125mm) compared to C-horizon soils (fraction <2mm) and O-horizon soils (fraction <2mm) from the same area and overbank sediments (fraction <0.063mm, N= ) from the whole of Norway. Top- and bottom samples of overbank sediment aqua regia extraction, O-horizon soil samples conc.HNO<sub>3</sub> extraction, C-horizon soil samples aqua regia extraction and overbank sediments Norway 7N HNO<sub>3</sub> extraction.

Element mg/kg	Catchment 2 (N=5)			Catchment 5 (N=9)			Catchment 7 (N=13)		
	min	med	max	min	med	max	min	med	max
Ag	0.09	0.25	0.96	0.01	0.02	0.04	<0.01	0.02	0.06
Al	16800	20600	21500	7210	8940	14800	9110	12400	18800
As	0.7	12	139	1.4	2.5	6.4	0.05	0.4	1.2
B	<3	<3	3	<3	<3	4	<3	<3	4
Ba	136	191	237	31	45	79	30	38	61
Bi	0.13	0.16	0.57	0.05	0.07	0.09	0.08	0.14	0.16
Ca	4640	6350	7160	4560	5800	6810	1380	1770	2330
Cd	0.17	0.22	1.82	0.02	0.05	0.06	<0.01	0.01	0.12
Co	19	61	641	6	7	13	3	5	6
Cr	116	121	149	19	24	42	44	63	81
Cu	88	496	3440	22	32	52	3	6	11
Fe	22300	28100	51600	12100	14900	27200	6010	8120	20400
Hg	0.06	0.16	0.2	0.01	0.02	0.03	0.01	0.03	0.1
K	700	900	1300	500	700	1400	200	300	500
La	23	28	33	16	22	25	16	23	31
Li	8	14	15	6	7	14	7	13	18
Mg	4700	5140	5280	3890	4850	8760	1470	2360	3240
Mn	167	444	6300	141	176	274	34	56	69
Mo	2.4	4.6	19.4	<1	<1	1.1	<1	<1	<1
Na	260	340	420	220	300	350	90	110	130
Ni	186	722	5880	12	15	26	14	20	28
P	918	1010	1090	934	1090	1140	293	367	867
Pb	1.4	1.6	31	1.4	1.6	1.9	1	1.8	6.2
S	1850	3870	5710	230	828	1930	280	571	2290
Sb	0.09	0.16	1.04	<0.01	0.02	0.05	<0.01	0.01	0.06
Sc	1.6	2.3	2.8	3.1	3.9	5.7	0.6	1.8	2.3
Se	2.9	3.7	7.7	0.22	0.39	0.48	0.4	0.63	1.25
Si	240	250	270	200	230	250	200	210	270
Sr	33	37	40	10	13	15	9	13	16
Te	0.05	0.7	4.8	<0.01	<0.01	0.01	<0.01	<0.01	0.03
Th	<5	<5	<5	5	7	9	<5	<5	<5
Ti	478	695	827	821	949	1530	308	600	787
V	56	90	111	33	44	71	26	32	43
Y	9	11	12	7	9	11	3	4	7
Zn	61	63	122	29	37	63	8	16	25
LoI - %	27	30	32	<0.1	3.4	4.2	7.6	13.3	23

**Tab. 5.2.3: Minimum, median and maximum content of all analysed elements in overbank sediment profiles from C2, C5 and C7. Grain size fraction <0.063mm, aqua regia extraction.**

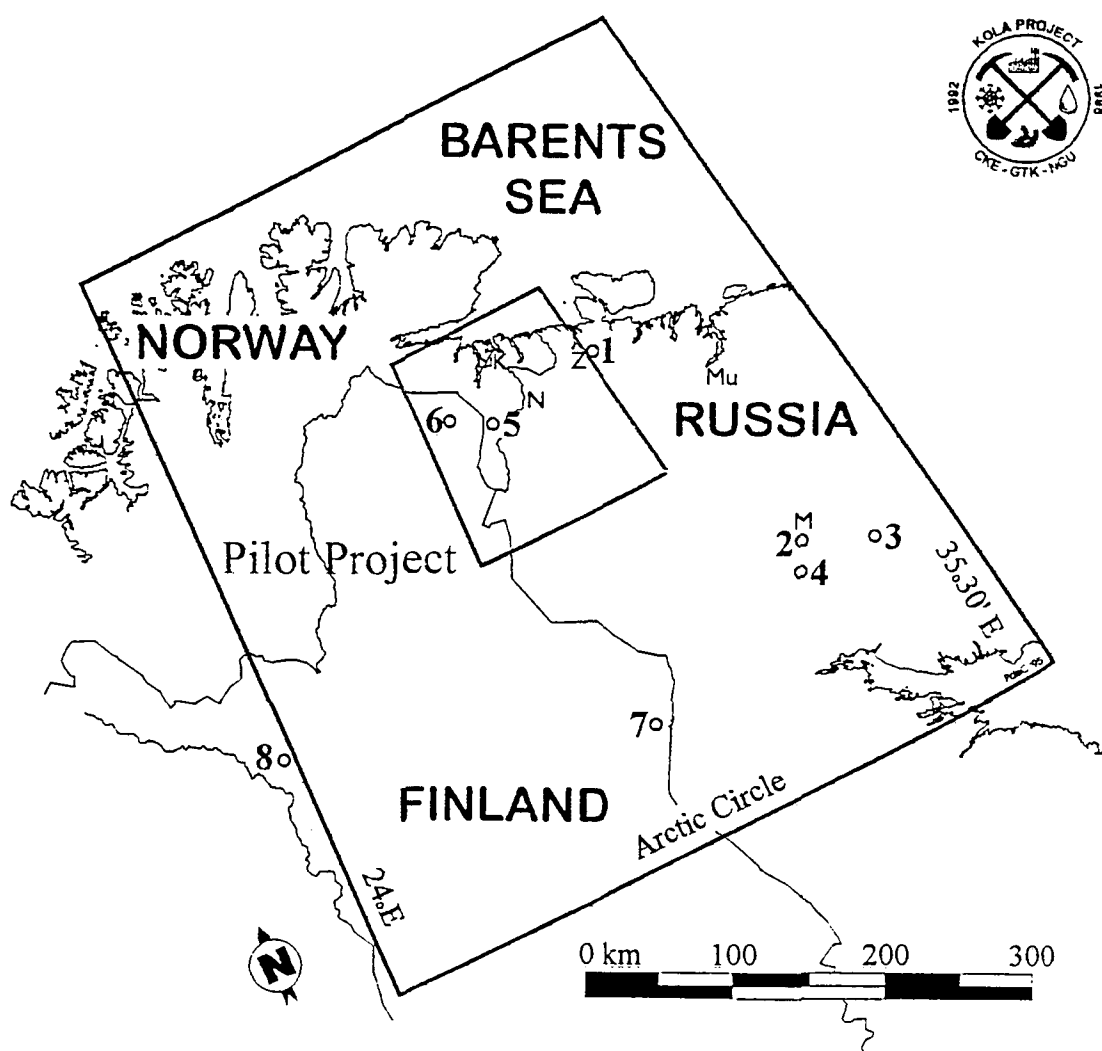
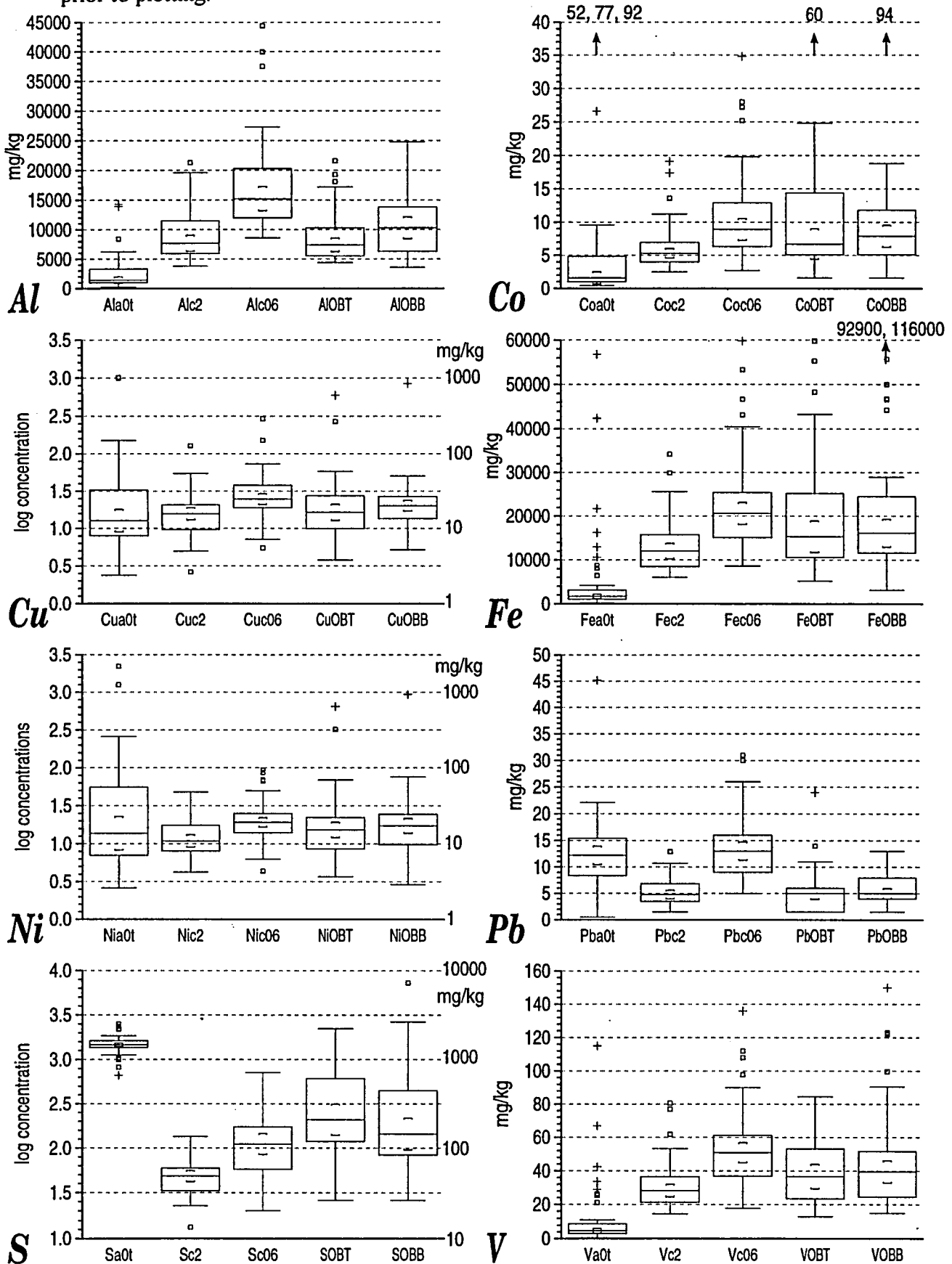


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Fig. 5.2.2: Boxplot comparison (Velleman and Hoaglin, 1989) of element contents and variations in O-horizon soils ("a0t")(conc. HNO<sub>3</sub>-extraction), the C-horizon sieved at <2mm ("c2"), the same sieved at <0.063mm ("C06")(both aqua regia extraction) and top- ("OBT") and bottom- ("OBB") part of overbank sediment profiles (fraction <0.125mm, aqua regia extraction) from the pilot project area. Note that data for some elements were log-transformed prior to plotting.





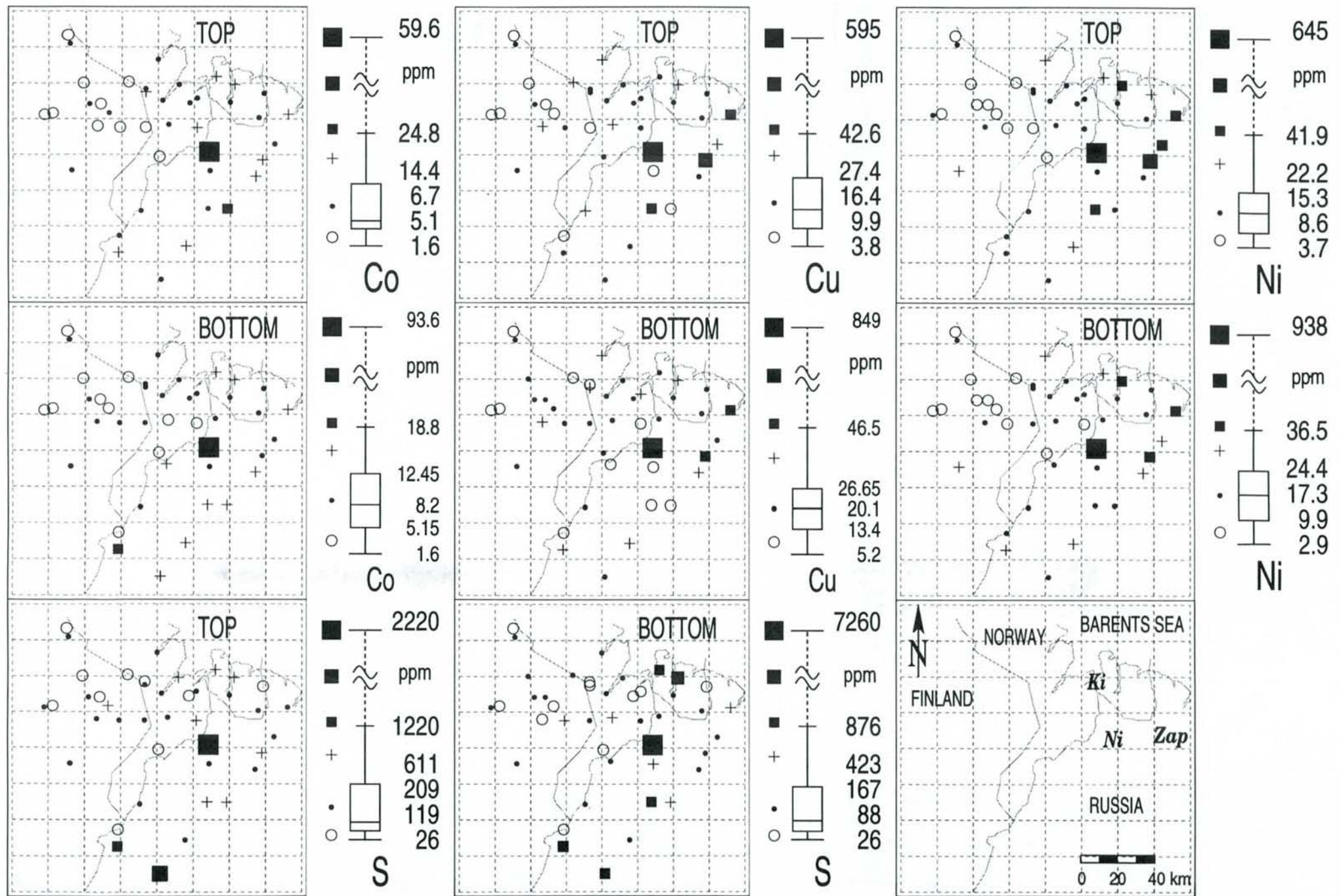
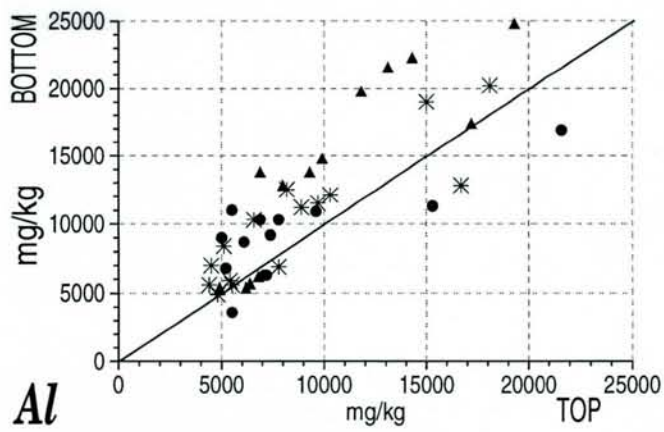
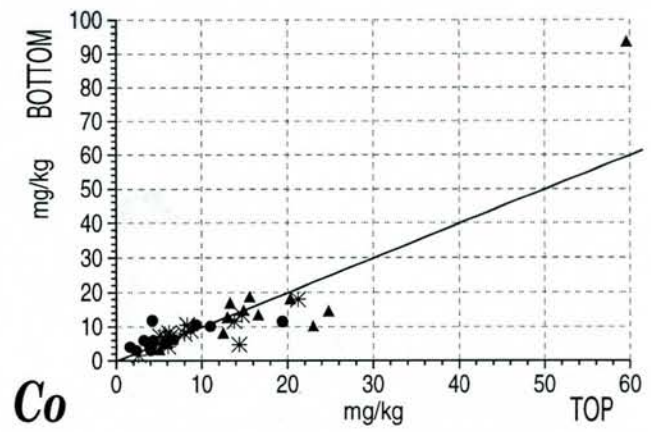


Fig. 5.2.3: Maps showing the regional distribution of Co, Cu, Ni and S in top(0-10cm)- and bottom(>40cm) samples of overbank sediment profiles in the pilot project area.

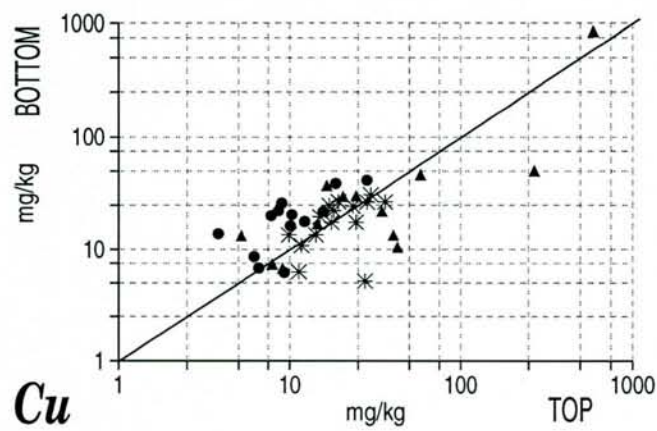
Fig. 5.2.4: XY-Diagrams comparing analytical results of top- and bottom- samples of overbank sediment samples from the pilot project area. The lines indicate a ratio of 1:1.



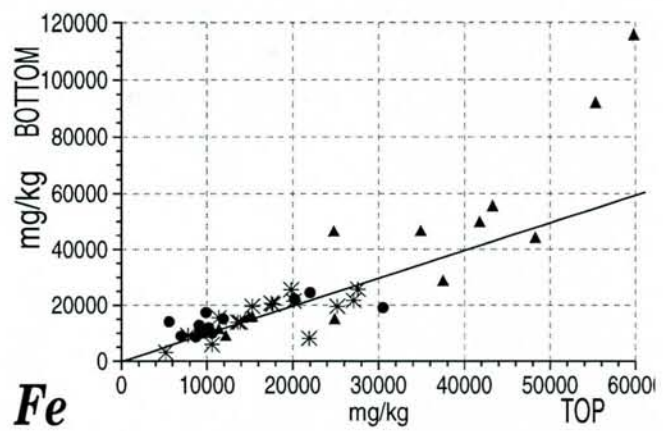
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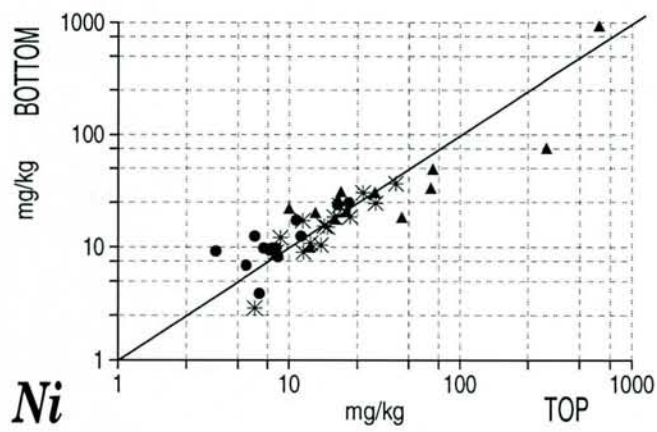
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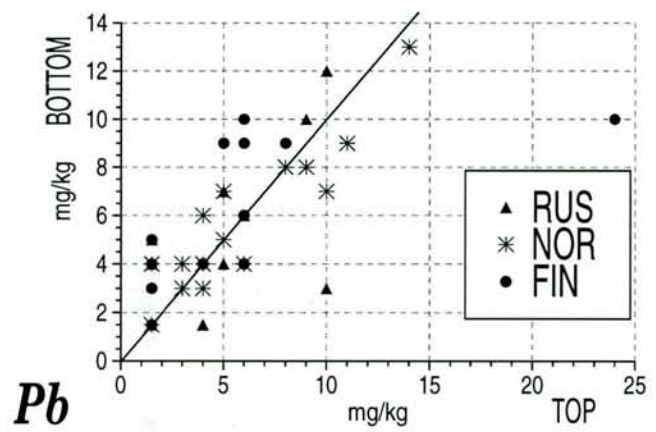
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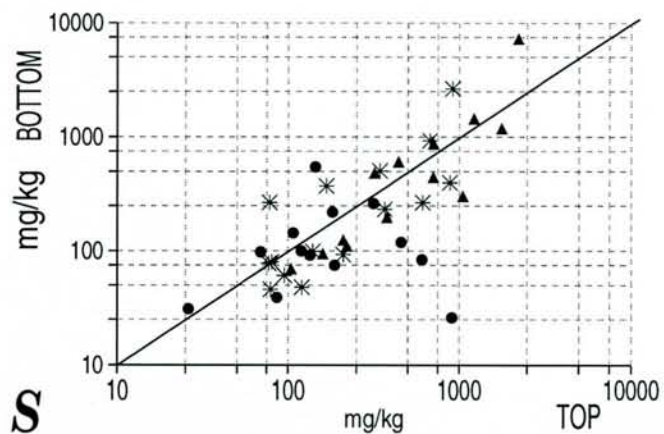
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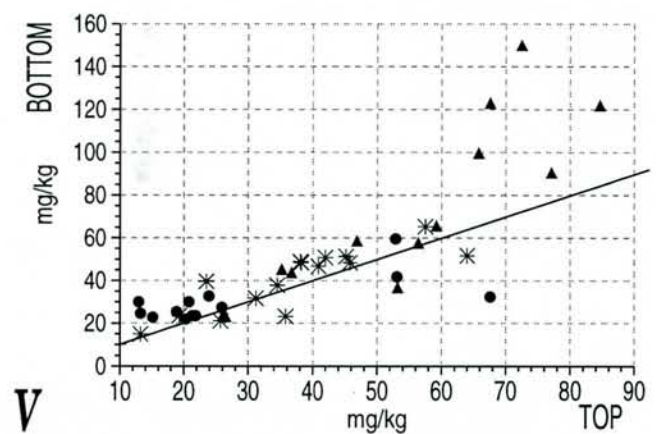
**Ni**



**Pb**



**S**



**V**



Fig. 5.2.5: Depth profiles of Co, Cu, Ni and S-content in C2 (strongly polluted) compared to C5 (low level pollution) and C7 (no pollution).

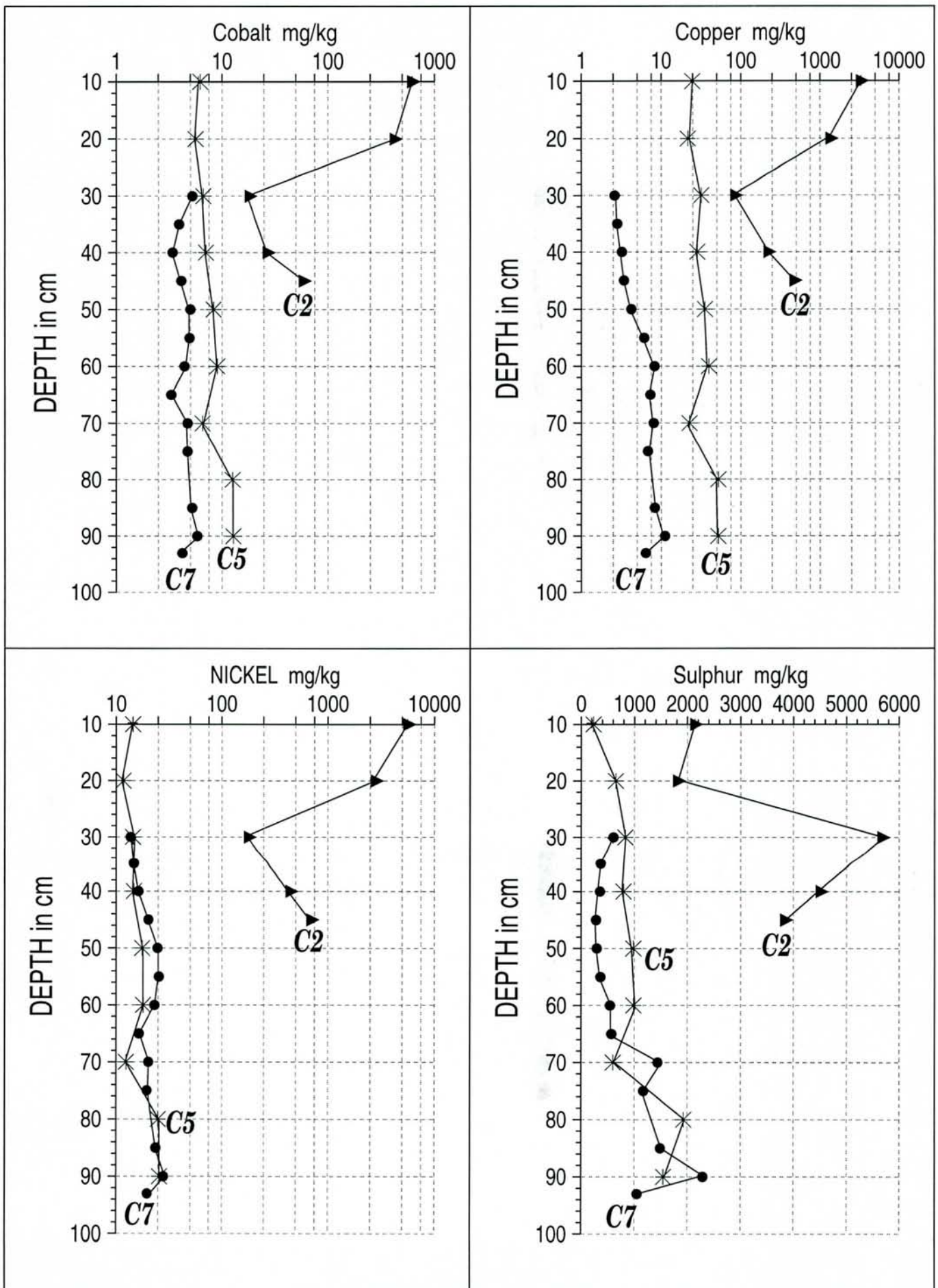
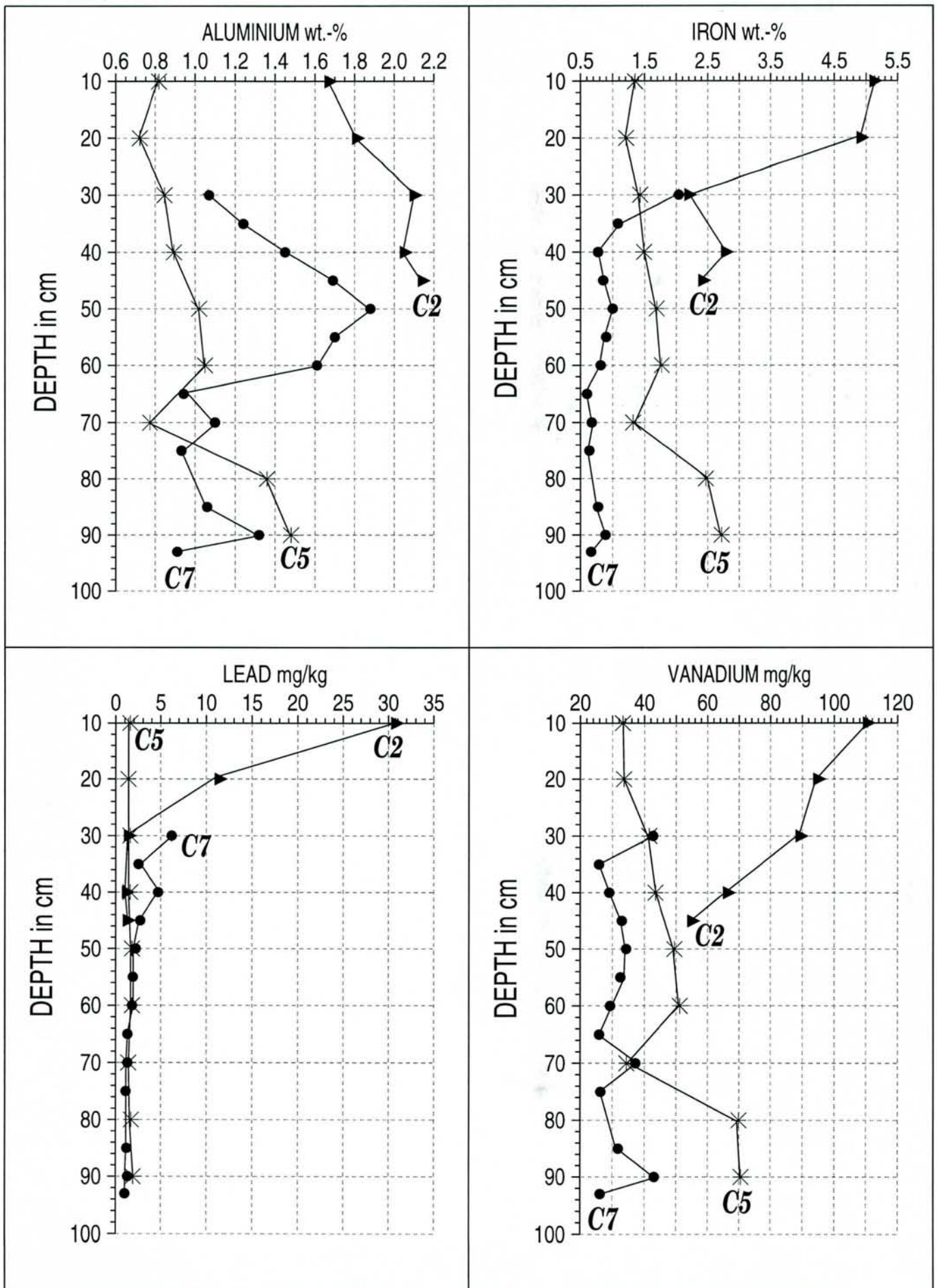


Fig. 5.2.6: Depth profiles of Al, Fe, Pb and V-content in C2 (strongly polluted) compared to C5 (low level pollution) and C7 (no pollution).



## 6. VEGETATION

### 6.1 Local versus regional variability of feather moss chemistry in pristine and technogenic parts of the Barents Region

*Matti Äyräs, Jo Halvard Halleraker, Vladimir Pavlov and Igor Bogatyrev*

#### ABSTRACT

Terrestrial mosses (*Hylocomium splendens* and *Pleurozium schreberi*) were sampled twice during 1994 from seven areas restricted to catchments (12 - 35 km<sup>2</sup>), and analysed for 38 elements by use of advanced techniques (ICP-AES/MS and AAS). The studied catchments are situated at various distances from the Barents sea, and from major industry centres like Ni-Cu smelters and Fe smelters and from dust sources like open pit mines. Three of the areas were in the Kola Peninsula (NW Russia), one in Pasvik (NE Norway) and three in Finnish Lapland. For the majority of the elements studied, the concentration level in moss was highest in the Russian catchments on the Kola Peninsula, where the median content decreases with increasing distance to the smelters for As, Co, Cr, Cu, Fe and Ni. The effect of pollution is also seen in the Norwegian catchment and the one in the NE part of Finland. The other Finnish catchments are located in so-called background areas, unaffected by local emissions. In addition to the well-known impact from the biggest Ni-Cu smelters in Europe, this study shows a major element contribution of Al, La, Si, Sr, Th, Ti and V in the moss, obviously connected to dust from open pit mines and tailing disposals in the Kirovsk/Apatity area. The variation between catchments is by far the most important factor for total variation, while seasonal variation for most elements is in general negligible. The content of B, Ca, K, Mn, Rb, Mo, Na, Sb and Zn in feather moss indicates that it must be used cautiously as a biomonitor of the depositions of these elements, due to high local versus regional variability as compared to variability in this region. This finding illustrates the drawbacks of using living sampling media; additionally, mosses are sensitive to pollution and therefore disappear in the most heavily polluted areas. The Cu:Ni ratios show the characteristic pattern (fingerprints) of emissions from Pechenganikel, as distinguished from Severonikel emissions. Compared to earlier studies using moss as a biomonitor, the concentration levels of Cd, Cr, and Pb have changed slightly. The values of Cu, Ni, Fe, V, and Zn are more or less at the same level showing the severe impact of smelters. In the background areas, the concentration levels seem to have decreased slightly during the last decade.

#### INTRODUCTION

The western part of the Kola Peninsula in NW Russia has been a region of major industrial activity such as mining and ore smelting for several decades (Kozlov et al., 1993). One visible result of the substantial emissions of sulphur, aluminium and heavy metals is the increasing damage of the local terrestrial ecosystem as measured by remote sensing, a phenomenon described as «technogenic deserts» by the Russians. These technogenic deserts have become popular as objects for scientific investigation. Several studies have measured the extent of pollution from the industrial centres at the Kola peninsula, e. g. in snow (Jaffe et al., 1995), in air (Sivertsen et al., 1992) and a multi-media survey (Niskavaara et al., 1996).

Carpet-forming mosses have proved to be suitable as bioindicators of inorganic substances at both a local and regional scale, and have therefore been widely used (Rühling and Tyler, 1973; Steinnes, 1995). A monitoring program using moss analysis in northern Europe has been carried out at five

year intervals, in 1985 and 1990, and again in 1995 (Rühling, 1994). Several other regional samplings of terrestrial mosses have been carried out in parts of the Barents region. Mosses were collected in eastern Finnish Lapland in 1982-83 with a sample density of 1 sample per 60 km<sup>2</sup> and repeated in all of Finnish Lapland in 1990 with sampling density of 1 sample/ 300 km<sup>2</sup> (Niskavaara and Lehmuspelto, 1992). Transect sampling has been carried out in Finnish Lapland and the Kola Peninsula (Mäkinen, 1994a and b), and in a national survey of Norway since 1977 (Steinnes et al., 1980). In addition, local scale studies have been carried out in Finnish Lapland (Ukonmaanaho, 1991; Äyräs and Niskavaara, 1992a; Äyräs and Niskavaara, 1992b), and in the border area between Norway, Finland and Russia (Äyräs et al., 1996), and on the Norwegian side of the border area (Schjoldager et al., 1983).

The Central Kola Expedition (CKE), the Geological Survey of Norway (NGU) and the Geological Survey of Finland (GTK) are carrying out an collaborative ecogeochemical project in the western part of the Kola Peninsula, Finnish Lapland and Finmark county in northern Norway. A pilot project was carried out in 1992-93 to harmonise field and laboratory methods and test different sampling media (Chekushin et al., 1993). In 1994, ten different sampling media, including moss, were collected in eight catchments in the Barents region of the three countries (Fig. 6.1.1). The main aims of this study are to evaluate element input and cycling in the catchments, and to form a basis for interpreting regional ecogeochemical mapping of 188,000 km<sup>2</sup> of the Barents region sampled in 1995. For more details and results of the project see its site on the World Wide Web, URL:<http://www.ngu.no/Kola>.

In this paper, we will focus on the chemical composition of terrestrial moss samples, and compare seven selected small areas, limited to catchments in the Barents region. Seasonal, vertical, and local versus regional variations as a result of different sources of the 38 elements are discussed in this paper. The results are linked to previous investigations using moss as biomonitors, and to other analysed media sampled simultaneously at the same localities.

## MATERIAL AND METHODS

### *Study area and sampling*

Eight small areas (hereafter abbreviated as C1 - C8) limited to catchments, 12 - 35 km<sup>2</sup> in size, were selected according to the principles of AMAP (Landers et al., 1995). Four of the catchments are situated in the Kola Peninsula: C1, 20 - 25 km NE of the concentration plant in Zapoljarnyi; C2, Monchegorsk (near the town of Monchegorsk); C3, Kirovsk (near the town of Kirovsk); and C4, Kurka (25 km south of Monchegorsk). One is in Norway, C5, Skjellbekken (near the Russian border south of Kirkenes); and three are in Finland: C6, Kirakka (near the Norwegian border); C7, Naruska (in eastern Finland near the Russian border); and C8, Pallas (in the western part of Finland) (see Fig. 6.1.1 and Tab. 6.1.1). The distance to major industrial centres varies from 5 to more than 150 km, and the distance to the Barents sea varies from 30 km (C1) to more than 300 km (C7). The annual precipitation in 1994, as measured at the nearest meteorological stations, varied from 386 mm (near C6) to 513 mm (near C7). The nature of the catchments was described (Chekushin et al., 1995; Reimann, 1995) and large amounts of field information were collected during sampling (Äyräs and Reimann, 1995).

Altogether, 79 moss samples were collected from 7 of the 8 investigated catchments. The three newest annual shoots of mainly *Hylocomium splendens*, but also *Pleurozium schreberi* were collected twice during 1994 from 2 to 8 sites within each catchment (Tab. 6.1.1). The sampling took place in July and at the end of September through the beginning of October in Russia (C1, C3 and C4), at the end of May and September in Norway (C5), and in June and August in Finland (C6 -



C8). Every effort was made to avoid mixing the samples with soil or litter, and sites under trees, bushes or dense vegetation were avoided wherever possible. The samples were collected as described in detail by Äyräs and Reimann (1995) in accordance with methods used in the European moss project (Rühling, 1994).

The samples were pre-dried in the field and then fully air-dried at  $< 40^{\circ}\text{C}$  and milled after arrival to GTK's laboratory in Rovaniemi. The moss samples (0.500 g) were dissolved with 10 mL of concentrated nitric acid in a microwave oven, and diluted to 50 mL with water (US EPA standard 3051) before being analysed for 38 elements by ICP-AES, ICP-MS and cold vapor AAS at GTK's accredited laboratory (Niskavaara, 1994).

## RESULTS

### *Concentration levels*

The results of the analysis of all 38 elements are summarised in Table 6.1.2. Only for Bi, Be, Li, La, Se, Sb, Th and U were more than 25% of the samples below the detection limits. The variations of median between the 7 catchments varied from less than 2 (Zn<K<Ca), through 2-5 (Hg<P<S<B<Rb<Pb<Mg<Mn<Si<Cd) up to 32 (Cu), 74 (Co) and even 185 (Ni) orders of magnitude. More or less the same order of the elements was found by dividing the maximum by the minimum values in the dataset. In general, the essential nutrient elements for plants varied less than the known pollutant in the area, both between and within the catchments.

ANOVA analysis (Krumbein et al., 1965) was conducted to estimate the influence of the three main factors on the variability of the element content in the moss samples, namely between catchments (R1), between sampling points within each catchment (R2) and seasonal variation (R3).

The results presented in Figure 6.1.2 indicate that the variation between the catchments (R1) is by far the most important factor influencing to the variation of element levels. The seasonal variation for most elements is in general very low. The content of the nutrients P, K, and Zn increases for several of the studied areas (C5, C1, C3) towards the autumn, while the opposite happens with Hg in C3, C4 and to an extent in C5. However, the total variations of these elements in the dataset are narrow (within the range of  $\text{max./min} < 5$ ). More than 10 -17 % of the relative variability is due to the variability within the catchment (R2) for Ag, As, Cd, Cr, Co, Cu, Fe, Mg, Ni and S. Less than 75 % of the variability can be explained for B, Mn, Mo, Na, Rb and Sc after having included factor interactions in the same ANOVA analysis, and 75 - 80 % can be explained for Bi, Ca, P, Sb, Se and Si. Regional patterns for these elements are therefore affected by uncertainties, and must be evaluated cautiously.

The thorium and uranium contents are mostly below the analytical detection limit (Tab. 6.1.2). Elevated levels of both Th and U are found in C3, C4 and C6, while in C8 only elevated U is found. In two locations in the Kirakka catchment, U levels are extremely high (0.49 and 0.62 ppm).

### *Comparison between catchments*

It was only possible to collect a few moss samples ( $n=4$ ) from C1 and C4, and no moss was found at all in C2 as a result of the heavy loading of pollutants, which causes vegetation damage in these catchments (Tab. 6.1.1). Due to the lack of samples, exact comparison between catchments is somewhat difficult. It is clear that for the majority of the studied elements, the concentration level in mosses is highest in the catchments at the Kola Peninsula (C1 - C4) (Tab. 6.1.2). The maximum contents were found on the Russian side, except for Mn and Zn (C5), Mo (C6), Na (C7), Rb (C8), Tl (C6, C8) and U (C6), all found in the Norwegian or Finnish catchments.

The catchments run C1 or C4 > C3 > C5 > C6 > C7 > C8 in decreasing order of median content of the main pollutants As, Co, Cr, Cu, Fe and Ni (Fig. 6.1.3), which have an internal correlation of 0.86 - 0.97 for the log-transformed data. This catchment order also corresponds to increasing distance to the main smelters.

The median values of the macro-nutrients calcium and magnesium are more or less similar for all the catchments. The values of Ca and Mg are decreasing in the order C5 (Skjellbekken), C6 (Kirakka), C8 (Pallas) and C7 (Naruska) as shown by boxplot comparisons in Figure 6.1.4. The median values of manganese are highest in Norway and Finland. The median values of sodium, potassium and phosphorus are quite similar, reaching the highest values either in Zapoljarniy or in the Kirovsk catchment.

For Ca, the variation within each catchment is higher than that between catchments. The highest K, Na and P contents are in Zapoljarniy (C1) and Kirovsk (C3), mainly due to the dust from the open pit mines. The same situation is also found for Si. High contents of La, Sr, Ti, V, and Y found in the Kirovsk and Kurka catchments are due to dust from several open pit mines in the Kirovsk area (Chekushin et al., 1995).

The influence of both mining and smelting activities are severe and seem to overwhelm most of the other element sources in the Russian areas. In order to sort out the most severe element contributors in the area and group the elements, Principal Factor Analysis (SMC) by VARIMAX rotation was done with Nlog transformed data for 21 selected elements. The different factors are shown as x-y plots in Figure 6.1.5, both for the whole dataset (explanations F1-F3: 0.8249) and F1:F2 for C5-C8 (explanations F1-F3: 0.757). Only factor loadings higher than the absolute value of 0.5 ( $> |0.5|$ ) are entered. The elements Ag, Ba, Mg (+/-), Rb (+/-) and S enter into factors indicating several sources. For the whole dataset, F1 seems to represent mainly an anthropogenic factor mixed with geogenic dust, F2, a geogenic factor, and F3, a biological/nutrient factor. The factor analysis distinguishes the Russian catchments from each other. C5 and C6 end with an intermediate pollution status, while C7 and C8, with increasing distance to the pollution sources, have background conditions where local geological features are important and are therefore hard to distinguish from one another.

Ni versus Cu or Co as an x-y-plot shows good correlation. In C6 and C5, the Cu:Ni ratio closely resembles the pattern in Zapoljarniy (C1), showing the characteristic signs or fingerprints of emissions from Pechenganikel as distinguished from Severonikel emissions (Fig. 6.1.6). Sr vs. Ti outlines the impact of dust in the Russian catchments and in particular C3, while Mn vs. Zn have a quite narrow variation and underline the peculiarities in C5 (see Fig. 6.1.6).

Enrichment factors (Ef) are calculated as the median of each catchment divided by the median of catchment 7 (considered to be a representative background catchment due to typical alkaline bedrock and its position > 150 km away from the major smelting industry). Ef for 12 selected elements are presented in Fig. 6.1.7, showing that C7 and C8 are representative as background catchments with the overall lowest element levels, except for Rb. Figure 6.1.7 also illustrates the broad span in variability of element content between elements.

## DISCUSSION

Most mosses growing on the forest floor receive their nutrients only from leached dust-fall (dry depositions) and precipitation (wet depositions). Several studies have shown that a survey of the metal concentration in mosses is a useful way of identifying sources of airborne pollution and



mapping the extent of deposition of 8 - 9 heavy metals (Rühling et al., 1994) up to the 40 elements surveyed in Norway (Steinnes et al., 1993). Our moss results show that the selected catchments range from among the most heavily polluted areas in Europe (C1 and C4) for Ni, Co and Cu, through the intermediately polluted areas as C3, C5 and C6, to some of the most pristine background areas in Euro-Arctic territories (C7 and C8) in terms of deposition of heavy metals. This fact is clearly seen by comparing both range, median and enrichment factors from the catchments with previous studies (op. cit.). Such broad variation within distances of 150 km make this part of Europe an interesting area for research.

Not surprisingly, it is obvious that the Russian catchments are strongly affected by both the surrounding smelters and concentration plants and by dust from open pit mines. These catchments (C1-C4) have the highest enrichments for almost all the analysed elements, roughly similar to the airborne depositions found in the snow (Äyräs et al., 1995) and rain (Reimann et al., 1996a) analysed from the same area. C1 (Zapoljarnyi), the catchment closest to the Barents sea, and 10 km NE of the concentration plant has the highest median values of B, Cr, Fe, Hg, K, Mg, Ni, P and S.

When using the moss technique, it is important to consider sources and mechanisms other than atmospheric pollutants which may contribute significantly to the concentrations observed (Steinnes, 1995). In addition to the anthropogenic air pollution, the marine factor, transfer to moss by leaching from living or dead plant material, and geogenic input of mineral particles as windblown dust from local soil, especially in areas of sparse vegetation cover, affect the moss composition. The effect of pollution is seen even in the Skjellbekken catchment (C5, Norway), as elevated contents of As, Cd, Co, Cr, Cu, Fe, Ni, V, Zn and S, and in Kirakka catchment (C6, NE Finland), as Co, Cr, Cu, Fe, Ni, Zn and S. The other Finnish catchments, Naruska (C7) and Pallas (C8), are clearly in background areas, showing maximum contents for only a few elements. The high concentrations of Mn and Zn found in C5 probably indicate a certain vegetation type (e. g. localities more dominated by *Empetrum nigrum*) or sampling closer to certain higher plants than was done in the other catchments. The notably high Rb content in the most remote catchment, C8, is probably also due to contributions by higher plants (Steinnes et al., 1992).

Regional studies using moss as a biomonitor imply that randomly chosen sampling sites should be representative for an area of 300 - 1000 km<sup>2</sup> each. Local variability must be low compared to the regional variability. This is the case for elements like Al, V, Ti and Sr, among others. Several elements have very high regional variability but also high local variability (e.g. Fe, Ni, Cr, Ag, Co). However, the following elements have less than 60 % of the variability explained by regional variability between the selected catchments: Ca, Mn, Rb, Mo, Na, B, Sb, K, Zn and Hg (known macro- and micronutrients for plants, see Fig 2). The lack of correlation between several of these elements in vegetation taxa and loading is discussed and handled by Bogatyrev et al. (1996). In general, depositions of nutrients seems to be underestimated by use of moss as biomonitor. Several elements have essential functions in the moss, and selective element uptake occurs (Berg et al., 1995).

A distinct marine factor is not really distinguished in our study, despite our selection of areas ranging from 30 to 300 km away from the Barents sea. This is probably due to several factors: (1) several marine elements (e.g. B, Ca, K, Na, Mg) also are part of the emission spectre or geogenic dust as revealed by enriched content in C4 (see Tab. 6.1.2) and snow sampled near the industry of Nickel and Zapoljarnyi (Reimann et al., 1996b), and (2) Na, for instance, is easily exchanged with other cations, as found in coastal areas of Norway (Steinnes et al., 1992).

Of our selected sites, C1, C3 and C4 in particular are potentially affected by unknown portions of geogenic dust from vegetation free areas, as a result of vegetation damage or operating open pits and mine tailings within a radius of less than 20 km. The highest median and maximum contents of Al, La, Si, Sr, Ti, Th and V, found in C3 (Kirovsk catchments), are clearly due to dust from open-pit mines only 5 - 15 km away. For these elements, in addition to Be, Sb, Sc, U and Y, the geogenic dust factor seems to be far more important than a possible contribution from smoke stacks! The mosses are not washed before being analysed, and microminerogenic material probably plays an important role in the analysis, especially for mosses from areas of sparse vegetation coverage. Even small areas of this type can play an important role locally in moss chemistry. This fact should be stressed more in the field manual for the forthcoming use of moss as biomonitor for atmospheric pollution.

The absorption and retention of heavy metals in *Hylocomium splendens* generally follows the order Cu or Pb>Ni>Co>Zn (Rühling and Tyler, 1973). However, in the case of the metal cations which are less strongly absorbed, the competition for exchange sites with other cations present in precipitation may significantly influence the uptake and retention in the moss (Gjengedal and Steinnes, 1990; Berg et al., 1995). Our results show heaviest enrichment of Ni followed by Co and Cu. The relatively high enrichment of Co compared to Ni and Cu is also found in rain (wet deposits) sampled simultaneously (Reimann et al., 1996). The explanation could be that Co is deposited in a more soluble phase than Ni and Cu, since Ni and Cu have 10 - 25 times higher emission levels than Co according to official data from the smelters (see Tab. 1 in Reimann et al 1996a).

During the snow melting period, transport of soluble compounds from the soil into the moss tissue may affect the retention of metal cations in the moss. Steinnes (1995) considers the sampling dates of mosses as important. In contrast, our results show that the seasonal variability is negligible in regional studies for the majority of the elements. The seasonal effects seem only to be noticeable for Hg, K, P and Zn, all elements that have a narrow overall range. K and P are macronutrients for plants, and the increasing content towards the autumn probably reflects growth. Even though we collect the three outermost shots of moss, the content does not really give an integrated picture, but is affected by growthrate, etc. No correlation was found between altitude and content, probably due to low relief in our selected areas.

The metal ratios, e.g. Cu or Co to Ni content in moss, can be used as fingerprints of the pollution sources. The same phenomenon is also present for several heavy metals in snowsamples from the catchments (Äyräs et al., 1995). This is due to the fact that both materials are useful indicators of airborne pollution. The fingerprints are clearly seen in Zapoljarny, Skjellbekken and Kirakka catchments, where the ratios of Cu/Ni resemble each other closely, and distinguished from the ratios in C3 and C4 affected by the Severonikel smelter.

#### COMPARISON TO EARLIER STUDIES

The Moss Atlas of Northern Europe, compiled in 1990, shows the regional distribution of 9 heavy metals (Rühling et al., 1994). Our results show slightly lower Cd and Cr concentrations in the Monchegorsk and Kirovsk area, while in Norway and Finland the level is more or less the same. There has been a general decline in the Cd deposition since 1985, and the trend seems to be continuing. This is probably due to a decrease in smelting activity in the Kola Peninsula and changes in the origin of the ores used. However, the analytical data from different studies was derived through different analytical procedures, making exact comparisons difficult.

For Cu, Ni and V, the concentration levels we found were roughly the same as in the study cited above. The exception is the Kirovsk catchment, where the level of Cu in the current study was

about 30 µg/g, while levels in the Moss Atlas were well in excess of 30ug/g. The Cu and Ni contents in mosses in Skjellbekken and Kirakka are at about the same level. The effect of emissions is clearly seen in mosses collected by GTK in 1982-83 and 1990 in NE Finland, although the area with highest levels of these main pollutants has decreased in size (Niskavaara and Lehmuspelto, 1992). In the background areas (C7 and C8), the concentration levels of both Cu and Ni closely resemble the levels found in the European moss project, only slightly affected by long transported airborne pollution, as in the southern part of Norway (Steinnes et al., 1993)

The level of Pb concentrations in our study is lower than in the Moss Atlas (Rühling, 1994). There has been a general decline in the Pb deposition since 1985 due to increased use of low-lead petrol, as was found earlier by mapping projects in Lapland, carried out by GTK in 1982-83 and 1990 (Niskavaara and Lehmuspelto, 1992). So far, no published data have been found concerning the regional extend of Ag or Ti contents in feather moss from this area, so this enrichments of Ag in C4 (emissions from Severonikel) and Ti in C3 (open pit dust) are believed to be the first of their kind to be discovered.

## CONCLUSIONS

- (1) Our selected small areas (catchments) represent both some of the most heavily polluted areas (for levels of As, Cr, Cu, Fe and Ni in the Monchegorsk and Pechenganikel area) and some of the cleanest areas of Europe (for levels of Cd, Fe and Zn in Finnish Lapland), when comparing our results with previous studies carried out in Europe.
- (2) Detailed sampling of 1 - 2 sample pr. 5 km<sup>2</sup> is not necessary and seasonal variation is negligible in comparison to the regional variability for the majority of the analysed heavy metals.
- (3) Geogenic dust from local sources has a major influence on moss chemistry for elements like Al, La, Si, Sr, Th, Ti and V in mosses from the mining area of Kirovsk. Element contents in mosses reflect an uncertain mixture of both dry and wet deposits, entered into the samples by leaching from dry deposits or as microparticles in the unwashed moss samples.
- (4) Sampling distance to surrounding trees or special vegetation types are probably the reason for enriched content of especially Mn and Zn in Skjellbekken (C5 in Norway), and probably also for Rb in the background area (C8).
- (5) The content of B, Ca, K, Mn, Rb, Mo, Na, Sb and Zn in feather moss must be used critically as a indicator of the depositions of these elements. Factors other than anthropogenic emissions, like growth rate, competition with other elements, etc., seem to play a more important role for these elements in this region. Depositions of several elements are underestimated when using moss as biomonitor, illustrating the drawbacks of using living sampling media. Still, moss is a suitable bioindicator for a variety of elements in regional scale, despite the fact that the most commonly used species disappear in the most heavily polluted areas.

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## TABLES

Table 6.1.1 . Some key figures for the eight studied catchments.

Table 6.1.2. Summarised data of minimum (min.) median (med.) and maximum (max.) from each catchment, where the maximum median and single max. content are outlined by frames.

## FIGURES

Figure 6.1.1. Location of the studied catchments in the centre of the Barents region, Northern Europe. The outer frame illustrates the borders of the ecogeochemical mapping carried out in 1995. The eight studied catchments are shown as no. 1-8: 1: Zapoljarnyi, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, 8: Pallas. The letters indicate the following industry towns: **Ki**: Kirkenes, **N**: Nikel, **Z**: Zaplojarniy, **M**: Monchegorsk, **K**: Kirovsk, **Mu**: Murmansk.

Figure 6.1.2. Variability analysis (ANOVA) illustrated as estimates of the influence of three factors (R1-R3) for the analysed elements in feather moss sampled in two periods in 1994 at seven catchments in the Barents region.

Figure 6.1.3. Boxplot diagrams showing the variations of log transformed values of As, Co, Cr, Cu, Fe and Ni for the different catchments. C1 - C8 refers to catchment numbers (Fig. 6.1.1 and Table 6.1.1 ). Lines show the concentration in each sample from C1 (n=4) and C4 (n=4).

Figure 6.1.4. Boxplot diagrams showing the variations of Ca, K, Mg, Na, P, and Si for the various catchments. C1 - C8 refer to catchment numbers (Fig.1 and Tab 1). Lines shows the concentration in each sample from C1 (n=4) and C4 (n=4).

Figure 6.1.5. X-Y plots of factor analysis for the whole dataset C1, C3 C8 (F1:F2, F1:F3 and F2:F3) and for the remote catchments C5-C8 (F1:F2). C1 - C8 refer to catchment numbers in Fig. 6.1.1 and Table 6.1.1 .

Figure 6.1.6. X-Y plots of Ni vs. Cu and Ni vs. Co (impact of smelters), Sr vs. Ti (impact of geogenic dust) and Mn vs. Zn (biological interactions).

Figure 6.1.7. Enrichment factors for 12 selected elements, using the Naruska catchment (C7) as background (x-axis). Calculated as median from each catchment divided by median of C7.

Table 6.1.1 . Some keyfigures of the eight studied catchments.

No	Name	Co-or-dinates - outlet	Size (km <sup>2</sup> )	Samplin g altitudes (m.a.s.l.)	Moss samples total (loc.)	Moss spp.	Distance and position to nearest major industry (emission sources)
C1 (Ru)	Zapoljarnyi	69°27' N 31°03' E	19.0	27 - 74	4	(2) H, P	10 km NE of Zapoljarnij (roasting) 30 km W of Nikel (Ni - Cu smelter)
C2 (Ru)	Monchegor sk	67°50' N 32°54' E	22.4	128 - 507*	0*	*	5 km S of Severonikel (Ni - Cu smelter)
C3 (Ru)	Kirovsk	67°32' N 33°48' E	20.0	290 - 495	14	(7) H	2 km S of Kirovsk (open pit mines) 5 - 15 km N and W of several ap-ne
C4 (Ru)	Kurka	67°41' N 32°50' E	20.5	156 - 213	4	(2) H, P	25 km SW of Severonikel 20 km W of Apatity (ap-ne)
C5 (N)	Skjellbekke n	69°21' N 29°27' E	34.6	75 - 186	15	(8) H, P	30 km W of Nikel 50 km SW of Kirkenes (Fe- smelter)
C6 (Fin)	Kirakka	69°35' N 28°52' E	11.9	115 - 220	14	(7) H	60 km NW of Nikel 50 km W of Kirkenes
C7 (Fin)	Naruska	67°21' N 29°22' E	20.2	265 - 350	14	(7) H	65 km SW of Kovdor ( div. mines) 150 km W of Kandalaksha (Al-plant)
C8 (Fin)	Pallas	68°09' N 23°52' E	24.4	320 - 385	14	(7) H	150 km W of Kiruna (Fe- mines) 290 km SW of Nikel

Abbreviations: Moss spp; H; *Hylocomium splendens*, P; *Pleurozium schreberi*, loc.; Localities Ru; Russia, N; Norway, Fin: Finland, ap-ne; Apatite - nephelin industry

\* Please note that it was not possible to find any moss in C2 as a consequence of severe vegetation damage.

	C8			C7			C6			C5			C4			C3			C1		
	Min	Median	Max	Min	Median	Max	Min	Median	Max	Min	Median	Max	Min	Median	Max	Min	Median	Max	Min	Median	Max
Ag	<0.02	<0.02	0.03	<0.02	0.03	0.04	<0.02	0.03	0.03	0.03	0.04	0.07	0.36	0.54	0.85	0.07	0.10	0.14	0.07	0.09	0.10
Al	50	120	280	90	120	250	50	95	280	90	120	250	540	725	780	940	1345	1800	280	305	320
As	0.08	0.13	0.21	0.14	0.17	0.24	0.18	0.25	0.38	0.34	0.43	0.61	0.87	1.39	3.05	0.38	0.55	0.74	0.71	0.74	1.08
B	0.7	1.12	2.88	0.73	1.2	1.69	1.73	2.51	3.93	1.13	1.85	2.92	1.36	2.37	9.79	1.38	2.13	4.82	2.22	2.72	3.79
Ba	10.6	14.6	32.9	10.3	18.25	26.2	12	20.8	24.5	11.9	19.6	35.1	107	181	255	22.5	32.35	49.7	16.5	18.1	20
Be	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.18	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.2	<0.1	<0.1	<0.1
Bi	<0.02	<0.02	0.15	<0.02	<0.02	0.03	<0.02	<0.02	<0.02	<0.02	0.03	0.04	0.09	0.115	0.23	0.03	0.05	0.06	<0.02	0.03	0.05
Ca	1930	2465	3160	2040	2390	3490	2360	2750	3680	2080	2930	4150	3890	4360	8050	1980	2685	3930	2960	3110	3910
Cd	0.06	0.08	0.14	0.06	0.08	0.09	0.04	0.06	0.09	0.09	0.1	0.15	0.22	0.285	0.39	0.08	0.12	0.23	0.13	0.15	0.15
Co	0.08	0.13	0.33	0.12	0.16	0.18	0.26	0.40	0.53	0.78	0.91	1.48	5.62	9.675	18.9	0.58	0.86	1.17	7.61	8.97	9.15
Cr	<0.4	<0.4	0.71	<0.4	<0.4	0.57	<0.4	0.50	0.7	0.44	0.81	1.44	1.46	2.05	3.15	1.04	1.54	2.37	5.52	5.64	6.18
Cu	3.57	4.24	4.67	4.95	5.42	6.35	8.29	9.01	12.2	16	20.2	30	118	136	291	17.8	24.7	31.2	115	120	140
Fe	60	80	160	70	100	130	80	130	250	180	250	440	480	615	790	430	570	710	1740	1825	2010
Hg	0.06	0.08	0.09	0.04	0.07	0.09	0.05	0.13	0.15	0.05	0.13	0.16	0.07	0.105	0.15	0.06	0.11	0.14	0.09	0.12	0.15
K	3210	3755	4410	2950	3585	4110	2770	3215	3890	2770	4000	4780	2760	5235	7750	3520	4405	7030	4850	6005	7150
La	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	1.8	<0.7	<0.7	<0.7	2.1	4.3	8.1	4.2	5.15	10.5	<0.7	<0.7	<0.7
Li	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7	<0.7
Mg	710	830	1110	680	715	820	780	925	1230	640	940	1240	840	970	1210	470	625	1110	1800	1990	2190
Mn	220	406	532	179	475	758	304	405	852	342	643	1020	130	180	408	174	260	373	252	257	282
Mo	0.06	0.07	0.1	0.06	0.07	0.08	0.04	0.12	1.46	0.04	0.06	0.08	0.38	0.50	0.83	0.19	0.31	0.36	0.08	0.09	0.1
Na	<10	<10	70	<10	<10	170	<10	15	60	<10	20	70	<10	85	160	30	65	90	40	80	120
Ni	1.1	1.41	3.48	1.94	2.23	2.93	7.02	8.62	12.3	22.3	26.1	41.7	98.5	184	241	11.5	16.1	19.6	225	258	261
P	615	1140	1510	821	1005	1090	581	774	1400	620	1210	1520	906	1340	2080	1290	1615	2500	1520	1790	2060
Pb	2.29	2.84	5.89	2.98	3.56	4.29	1.46	2.22	3.05	2.07	2.69	4.09	6.07	6.62	9.61	3.93	6.43	8.82	2.36	2.45	2.60
Rb	11.6	17.5	22.7	8.08	11.2	19.6	4.97	8.67	14.9	5.81	10.8	17	1.66	6.59	9.19	11.3	14.8	21.6	5.51	10.1	14.9
S	640	760	839	676	714	799	687	757	886	647	884	1040	1120	1190	1720	758	947	1370	1350	1470	1610
Sb	<0.02	0.03	0.05	0.03	0.04	0.05	<0.02	0.03	0.05	0.03	0.04	0.07	0.13	0.16	0.27	0.06	0.1	0.15	0.06	0.12	0.20
Sc	<0.1	<0.1	0.1	<0.1	<0.1	0.1	<0.1	<0.1	0.1	<0.1	<0.1	0.1	<0.1	0.08	0.1	<0.1	<0.1	0.2	<0.1	0.10	0.10
Se	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	1.2	<0.5	<0.5	<0.5	<0.5	<0.5
Si	40	60	120	60	70	150	40	70	150	40	70	280	70	155	190	150	215	280	120	140	160
Sr	3.25	6.02	11.8	5.86	9.63	26.9	3.72	7.86	19.1	3.42	6.15	7.93	13.2	21.1	34.9	65.1	121	154	10.5	14.7	15.5
Th	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	0.05	<0.04	<0.04	<0.04	<0.04	<0.04	0.04	0.09	<0.04	0.10	0.13	<0.04	<0.04
Ti	1.8	2.5	3.8	2.5	3.35	5.3	1.7	3.1	4.5	3.1	4.8	10	12.9	19.7	26.3	34.2	53.7	90	18.3	19.9	23.3
Tl	<0.01	0.03	0.18	0.02	0.03	0.06	0.02	0.04	0.11	<0.01	0.02	0.03	0.02	0.025	0.04	0.02	0.04	0.07	<0.01	0.01	0.02
U	<0.01	<0.01	0.02	<0.01	<0.01	<0.01	<0.01	0.05	0.62	<0.01	<0.01	<0.01	0.07	0.1	0.24	0.07	0.09	0.1	<0.01	<0.01	<0.01
V	0.67	0.95	1.6	1.04	1.25	1.56	0.73	1.05	1.31	1.06	1.23	1.65	4.45	4.68	6.54	7.83	12.8	19.1	2.95	3.06	3.16
Y	<0.1	<0.1	0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.3	<0.1	<0.1	<0.1	0.3	0.75	1.8	0.7	0.95	1.4	<0.1	<0.1	0.1
Zn	26.6	34	47.4	20.4	26.6	39.5	18.7	29.4	43.6	22.1	38.3	90.7	24.7	30.6	37	18.5	32.2	47.9	24.6	35.5	43.9

Table 6.1.2. Summarised data of minimum (min.) median (med.) and maximum (max.) from each catchment, where the maximum median and single max. content are outlined by frames.



*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
 Catchment locations

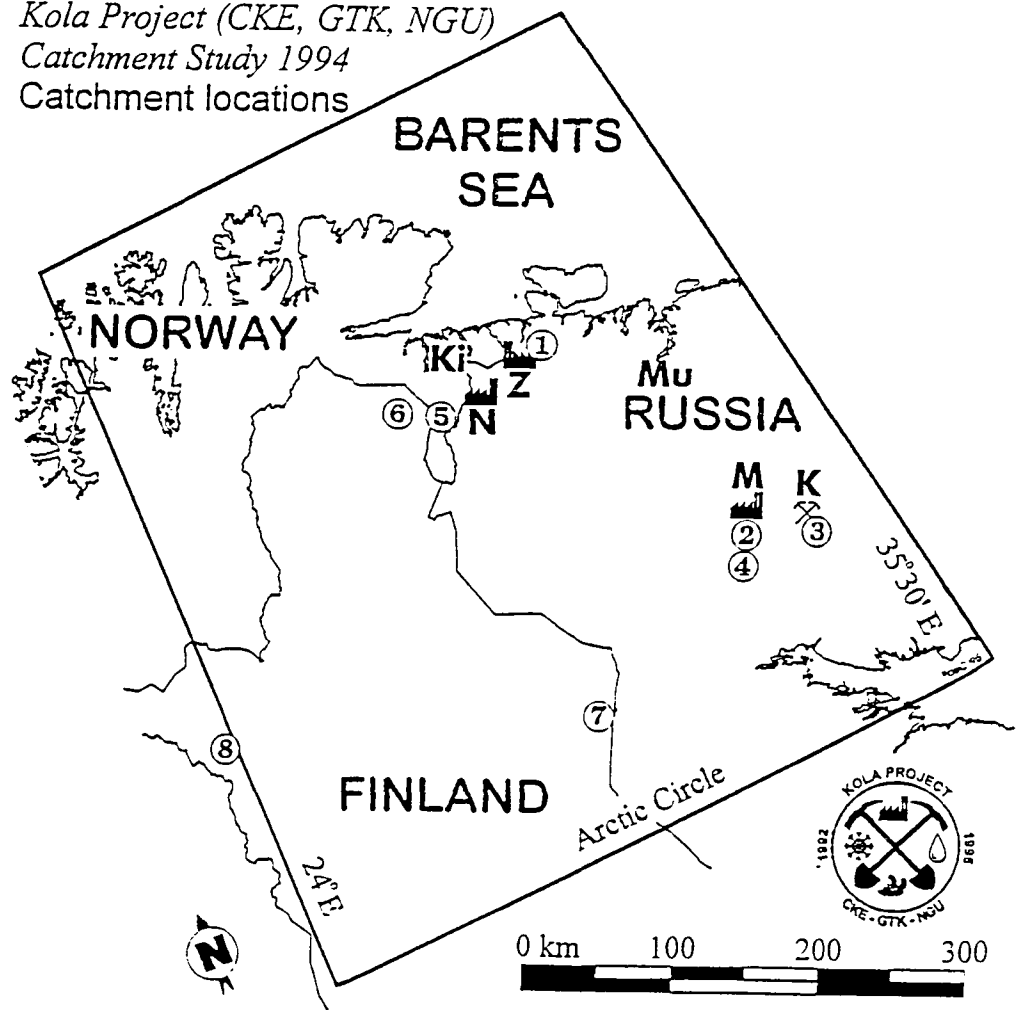


Figure 6.1.1. Location of the studied catchments in the centre of the Barents region, Northern Europe. The outer frame illustrates the borders of the ecogeochemical mapping carried out in 1995. The eight studied catchments are shown as no. 1-8: 1: Zapoljarnyi, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, 8: Pallas. The letters indicate the following industry towns: **Ki**: Kirkenes, **N**: Nikel, **Z**: Zaplojarniy, **M**: Monchegorsk, **K**: Kirovsk, **Mu**: Murmansk.

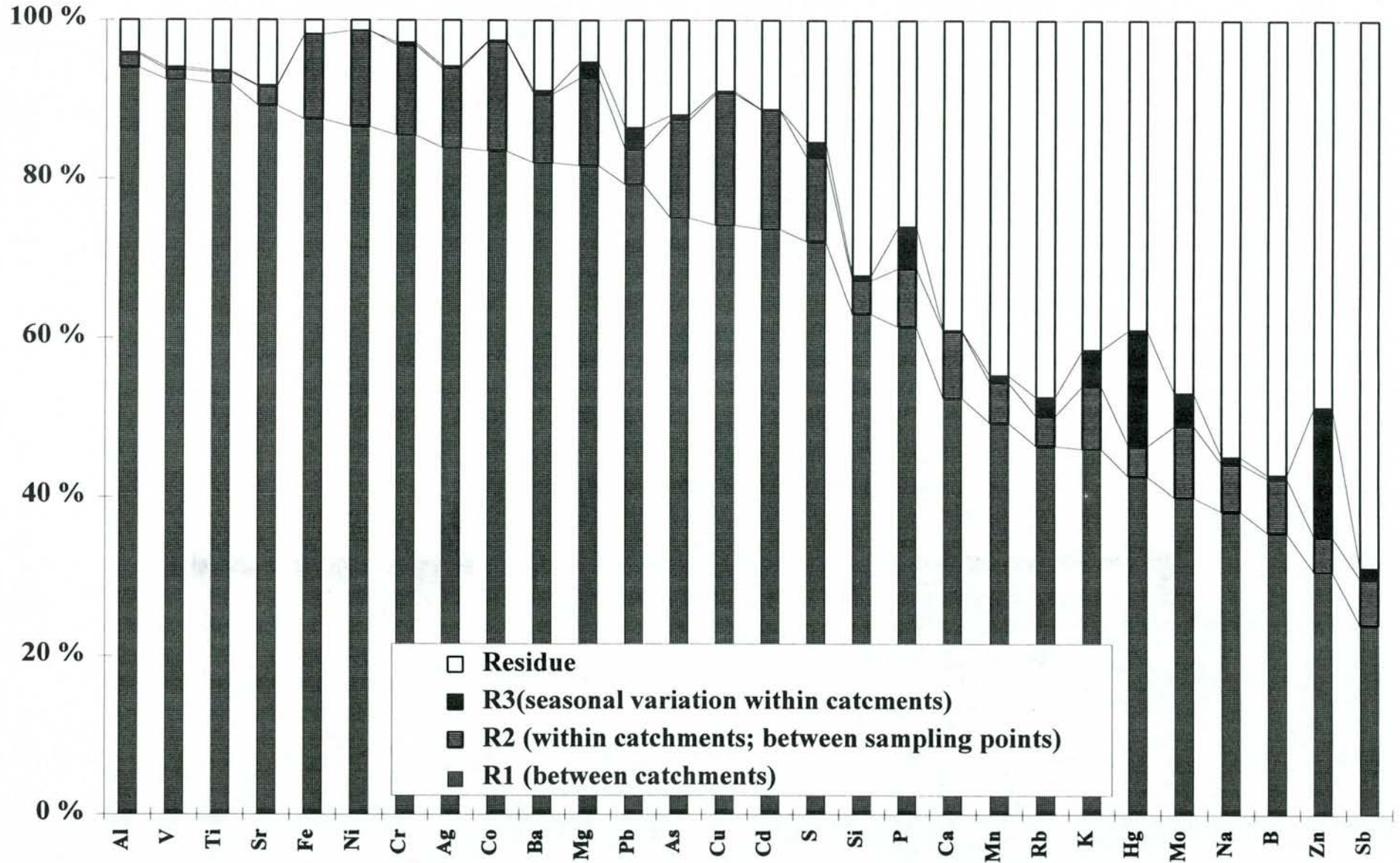


Figure 6.1.2. Variability analysis (ANOVA) illustrated as estimates of the influence of three factors (R1-R3) for the analysed elements in feather moss sampled in two periods in 1994 at seven catchments in the Barents region.

Figure 6.1.3. Boxplot diagrams showing the variations of log transformed values of As, Co, Cr, Cu, Fe and Ni for the different catchments. C1 - C8 refers to catchment numbers (Fig. 6.1.1 and Table 6.1.1). Lines show the concentration in each sample from C1 (n=4) and C4 (n=4).

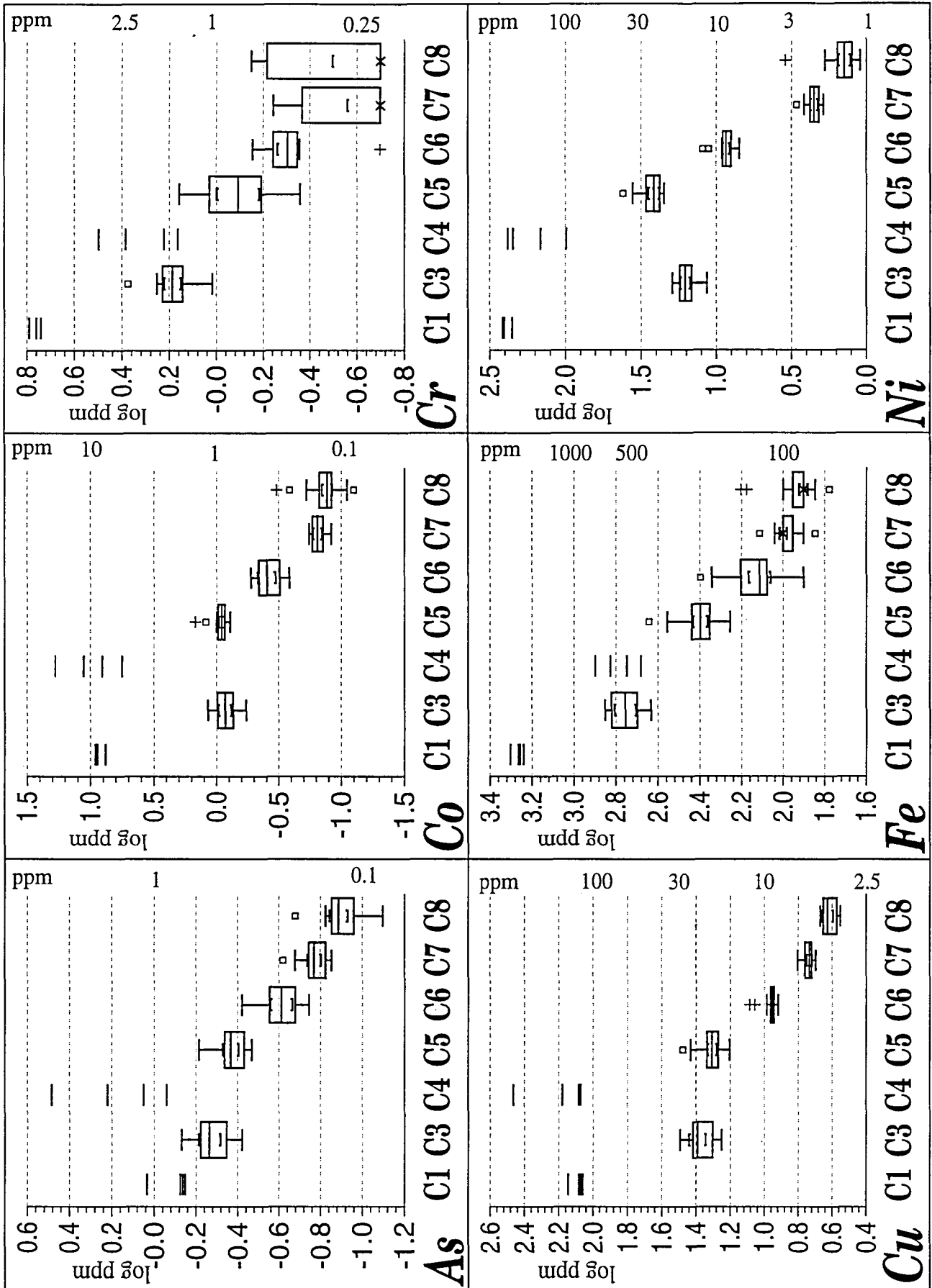


Figure 6.1.4. Boxplot diagrams showing the variations of Ca, K, Mg, Na, P, and Si for the various catchments. C1 - C8 refer to catchment numbers (Fig.1 and Tab 1). Lines shows the concentration in each sample from C1 (n=4) and C4 (n=4).

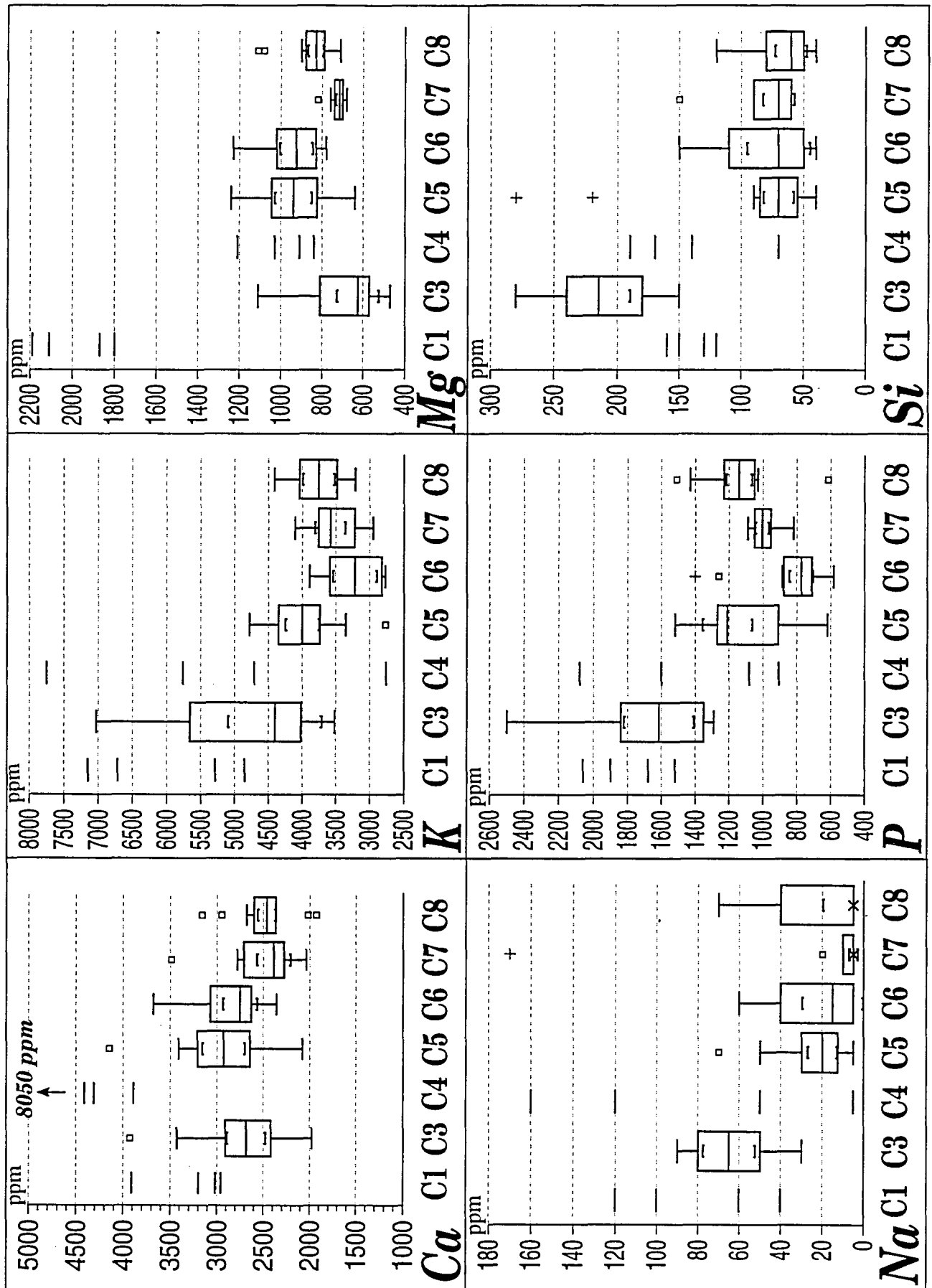


Figure 6.1.5. X-Y plots of factor analysis for the whole dataset C1, C3 C8 (F1:F2, F1:F3 and F2:F3) and for the remote catchments C5-C8 (F1:F2). C1 - C8 refer to catchment numbers in Fig. 6.1.1 and Table 6.1.1 .

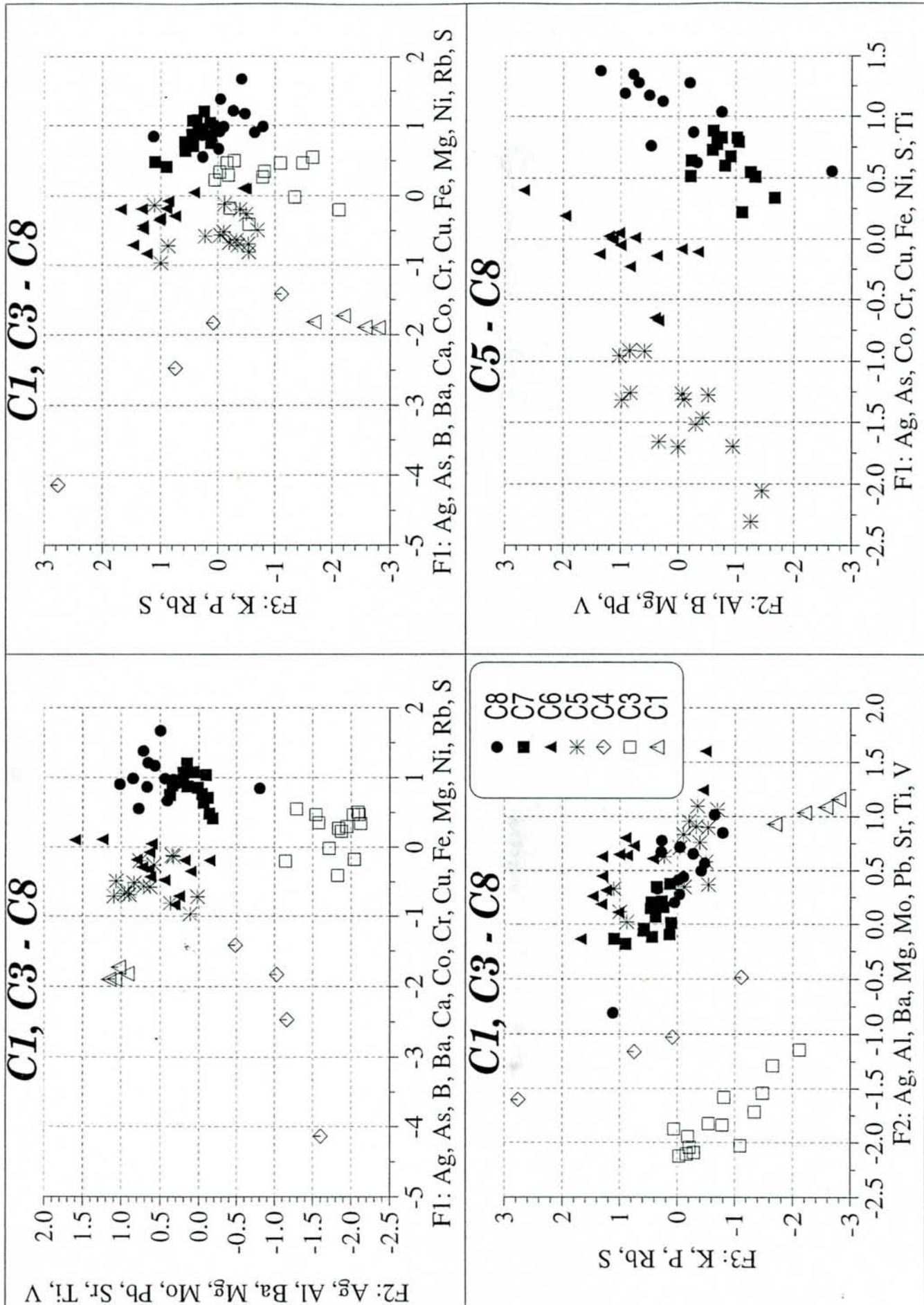




Figure 6.1.6. X-Y plots of Ni vs. Cu and Ni vs. Co (impact of smelters), Sr vs. Ti (impact of geogenic dust) and Mn vs. Zn (biological interactions).

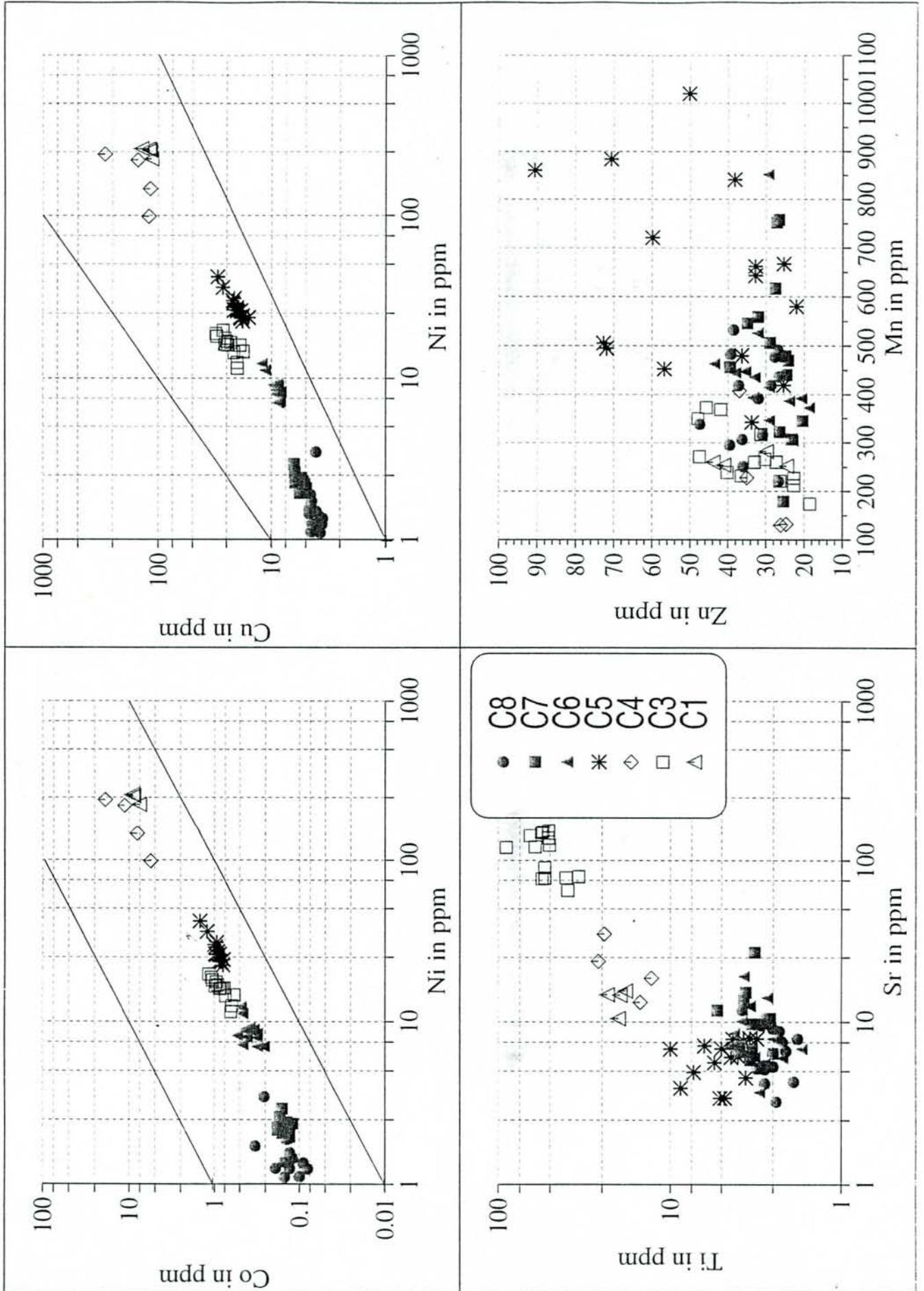
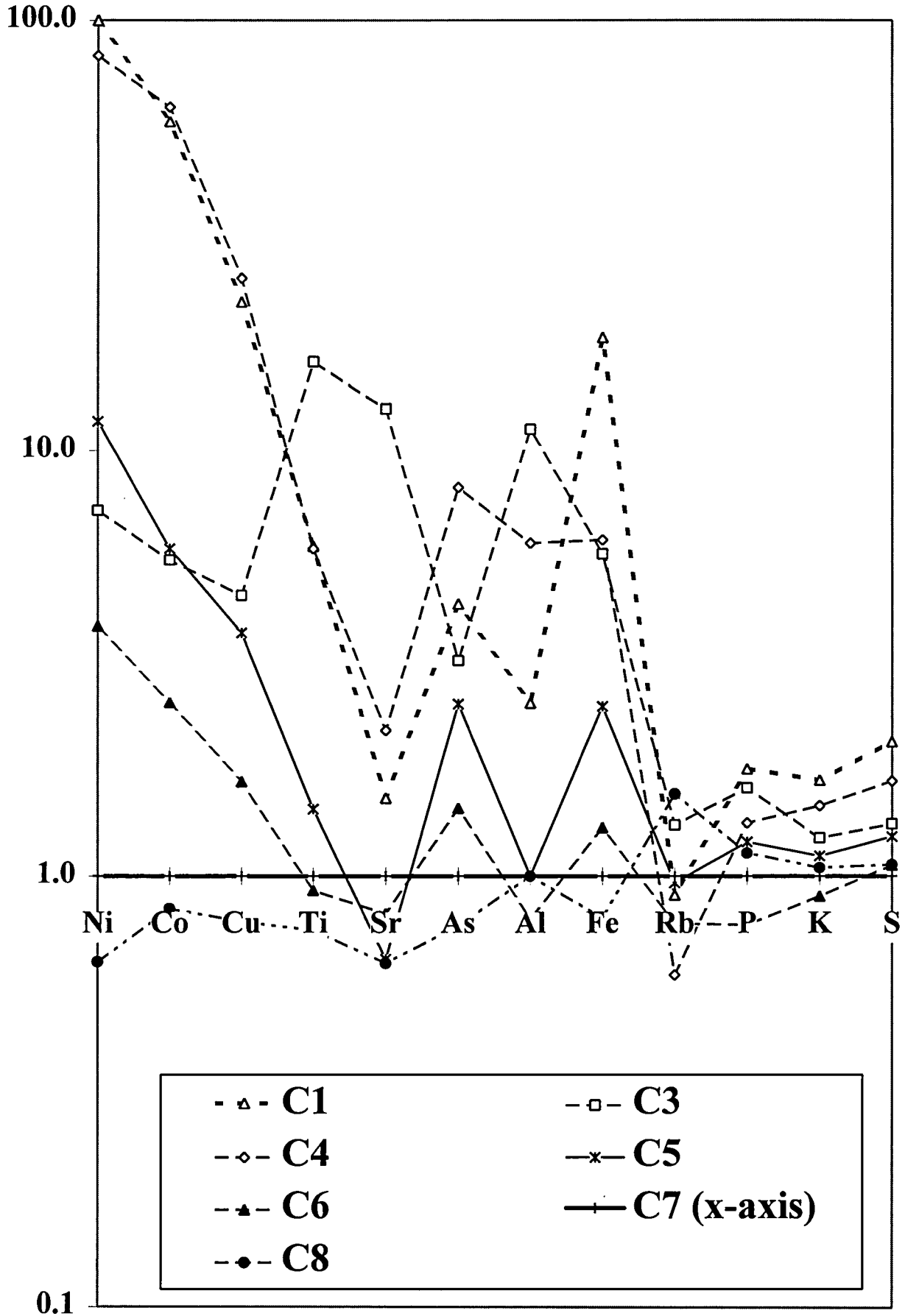


Figure 6.1.7. Enrichment factors for 12 selected elements, using the Naruska catchment (C7) as background (x-axis). Calculated as median from each catchment divided by median of C7.



## 6.2 Ground Vegetation (crowberry, lichens and mosses) as bioindicators of atmospheric pollution in the Barents Region

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### ABSTRACT

The relationship between the chemical composition of three vegetation taxa and annual element depositions has been studied in eight selected catchments located on the Kola peninsula (Russia) and in adjacent areas of Finland and Norway. All three vegetation taxa are strongly affected by the main pollutants in the Barents region, and moss shows the highest sensitivity to airborne contaminants. However, in the areas with the heaviest depositions, moss seems to reach saturation for As, Co, Cu and Ni. No correlation was found between depositions and uptake in the bioindicators for Ca, Cd, Mo and Zn, indicating the importance of essential functions, other sources or a lack of deposition sensitivity. Deposition sensitivity limits are found for uptake of at least Al, Co, Cr, Mo, Ni, S, Sr and V in the moss. Crowberry survives even in the so-called technogenic desert, and therefore has an advantage as a bioindicator. Radiocesium in lichens from the Kola catchments showed generally low levels.

### INTRODUCTION

The Central Kola Expedition, in co-operation with the Geological Surveys of Finland and Norway, are carrying out a joint ecogeochemical mapping project in the western part of the Murmansk region (Russia) and contiguous areas of Finland and Norway, covering a total area of 188,000 km<sup>2</sup> (see <http://www.ngu.no/Kola>). As one part of this project, a detailed study of eight selected catchments (each 11-35 km<sup>2</sup>) was carried out during 1994. These catchments were chosen to be spread over the whole project area at varying distances from the main sources of air pollution in the Barents region (see Fig. 6.2.1). A wide range of sampling media was collected and analysed from several localities within each catchment. More than 38 inorganic substances were determined using advanced analytical techniques. One of the major aims of this study was to identify the variations in element distribution in different media affected by varying levels of contamination compared to the influence of geological factors within the study area.

During the last decades, the term bioindicators or biomonitors has gained increasing popularity. Bioindication is generally defined as the use of an organism to obtain information on the quality of its environment (Wittig, 1993). Several studies have been carried out in the vicinity of industrial centres within the Barents region by using the chemical content of various plants as an indication of the pollution status. In the terrestrial ecosystems in these Arctic regions, mosses have been used with success in regional environmental studies more frequently than any other vegetation type (see e. g. Rühling and Tyler, 1973; Steinnes, 1995), but lichens are also used to assess air quality with respect to heavy metals (e. g. Takala et al., 1994; Nash and Gries, 1995) or radiocesium fallout (e. g. Rissanen and Rahola, 1990).

Lower plants like mosses or lichens have several advantages as bioindicators when compared to higher plants, due to their lack of roots, lack of variability of morphology through the growing season and their lack of cuticula (in the case of lichens and most moss species). In addition, they have a high surface to volume ratio and exhibit ion exchange properties. Berg et



al. (1995) found generally significant correlations between mosses and wet deposition of elements transported over long distances, and have estimated the uptake efficiencies of Pb by use of the moss species *Hylocomium splendens* to vary from 65 % to nonexistent at background stations in Norway. The relative efficiencies of different moss and lichen taxa in retaining airborne trace elements have also been evaluated by Steinnes (1993), by comparing the chemistry of mosses (e. g. *Hylocomium* sp.) and lichens (*Cladonia* sp.) sampled simultaneously in Norway. He found an apparently lower uptake of the analysed heavy metals (Cd, Cu, Pb and Zn) in *Cladonia* sp. relative to *Hylocomium*.

Several other studies of the chemical composition of higher plants have been carried out in both heavily polluted and more pristine parts of the Euro-Arctic territories. Nickel and copper content in berries from the Monchegorsk area have been found to exceed health standards (Barkan et al., 1993). Aamlid and Skogheim (1993) also found regional patterns of heavy metals in berries from northeast Norway close to the Nickel smelter. Needles from coniferous forests have been investigated intensively by several Russian scientists (Lukina and Nikonov, 1992; 1993) but a drawback of conifers is that they disappear in the most heavily polluted sites. In contrast, leaf trees such as birch survive even within the technogenic deserts. Although birch leaves accumulate less contaminants than pine needles, they have been found to be useful as bioindicators, and an abrupt decrease in content of the main contaminants (Ni and Cu) has been reported by Kozlov et al. (1995) around the Severonikel enterprise.

Mosses are widely used as bioindicators, and several studies have found moss to be an efficient accumulator of contaminants, but this plant taxa is not important as food for vertebrates. Only lemmings and a few other animals living in cold climates eat mosses (Slack, 1988). In contrast, both reindeer lichens (*Cladonia* sp.) and berries (e.g. crowberries) have a direct link to the food chain as they are consumed by reindeer or man. This makes them especially interesting in environmental studies. The ecology of lichens and mosses make them sensitive but also vulnerable to heavy depositions of pollution. Crowberries have therefore been chosen as a supplement in this study.

In this paper we focus on 1) the chemical composition of various ground vegetation sampled simultaneously, 2) the response of this vegetation to the local depositions and 3) an evaluation of their properties as bioindicators of airborne pollution.

## MATERIAL AND METHODS

### *Study area*

The eight catchments (C1 - C8) chosen for this study are situated at various distances from the main pollution sources in the European North (see Fig. 6.2.1 and Tab. 6.2.1). Four of them are in Russia: C1, Zapoljarnyi (10 km NE of the concentration plant in Zapoljarnyi); C2, Monchegorsk (5 km S of "Severonikel" smelter complex in Monchegorsk); C3, Kirovsk (surrounded by major mining activity); and C4, Kurka (25 km S of Monchegorsk). One is in Norway, C5, Skjellbekken (31 km SW of the Nickel smelter and S of Kirkenes); and three are in Finland: C6, Kirakka (near the Norwegian border, 57 km NW of Nickel); C7, Naruska (in E Finland 65 km NE of the town of Kovdor and about 100 km NE of the town of Kemijarvi); and C8, Pallas (in W Finland). The annual precipitation in 1994, as measured at the nearest meteorological stations, varied from 386 mm (near C6) to 513 mm (near C7). A detailed description of catchments is given by Chekushin et al. (1995) and Reimann et al. (1995).

## SAMPLING

The samples were collected according to the field manual by Äyräs & Reimann (1995). Where possible, seven localities were chosen within each catchment for sampling vegetation. Samples were collected twice during the summer of 1994. The sampling took place in July and at the end of September or beginning of October in Russia (C1, C3 and C4), at the end of May and September in Norway (C5), and in June and August in Finland (C6 - C8).

The sampling localities were spread over each catchment. Every effort was made to avoid contaminating the samples with soil or litter, and places underneath trees or bushes were avoided wherever possible. Only the three outermost shoots of the mosses *Hylocomium splendens* and *Pleurozium schreberi* were sampled as the main taxa of vegetation from each catchment. In addition, the soil-free, upper living portions (ca. 1-2 cm) of ground lichens (*Cladonia* sp. and *Cetraria* sp.) and the upper living portions (5 - 6 cm) of crowberry (*Empetrum nigrum*) were collected from the same localities in the Russian catchments (C1 - C4). However, due to the heavy depositions of pollutants in this zone, no localities with lichens or mosses could be found in C2 near Monchegorsk, and only two moss localities could be found in both C1 and C4 (Table 6.2.1).

## CHEMICAL ANALYSIS

The unwashed samples were pre-dried in the field and then fully air dried at  $< 40^{\circ}\text{C}$  and milled upon arrival at GTK's laboratory in Rovaniemi. Then samples were dissolved with 10 mL of concentrated nitric acid in a microwave oven, and diluted with water to 50 mL (US EPA standard 3051) before being analysed for a variety of elements by ICP-AES (crowberry was analysed with this method for all elements), ICP-MS and cold vapor AAS at GTK's accredited laboratory (Niska-vaara, 1995). For three of the Russian catchments (C1, C3 and C4),  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations were determined in lichen using a High Purity Germanium semiconductor and a multichannel pulse-high analyser at the Finnish Centre for Radiation and Nuclear Safety (STUK) in Rovaniemi.

## DATA ANALYSIS

The influence of three different factors on the total variability of the element concentrations in the vegetation were estimated by use of one-way ANOVA algorithm calculation (Krumbein and Graybill, 1965.) available in the STATGRAFICS program. ANOVA analysis was used to estimate the influence of the three main factors on the variability of the element content in the moss samples, namely between catchments (R1), between sampling points within each catchment (R2) and finally seasonal variation (R3).

The total variation of element concentrations is estimated as the sum of element concentration squares deviated from the general mean. The variation, associated with different values of the factors studied, is calculated as the difference between the total variation and the sum of squares of deviations from partial (local) means. The contribution of the factor is calculated as the ratio (in %) of variation connected with the factor to the total variation. The residue is the result of factor interactions and unexplained contributions (detection limits etc.), and is included to make the total sum to 100 %.

In several of the approaches in this paper, we have used arithmetic mean values from each catchment. For analytical results below the detection limit, half the detection limits is used for calculations and indicated by italics in the statistical summary as shown in Table 6.2.2.

## RESULTS AND DISCUSSION

The results of the chemical analysis of 38 elements in the three vegetation taxa are presented as the arithmetic mean for each catchment in Table 6.2.2. In general, moss, lichen and crowberry show similar variation between catchments. The catchments represent different level of contamination due to the varying distances to the pollution centres. The different concentration levels observed in the mosses are described comprehensively by Åyräs et al. (1996). The highest levels of Cr, Fe, Mg, S and Sc are in C1 near Zapoljarnyi for all three taxa. Ag, Ba, Bi, Cd, Na, Sb and Se have the highest concentration in mosses and lichens within the impact zone of the “Severonickel” complex (C4). In the almost vegetation-free area of C2 near the “Severonickel” complex, there are no results for moss and lichen, but contents of As, Co, Cu, Mo, Ni and Pb in crowberry are higher than for all the other vegetation samples in our study. The highest levels of Al, Be, Ca, K, La, Sr, Th, Ti, Tl, V and Y are seen for mosses, and Ca and K in crowberries from C3 near the big open - pit Apatite mines in Kirovsk. Most of these elements are typical of the alkaline bedrock of the Hibiny mountain. The lowest concentrations of elements, except for Rb, are found in C7 and C8. These catchments are situated far away from major industrial centres, and therefore represent a background area.

The results of the ANOVA variability analysis are shown in Figure 6.2.2, which indicates that the main component of variability is the difference between the catchments (R1) in all three taxa for Co, Cr, Fe, Mg, Ni, P, Pb and Sr. The same factor also has a major influence in mosses and lichen for Ag, La and Y, in mosses and crowberry for Cu and V, in mosses only for Al, As, Ba, Cd, S and Ti, in lichen and crowberry for Mo and Zn, and in lichen only for K. This factor has little influence on levels of Ca, Mn and Na. The effect is greatest for most elements in moss. In general, R1 plays a minor role for the chemical variability for lichen and crowberry, because these samples were taken from only the most polluted catchments, but for elements like Ag, Co, Cr, Cu, La, Ni, Sr, V and Y, the between-catchment variations for lichen and crowberry are as large as the variation observed for the moss data. For K, Mo and Zn, this factor (R1) gives a broader variation for lichen and crowberry than for moss. Contributions of seasonal variations (R3) and variations within the catchments (R2) are generally very low for all three taxa of vegetation. However, notably large variation within each catchment (R2) is seen for Ca, S and Ti in lichen, and S in crowberry. The seasonal variations are highest for K in crowberry, for P and Zn in moss and Pb in lichens. The differences in cause of variation for each taxa can partly be explained by the fact that mosses were sampled from areas ranging from extremely polluted to unpolluted ecosystems, while lichen and crowberry are not sampled from such a wide range of environments (see Tab. 6.2.1).

### *Radiocesium in ground lichen.*

The radiocesium levels of the lichens samples have previously been described in detail (Halleraker et al., 1995), and the mean contents are shown in Table 6.2.2. The levels of  $^{134}\text{Cs}$  (half-life of only 2 years) and to a lesser extent  $^{137}\text{Cs}$  are low compared to previous investigations of radiocesium in reindeer lichens in both Norway and Finnish Lapland, where the impact from the Chernobyl accident in 1986 was more severe (National Institute of Radiation Hygiene, 1986; Rissanen and Rahola, 1990). No significant differences in radiocesium levels were found between the catchments, but an especially high  $^{137}\text{Cs}$  concentration (550 Bq/kg) was observed in one lichen sample (*Cladonia sulphurina*, a species not consumed by reindeer) from C4.

The nitric acid extractable content of Th and U was analysed for all three taxa of vegetation by ICP-MS and ICP-AES. In general, the concentrations are very near the detection limits, but

enriched content of these toxic heavy metals was observed in many of the catchments, with the maximum in moss from C3 (Th) and C4 (U).

#### *Bioindication properties of studied vegetation*

For a terricolouse bioindicator of atmospheric fallout to be useful, it must show a strong correlation between its content and the actual deposition of the same element. Several previous investigations, including both laboratory experiments (Gjengedal and Steinnes, 1990) and field studies (Berg et al., 1993), have provided knowledge about the connection between wet depositions and element content in mosses.

The X-Y diagrams in Figure 6.2.4a and 6.2.4b illustrate the relationship (correlation or lack of correlation) between the mean element contents in plants and the calculated annual deposition of 17 selected elements from each of the catchments. Please note that logarithmic scales are used on both axes, except for the Ca, Pb, S and Zn diagrams. Mean annual deposition is used as an estimate of airborne deposition intensity for each catchment. The annual depositions in  $\text{kg}/\text{km}^2$  are calculated based on both snow (to reflect deposition during the previous winter) and rainwater (monitoring from May to October), sampled from several localities within each catchment during 1994. These calculations include both dry (filter residue of snow) and wet (meltwater of snow and filtered rainwater) depositions. A more comprehensive presentation of the methods and the results of the calculations are given by Chekushin et al. (1996).

Although element variations show similar tendencies for moss, lichen and crowberry, the concentration levels differ significantly between the taxa. This fact is related to differences in their physiology and ecology, such as their structures, growth rate and ion exchange properties. It is possible to pick out groups of elements with similar correlation of contents between moss, lichen and crowberry. Levels of Al, Co, Cr, Cu, Fe, La, Ni, Pb, Sc, Ti, V, Y and Zn are higher in moss than in lichen, and higher in lichen than in crowberry. This tendency allows us to qualitatively compare the sensitivity of these vegetation taxa to the pollutants, showing that the mosses have the best accumulation potential for the majority of the elements including all the main pollutants except S. The order is moss > crowberry > lichens for P and Sr, while contents of Ba, Ca, K, Mg, Mn, Mo and S are more or less similar in moss and crowberry, but lower in the lichen samples. It is interesting to notice that most nutrients have a different relative concentration in lichens compared to the other taxa. Physiological differences obviously play a major role in determining the nutrient content, while the ion exchange properties are more important for the first group of elements. For the main pollutants in this area, moss has the best accumulation potential of the studied taxa. The same conclusion was reached by Steinnes (1993) when comparing Cd, Cu, Pb and Zn content in *Hylocomium splendens* and *Cladonia* sp. from the Norwegian survey in 1976.

The following features, which characterise the properties of bioindicators, are illustrated in Figure 6.2.3, and are estimated on the basis of the x-y diagrams in Figure 6.2.4 a and b for each element.

**Background** is defined as the minimum content of an element, the level typical for plants growing far away from local pollution sources. However, in general, background level is hard to define due to the fact that some area naturally have anomalously high element content.

**Deposition sensitivity** is the minimum value of annual deposition at which element concentration in plants starts to exceed the background level (or the analytical detection limit, if that is higher).

**Contrast ratio** is the degree of change in element contents resulting from varying deposition intensity. This term corresponds to the angle between the trend line (estimated) and the horizontal axis. The bigger this angle is, the higher is the contrast ratio.

**Saturation limit** is the maximum uptake of an element by a bioindicator, where an additional deposition of an element does not lead to an increased concentration in the bioindicator. This phenomena has previously been described in the literature as the barrier effect (Kovalevskii, 1979).

The features defined above describe the relationship between depositions of an element and the content of the same element in the bioindicator. The trend lines in Figure 6.2.4a and 6.2.4b are only estimates (not calculated) outlining the main tendencies in varying element contents versus depositions.

**Aluminium (Al):** The contents of Al are stable in the background catchments, where the annual depositions are less than 10 kg/km<sup>2</sup>. The level in the mosses is 0.12 - 0.14 g/kg, which seems to represent background. The sensitivity and contrast ratios are slightly higher for moss than for lichen and crowberry.

**Arsenic (As):** The saturation limit is clearly defined in Figure 6.2.4a for moss and lichen. The level is essentially the same for both taxa and is reached when depositions exceed 1.0 kg/km<sup>2</sup>. Berg et al. (1993) calculated the uptake efficiencies of As in moss to be very low (10 - 25 %) in background stations in Norway. Background concentrations in moss are about 2.5 mg/kg. The sensitivity of moss is slightly higher than that of lichen, while the contents of As in crowberry are below the analytical detection limit of 5 mg/kg (ICP-AES).

**Cobalt (Co):** The deposition sensitivity of Co for moss is almost 10 times higher than for lichen and crowberry, while the contrast ratio is essentially the same for all three taxa. The sensitivity limit for moss is distinguished clearly when the level of deposition is less than 0.03 kg/km<sup>2</sup>, which gives a background content of 0.15 mg/kg. The saturation limits for moss and lichen are reached when the level of depositions exceed 4.5 kg/km<sup>2</sup>. No saturation limit was reached for Co in crowberry within the range of depositions in our study. The contents of Co in the bioindicators increase uniformly with increasing depositions until the saturation limit of moss and lichens are reached.

**Chromium (Cr):** The deposition sensitivity of Cr is higher for moss than for lichen and crowberry, while the contrast ratio for all three taxa is the same in areas of low to middle depositions. A tendency to reach saturation limit can be seen for moss in areas affected by the highest depositions.

**Copper (Cu):** The reaction of the bioindicators to this element is similar to their reaction to cobalt. Moss has the best deposition sensitivity. Saturation limits are seen for both moss and lichen when depositions exceed 50 kg/km<sup>2</sup>, but once again not for crowberry. The contrast ratio is slightly higher for lichen than for moss and crowberry.

**Iron (Fe):** Deposition sensitivity of moss is 10 times higher (as is true for Co) than for lichen and crowberry, which are almost the same. The limit of sensitivity for moss occurs at depositions near 10 kg/km<sup>2</sup>, while the background content is about 0.1 g/kg. Crowberry has the highest contrast ratio. A decreasing contrast ratio is observed in moss affected by high-level depositions. The reason is probably the proximity to the saturation limit. Iron is the only macronutrient which shows the highest contents in moss relative to the other taxa in addition to a good correlation to the annual deposition.

**Manganese (Mn):** A negative correlation was found between deposition and the Mn-content in all the plant taxa. Such a behaviour has previously been explained by the negative correlation between Fe and Mn levels in plants growing on acidic soils (Kabata-Pendias and Pendias, 1989). This tendency was also observed for different taxa of vegetation (Lukina and Nikonov, 1992; 1993) in the zone influenced by the “Severonikel” plant. The increasing iron contents in plants from contaminated areas leads to a decrease of Mn. This situation is illustrated in more detail in Figure 6.2.5. This diagram shows the decreasing Fe/Mn-ratio with increasing distance from the main pollution sources, except for C3 where local dust sources of both elements probably disturb this picture.

**Magnesium (Mg):** The correlation between Mg-depositions and uptake in moss or crowberry is practically nonexistent, and only the highest depositions in C1 (more than 70 times higher than C8) are reflected to a small degree by the bioindicators. The contrast ratio of lichen is slightly higher than for moss and crowberry. The nutrient factor seems to be more important than the deposition, leading to a constant level of ca. 1 g/kg in both moss and crowberry regardless of the depositions.

**Molybdenum (Mo):** A correlation does, on the other hand, exist between depositions and Mo contents in lichen. Mo contents in moss represent background conditions except for C4 (which experiences depositions 13 times higher than C5 or C6). The relatively high contents of Mo for C3 and C6 are not connected with atmospheric fallout. They can be considered to be the local background levels for these areas. These values show good correlation with high levels of Mo in stream water and organic stream sediments from C3 and C6, and probably reflect local dust contributions (Chekushin et al., 1995). This is a good example to illustrate how wrong it would be to consider only atmospheric depositions in determining general background levels. Mo contents in crowberry are below the analytical detection limit and therefore excluded from the diagram.

**Nickel (Ni):** The bioindicators react to this element in much the same manner as to the other main pollutants, Co and Cu. A clear tendency to reach saturation limits is seen for both moss and lichen when the depositions exceed 70 kg/km<sup>2</sup>. However, no saturation limit was reached for crowberry, despite the fact that this species survives in some of the most extreme ecosystems in Northern Europe for Ni depositions. The limit of sensitivity for the moss is at the level of depositions near 0.5 kg/km<sup>2</sup>, giving a background content of 2.0 mg/kg.

**Lead (Pb):** Deposition sensitivity for moss is considerably higher than for lichen and crowberry, while the contrast ratio for lichen is higher than for moss. The contrast ratio of crowberry is high at the heaviest deposition (C2), but decreases rapidly to less than the analytical detection limit of 3 mg/kg (ICP-AES) when the depositions decrease.

**Sulphur (S):** The S concentrations in moss from catchments with background depositions are at approximately the same level, and ca. 800 ppm is probably the background level in moss. The contrast ratio for moss is only distinct when comparing catchments affected by the middle to highest depositions (C1). However, surprisingly, the total range between the highest and the lowest annual depositions varies by only a factor of 12 in our selected areas (Chekushin et al., 1996), despite the enormous known emissions of SO<sup>2</sup> from both the Pechenganikel and the Severonikel enterprises. Crowberry also reflects the varying depositions for Russian catchments, but its contrast ratio is far lower than for moss. The S-concentrations in lichen are only slightly enriched in C1, and lichen is therefore not very suitable for monitoring S depositions.

**Strontium (Sr):** The background contents (7 - 9 mg/kg) are in the same range for both moss and crowberry when depositions are less than 0.5 kg/km<sup>2</sup> and 5.0 kg/km<sup>2</sup> respectively (a factor of 10!). This depositions can also be estimated as the limit of sensitivity. The contrast ratios of moss and lichen are practically equal, but the accumulation potential is approximately 10 times higher for moss than for lichens.

**Vanadium (V):** Correlations have been found between V-depositions and the V-uptake in crowberry and moss, and to a small extent in lichen. However, the relationship between depositions and V-content in the moss gives an unclear picture, but the limit of sensitivity for the moss is at the level of depositions near 1.0 kg/km<sup>2</sup>, giving a background content of less than 0.6 mg/kg. Berg et al. (1995) indicated that V is one of several elements that shows general background levels.

A general lack of correlation between Ca, Cd and Zn contents in the bioindicators and the airborne deposition is illustrated in Figure 6.2.4a and 6.2.4b. All three elements have a narrow variation of depositions (ratio max. load/min. load < 14), which may partially explain the lack of correlation, since the limit of sensitivity to the depositions is probably much higher. Tendencies toward general background concentrations are found for at least Ca, Cd, Co, Cr, Mg, Mn, Mo, Ni, S, Sr, V and Zn in moss, but the number of samples from real background areas is small (only moss from C7 and C8). These findings correspond somewhat with the lack of correlation between uptake in moss and wet depositions in background stations of Norway which was found for several elements (Berg et al., 1993). Selective element uptake does occur, and the fact that bioindicators also need nutrients is probably the reason for this lack of correlation, in addition to very low depositions levels of Zn.

## CONCLUSIONS

All three of the studied vegetation taxa react to atmospheric fallout of most pollutants, but the degree of reaction differs considerably. Moss has better bioindicator properties relative to lichen and crowberry for most of the pollutants (Al, Co, Cr, Cu, Fe, Ni, Pb, S and Sr). The moss species *Hylocomium splendens* and *Pleurozium schreberi* show high sensitivity to depositions as well as a good contrast ratio and accumulation potential within most parts of the deposition range. However, the levels of elements like Ca, Cd, Mg, Mo and Zn in the moss samples do not correlate with the calculated annual deposition data from the respective catchments. The known negative relation between contents of Fe and Mn in plants in acidic soils is confirmed for the taxa of vegetation studied, shown as a negative correlation between Mn in the plants and the known depositions.

The levels of Co, Cr, Cu, Fe and Ni in moss seem to reach saturation limit in areas with the most intensive technogenic depositions. Therefore, the bioindicator properties of moss for these elements degrade in highly polluted areas, and both moss and lichen disappear from the most heavily polluted sites.

Crowberry has good bioindicator properties in the range from middle to high technogenic depositions for Co, Cr, Cu, Fe, Ni and S. It generally needs higher atmospheric input than moss before it reacts, but has a good contrast ratio. The advantages of crowberry compared to moss or lichens are a very high saturation limit (not really distinguishable in our material for any of the 17 studied elements), and the ability to survive even in the most polluted territories. The use of crowberry makes it possible to estimate even the highest technogenic depositions, which were found in areas where both moss and lichen have died out.

Mosses have a strong position internationally as a bioindicator of heavy metals. This article strengthens this plant taxa's position in this context. In addition, crowberry and in some cases, reindeer lichen could be useful bioindicators when studying the most polluted areas. These plant taxa give interesting additional information due to their direct link to the food chain.

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## TABLES

Table 6.2.1. Some key figures for the eight studied catchments, with numbers of sampled plots with various vegetation species within each catchment.

Table 6.2.2. Element contents (mean) in three taxa of ground vegetation. **The figures indicated by bold font show maximum element contents for each taxa**, while shaded figures show the absolute maximum contents. *Italic font indicates calculations including half the detection limit values.*

## FIGURES

Figure 6.2.1. Location of the studied catchments in the centre of the Barents region, Northern Europe. The outer frame illustrates the borders of the ecogeochemical mapping area. The eight studied catchments are shown as no. 1-8: **1**: Zapoljarnyi, **2**: Monchegorsk, **3**: Kirovsk, **4**: Kurka, **5**: Skjellbekken, **6**: Kirakka, **7**: Naruska, **8**: Pallas. **K**: Kirkenes, **N**: Nikel, **Z**: Zaplojarniy, **M**: Monchegorsk, **Mu**: Murmansk.

Figure 6.2.2. Estimation of the influence of different factors on element level variability in ground vegetation taxa, calculated in % by one-way ANOVA algorithm.

Figure 6.2.3. The main features for bioindicator properties, as illustrated by Ni content in moss from this study.

Figure 6.2.4a. Relationship between element concentrations in bioindicators and annual depositions (atmospheric fallouts) from eight catchments in the Barents region.

Figure 6.2.4b. Relationship between element concentrations in bioindicators and annual depositions from eight catchments in the Barents region.

Figure 6.2.5. Behaviour of the Fe/Mn ratio in plants at varying distances from the sources of pollution.

Table 6.2.1. Some key figures of the eight studied catchments, and the number of sampled plots with various taxa within each catchment.

No	Name	Co-ordinates - outlet	Size (km <sup>2</sup> )	Altitudes (m.a.s.l.)	Moss		Lichen		Crow- berry <i>En</i>	Distance and position to nearest major industry (emission sources)
					<i>Hs</i>	<i>Ps</i>	<i>Cl</i>	<i>Ce</i>		
C1 (Ru)	Zapoljarnyi	69°27' N 31°03' E	19.0	25 - 373	0*	4	10	4	14	10 km NE of Zapoljarnij (roasting) 30 km W of Nikel (Ni - Cu smelter)
C2 (Ru)	Monchegor sk	67°50' N 32°54' E	22.4	128 - 507	0*	0*	0*	0*	14	5 km S of Severonikel (Ni - Cu smelter)
C3 (Ru)	Kirovsk	67°32' N 33°48' E	20.0	240 - 1075	14	14	14	-	14	2 km S of Kirovsk (ap-ne mines) 5 - 15 km N and W of several ap-ne
C4 (Ru)	Kurka	67°41' N 32°50' E	20.5	152 - 466	2	2	5	7	14	25 km SW of Severonikel 20 km W of Apatity (ap-ne)
C5 (N)	Skjellbekke n	69°21' N 29°27' E	34.6	80 - 297	12	3	-	-	-	30 km W of Nikel 50 km SW of Kirkenes (Fe- smelter)
C6 (Fin)	Kirakka	69°35' N 28°52' E	11.9	110 - 200	14	-	-	-	-	60 km NW of Nikel 50 km W of Kirkenes
C7 (Fin)	Naruska	67°21' N 29°22' E	20.2	263 - 490	14	-	-	-	-	65 km SW of Kovdor (Fe - ap- ne) 150 km W of Kandalaksha (Al- plant)
C8 (Fin)	Pallas	68°09' N 23°52' E	24.4	303 - 500	14	-	-	-	-	150 km W of Kiruna (Fe- mines) 290 km SW of Nikel

Abbreviations: *Hs*; *Hylocomium splendens*, *Ps*; *Pleurozium schreberi*, *Cl*: *Cladonia spp.*, *Ce*: *Cetraria spp.*, *En*: *Empetrum nigrum*, Ru: *Russia*, N: *Norway*, Fin: *Finland*, ap-ne: *Apatite - nephelin industry*

\* Indicates that no samples of this taxa could be found within the catchment, while (-) indicates that no effort was made to find this taxa.

Catchment		1	1	1	2	3	3	3	4	4	4	5	6	7	8
Media		Moss	Lichen	Crowberry	Crowberry	Moss	Lichen	Crowberry	Moss	Lichen	Crowberry	Moss	Moss	Moss	Moss
#Samples		4	14	14	14	14	14	14	14	12	14	14	14	14	12
Ag	mg/kg	0.085	0.104			0.102	0.051		<b>0.57</b>	<b>0.772</b>		0.045	<i>0.021</i>	<i>0.029</i>	<i>0.009</i>
Al	mg/kg	303	211	122	<b>174</b>	<b>1396</b>	<b>277</b>	85	693	151	62.9	141	117	133	128
As	mg/kg	0.818	<b>1.75</b>	2.5	<b>2.86</b>	0.539	<i>0.456</i>	2.5	<b>1.68</b>	1.49	2.5	0.429	0.256	0.172	0.129
B	mg/kg	2.86	<b>0.661</b>	<i>21.5</i>	17.7	2.39	<i>0.412</i>	19.1	<b>3.97</b>	<i>0.412</i>	18.1	1.91	2.64	1.16	1.32
Ba	mg/kg	18.2	2.36	24.3	20.2	34.4	4.38	<b>31.4</b>	<b>180.75</b>	<b>5.87</b>	24.7	21.2	19.4	18.1	16.4
Be	mg/kg	0.02	0.006			<b>0.094</b>	<b>0.017</b>		0.039	0.012		0.01	0.055	0.027	0.028
Bi	mg/kg	0.033	<i>0.044</i>			0.044	<i>0.02</i>		<b>0.138</b>	<b>0.183</b>		0.026	0.012	0.019	0.021
Ca	mg/kg	3273	<b>373</b>	4894	3942	2714	234	<b>5349</b>	<b>5165</b>	353	4999	2915	2840	2486	2499
Cd	mg/kg	0.143	0.084	<i>0.25</i>	<b>0.275</b>	0.127	0.08	<i>0.25</i>	<b>0.295</b>	<b>0.122</b>	<i>0.25</i>	0.109	0.059	0.077	0.084
Co	mg/kg	<b>8.67</b>	6.87	5.34	<b>26.2</b>	0.857	0.304	<i>0.25</i>	11	<b>8.36</b>	3.31	0.955	0.391	0.152	0.152
Cr	mg/kg	<b>5.75</b>	<b>6.49</b>	<b>3.19</b>	2.16	1.55	0.639	<i>0.25</i>	2.18	1.32	<i>0.461</i>	0.878	0.499	0.377	0.424
Cs <sup>134</sup>	Bq/kg		<b>2.74</b>												
Cs <sup>137</sup>	Bq/kg		107				102								
Cu	mg/kg	124	138	85.7	<b>396</b>	24.1	6.33	6.74	<b>170</b>	<b>199</b>	67	20.8	9.32	5.54	4.18
Fe	mg/kg	<b>1850</b>	<b>1929</b>	<b>1096</b>	463	581	165	46.4	625	265	80	261	145	96.4	92.1
Hg	mg/kg	0.118	<b>0.179</b>			0.105	0.114		0.108	0.142		<b>0.122</b>	0.121	0.066	0.074
K	mg/kg	<b>6003</b>	1793	5221	4258	4734	2106	<b>6494</b>	5245	<b>2832</b>	5625	3991	3241	3537	3794
La	mg/kg	<i>0.35</i>	<i>0.382</i>	<i>0.35</i>	<i>0.35</i>	<b>5.74</b>	<b>2.56</b>	<b>0.375</b>	4.7	<i>0.617</i>	<i>0.35</i>	<i>0.35</i>	<i>0.554</i>	<i>0.35</i>	<i>0.35</i>
Li	mg/kg	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>	<i>0.35</i>
Mg	mg/kg	<b>1993</b>	<b>796</b>	<b>1551</b>	983	680	161	1294	998	209	991	925	951	724	849
Mn	mg/kg	262	33.8	270	143	269	23.2	464	225	<b>65.4</b>	<b>522</b>	<b>638</b>	435	481	384
Mo	mg/kg	0.088	0.086	<i>0.5</i>	<b>1.11</b>	0.295	0.06	<i>0.5</i>	<b>0.55</b>	<b>0.348</b>	<i>0.5</i>	0.061	0.249	0.069	0.074
Na	mg/kg	80	<i>80.4</i>	<b>50</b>	37.9	64.3	46.4	35.4	<b>83.8</b>	<b>131</b>	25.8	26	21.4	18.6	21.1
Ni	mg/kg	<b>250</b>	<b>189</b>	174	<b>348</b>	16	5.31	7.72	177	175	73.4	27.6	8.75	2.26	1.56
P	mg/kg	<b>1790</b>	628	800	587	1656	915	<b>1480</b>	1417	<b>1023</b>	903	1118	834	992	1137
Pb	mg/kg	2.46	1.54	<i>1.5</i>	<b>6.57</b>	6.43	2.64	<i>1.5</i>	<b>7.23</b>	<b>5.62</b>	<i>1.71</i>	2.76	2.29	3.54	3.21
Rb	mg/kg	10.2	3.49			15.2	7.96		6.01	<b>8.77</b>		10.8	9.23	12.4	<b>16.6</b>
S	mg/kg	<b>1475</b>	<b>891</b>	<b>1534</b>	1443	964	602	1337	1305	702	1323	891	772	722	746
Sb	mg/kg	0.123	0.015	3.5	3.5	0.103	0.027	3.5	<b>0.18</b>	<b>0.099</b>	3.5	0.041	<i>0.027</i>	0.039	<i>0.032</i>
Sc	mg/kg	<b>0.088</b>	<b>0.086</b>	<i>0.054</i>	<i>0.05</i>	<i>0.064</i>	<i>0.05</i>	<i>0.054</i>	<i>0.075</i>	<i>0.054</i>	<i>0.05</i>	<i>0.053</i>	<i>0.054</i>	<i>0.054</i>	<i>0.054</i>
Se	mg/kg	0.418	0.4			0.289	0.248		<b>0.588</b>	<b>0.76</b>		0.177	0.229	0.14	0.071
Si	mg/kg	140	<b>61.4</b>	<b>101</b>	82.9	<b>213</b>	53.2	56.4	142.5	45	58.6	89.3	79.3	77.9	67.9
Sr	mg/kg	13.8	1.58	9.34	8.6	<b>112</b>	<b>11.1</b>	<b>88.1</b>	22.6	2.23	7.24	6.02	9.08	10.4	6.77
Th	mg/kg	0.035	<i>0.032</i>	2.5	2.5	<b>0.089</b>	<b>0.051</b>	2.5	0.05	<i>0.023</i>	2.5	0.021	0.021	0.021	0.014
Ti	mg/kg	20.4	<b>31.5</b>	11.8	<b>16.3</b>	<b>54.1</b>	24.8	2.764	19.6	12.2	1.84	5.28	3.14	3.41	2.56
Tl	mg/kg	0.015	0.009			<b>0.038</b>	0.007		0.028	<b>0.013</b>		<i>0.023</i>	0.061	0.031	<i>0.04</i>
U	mg/kg	0.01	0.015			0.087	<b>0.024</b>		<b>0.128</b>	0.014		<i>0.005</i>	<i>0.121</i>	<i>0.01</i>	<i>0.008</i>
V	mg/kg	3.06	<b>2.21</b>	1.42	<b>4.34</b>	<b>12.9</b>	1.65	0.346	5.09	1.93	0.796	1.26	1.05	1.28	0.99
Y	mg/kg	0.063	<i>0.075</i>	<i>0.05</i>	<b>0.071</b>	<b>0.993</b>	<b>0.464</b>	<i>0.054</i>	0.9	<i>0.138</i>	<i>0.05</i>	<i>0.05</i>	<i>0.082</i>	<i>0.05</i>	<i>0.057</i>
Zn	mg/kg	34.9	27.3	10.1	10.3	33.7	16.9	<b>16.8</b>	30.7	<b>44.1</b>	12.5	<b>48</b>	29.5	27.6	33.9

Table 6.2.2. Element contents (mean) in three taxa of ground vegetation. The figures indicated by bold font show maximum element contents for each taxa, while shaded figures show the absolute maximum contents. *Italic font indicates calculations including half the detection limit values.*

*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
Catchment locations

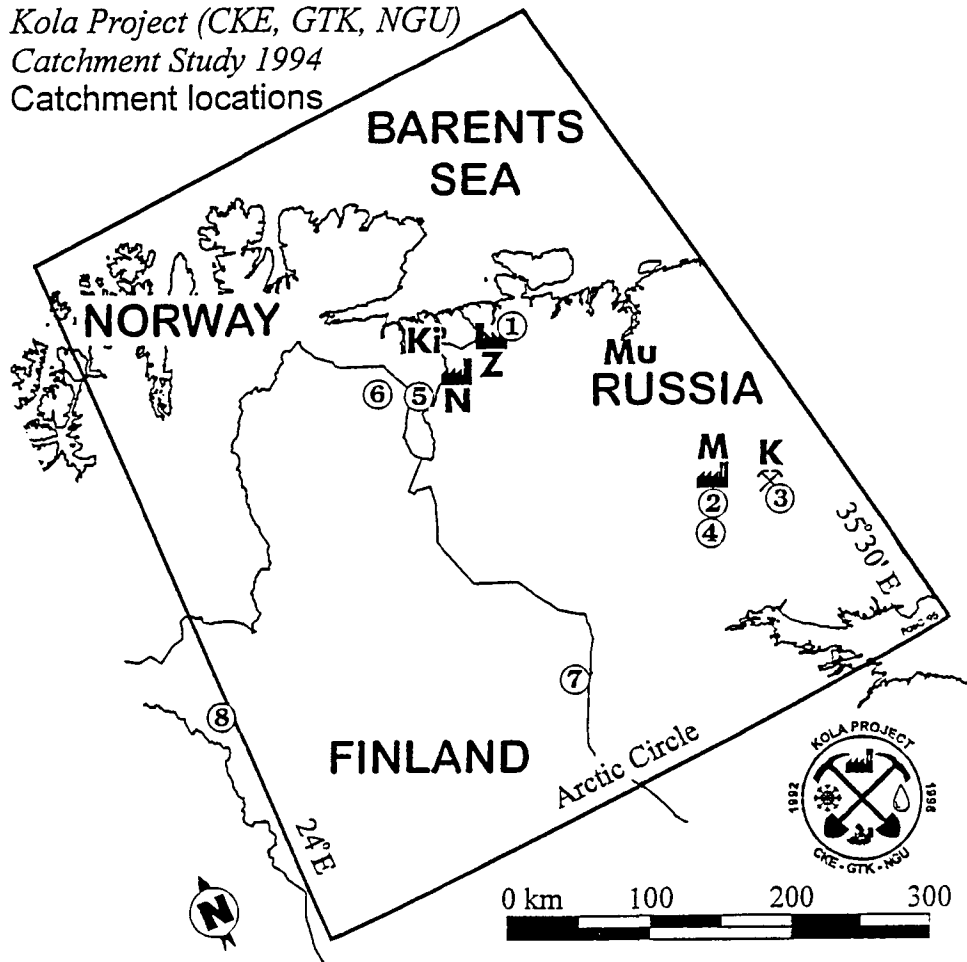


Figure 6.2.1. Location of the studied catchments in the centre of the Barents region, Northern Europe. The outer frame illustrates the borders of the ecogeochemical mapping area. The eight studied catchments are shown as no. 1-8: 1: Zapoljarnyi, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, 8: Pallas. K: Kirkenes, N: Nikel, Z: Zaplojarniy, M: Monchegorsk, Mu: Murmansk.



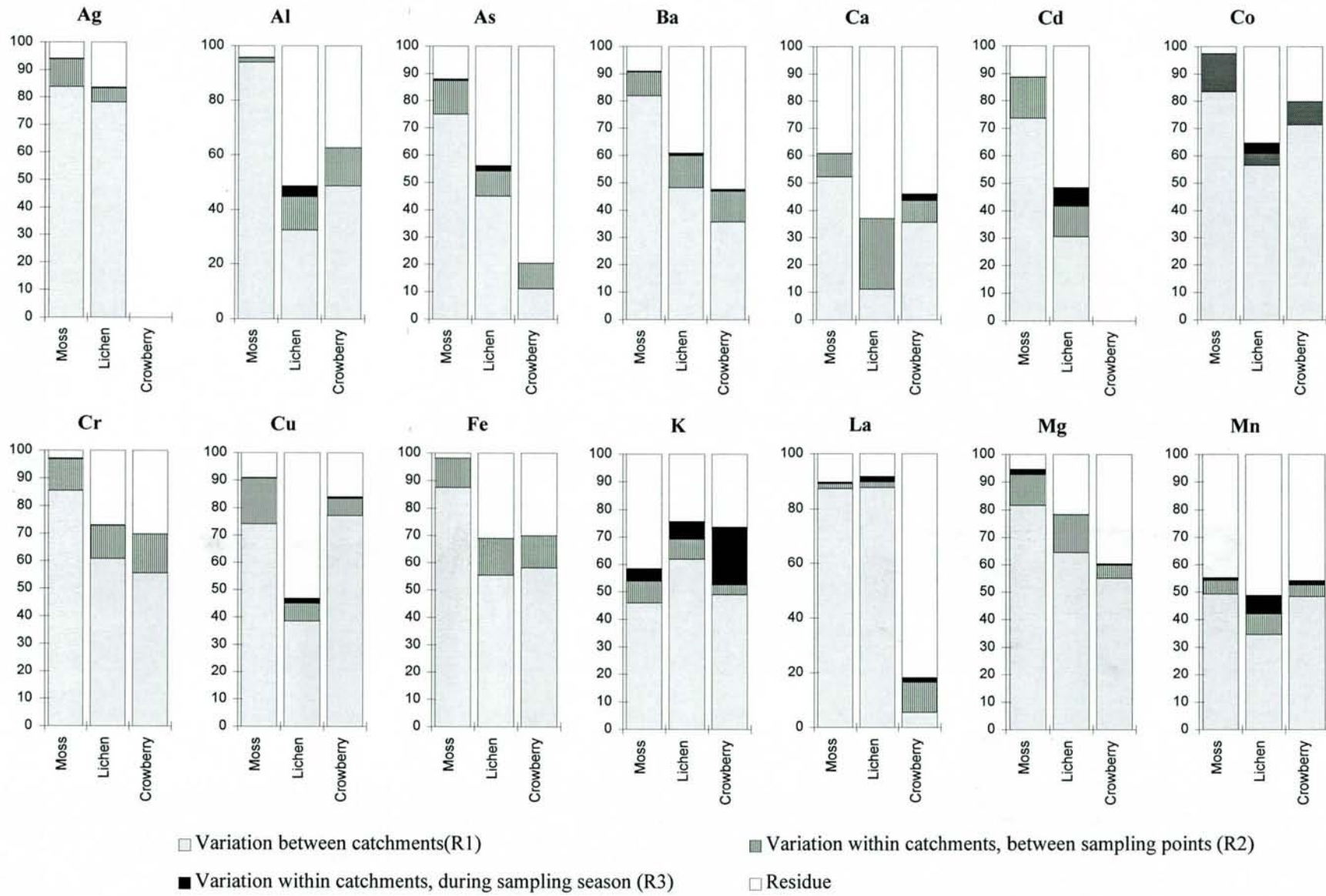
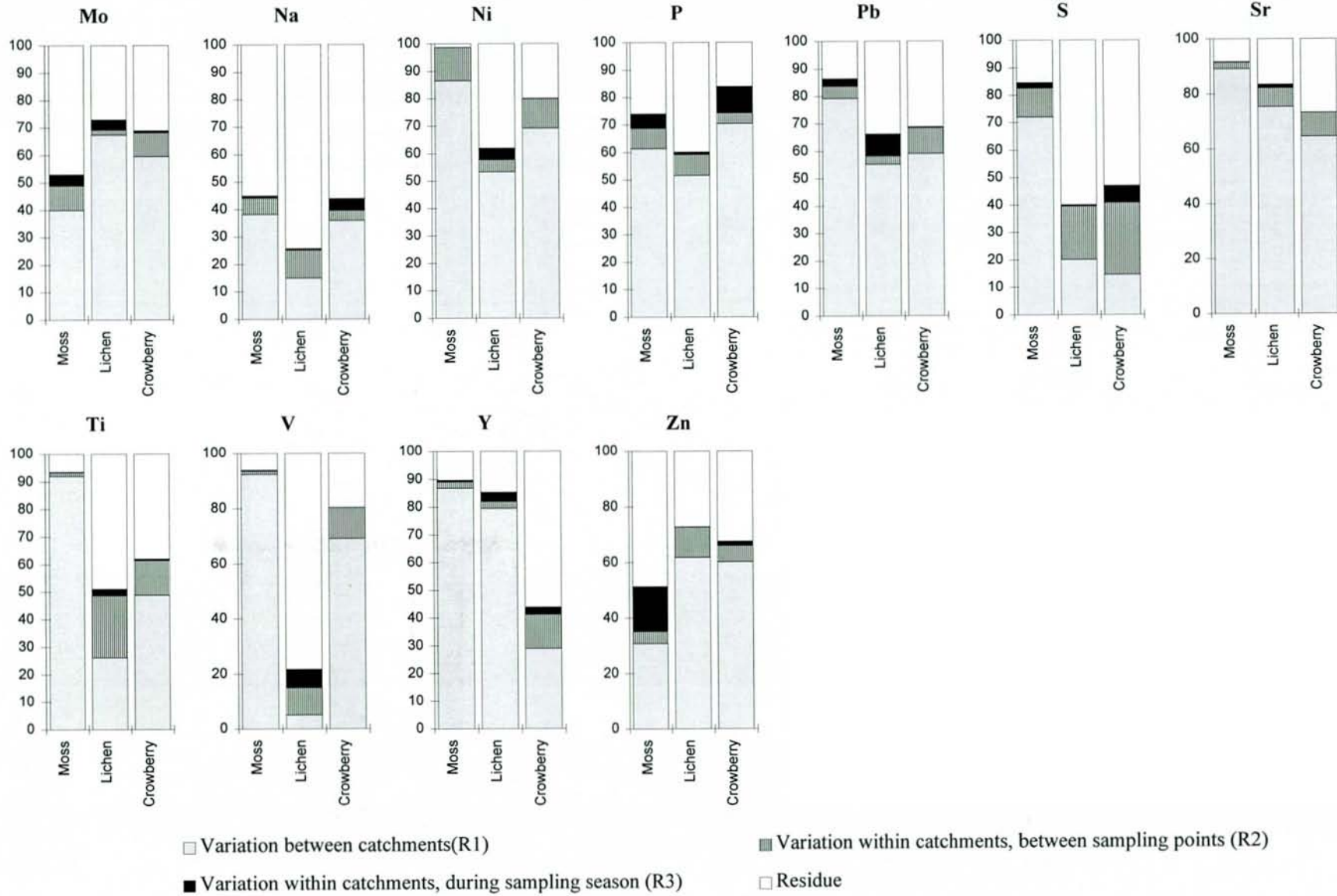


Figure 6.2.2. Estimation of the influence of different factors on element level variability in ground vegetation taxa, calculated in % by one-way ANOVA algorithm.





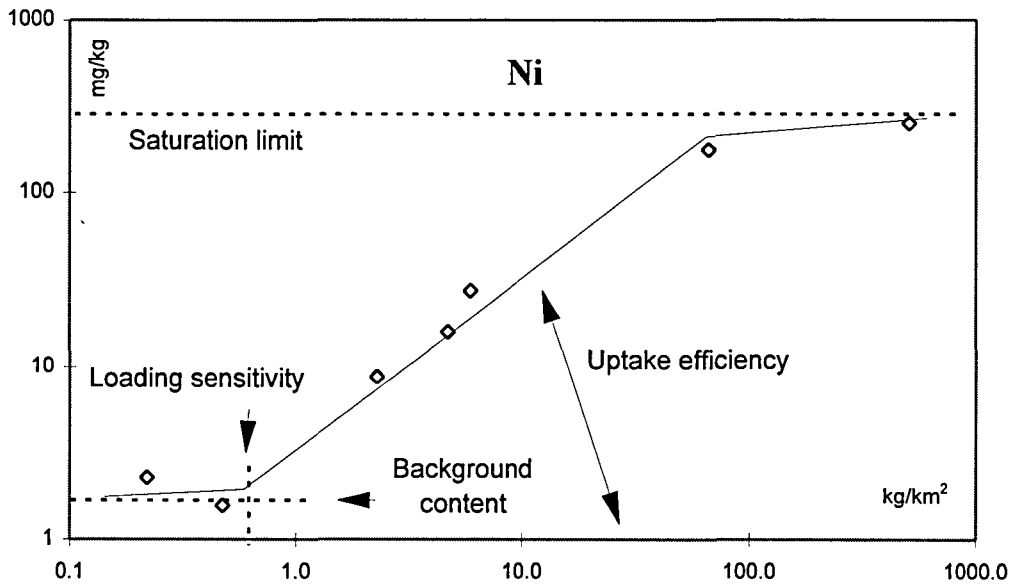


Figure 6.2.3. The main features for bioindicator properties, as illustrated by Ni content in moss from this study.

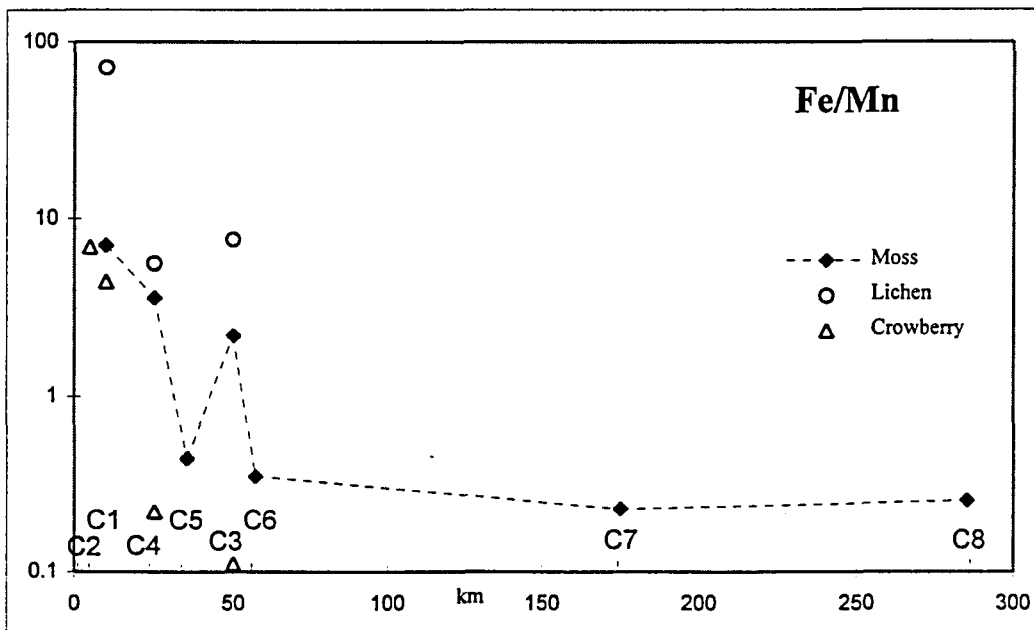
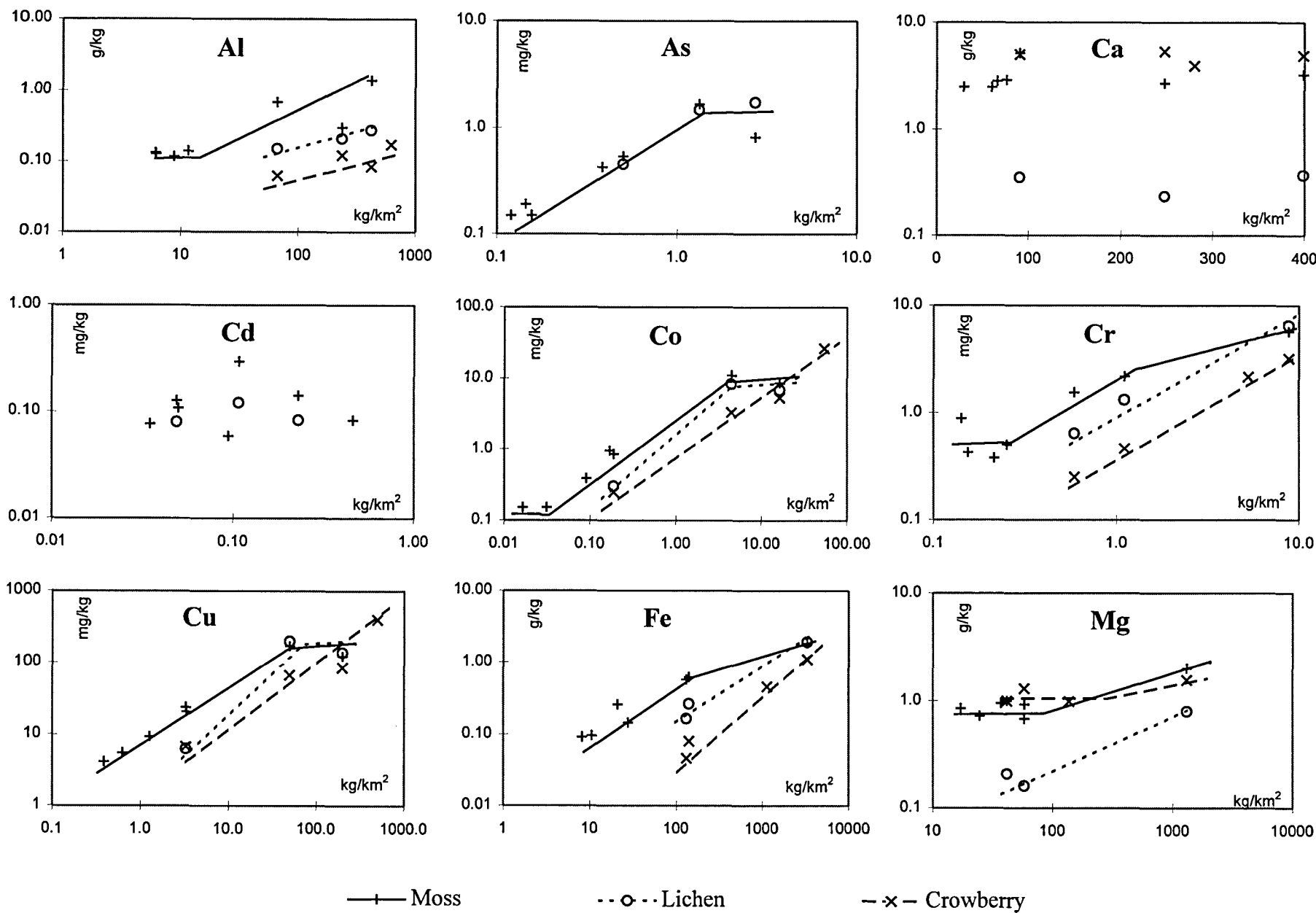


Figure 6.2.5. Behaviour of the Fe/Mn ratio in plants at varying distances from the sources of pollution.

Figure 6.2.4a. Relationship between element concentrations in bioindicators and annual depositions (atmospheric fallouts) from eight catchments in the Barents region.



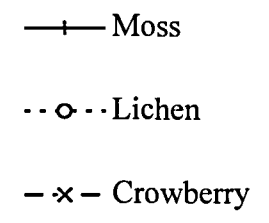
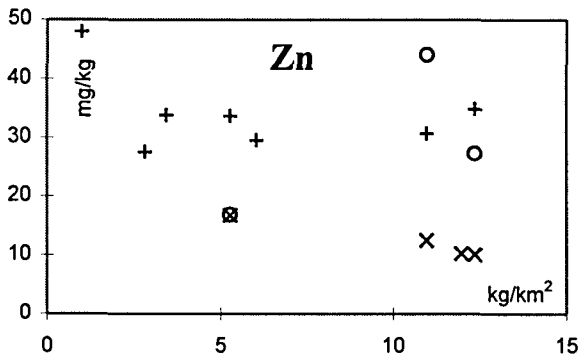
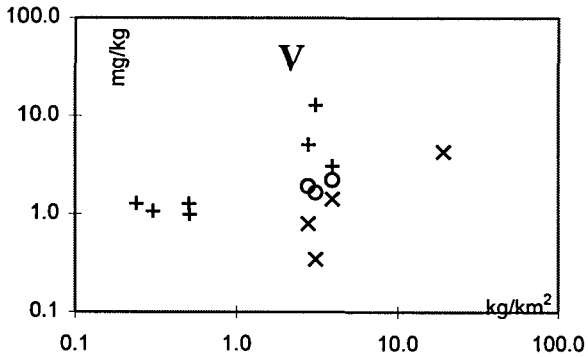
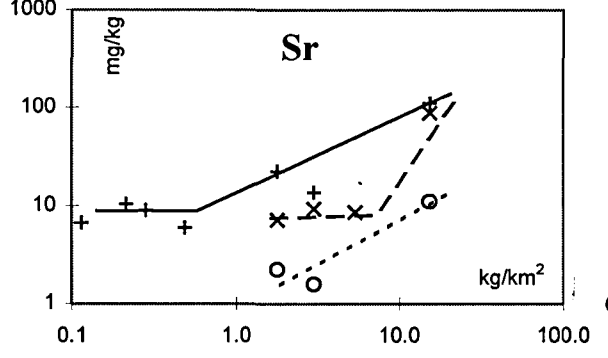
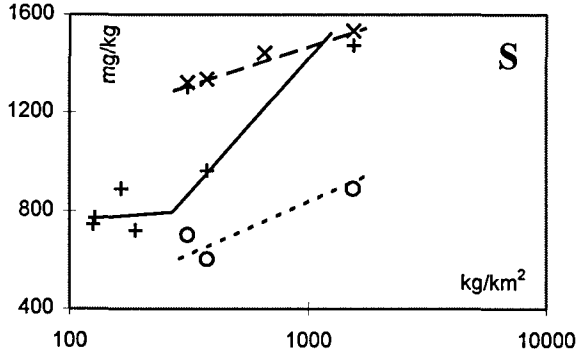
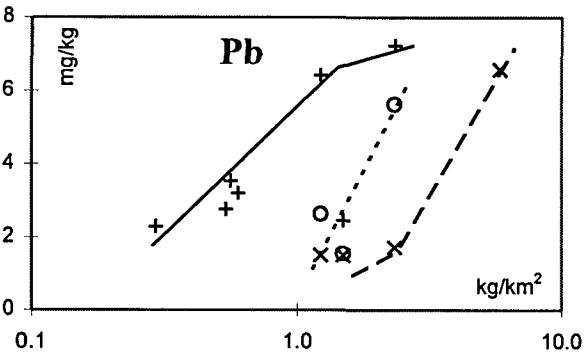
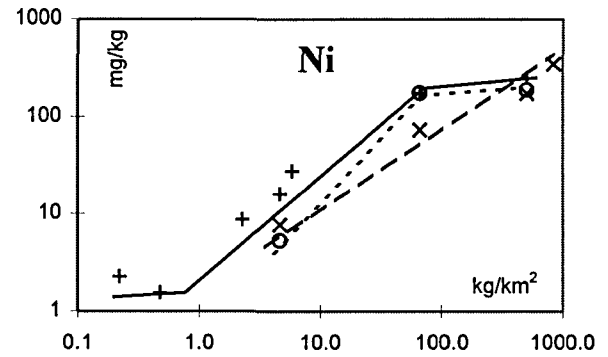
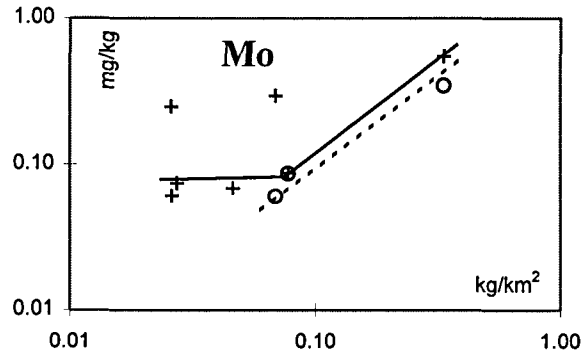
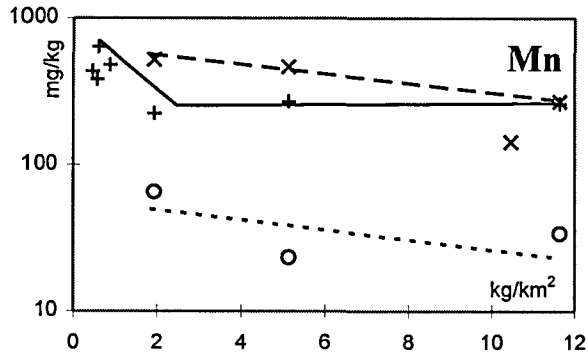


Figure 6.2.4b. Relationship between element concentrations in bioindicators and annual depositions from eight catchments in the Barents region.

### 6.3 Laser ablation ICP-MS analysis of tree ring profiles of pine and birch from N Norway and NW Russia

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#### ABSTRACT

Laser Ablation ICP-MS analysis of tree rings provides a rapid and sensitive method for investigating element concentrations and fluctuations in trees along time profiles. Time profiles obtained from pine and birch trees from a strongly polluted area in northwest Russia and a slightly polluted area astride the Norwegian-Russian border show that single trees behave rather individualistically in terms of heavy metal uptake.

#### INTRODUCTION

To be able to calculate element inputs into ecosystems and differentiate anthropogenic contamination from natural sources it is often important to have true background levels from pre-industrial times. However, these were not measured tens or hundreds of years ago, the necessary samples were not taken and analytical chemistry was not sufficiently advanced to provide us with these data. Consequently, we need samples that can give a retrospective view into the past, i.e. a sample medium where pre-industrial element levels have been preserved. A few media provide us with this opportunity, for instance lake sediments, ocean floor sediments, arctic ice cores and blanket bog peats. However, these only occur in very specific areas and age dating is often tedious and expensive.

Tree rings are a medium that could be very interesting for studying former element levels. Trees grow on a regular, stationary basis over several hundreds of years, show the element uptake of a living organism and their age is very easy to determine.

The first attempt to use tree rings to unravel pollution history was a study of the lead content of different growth rings in American elm (Ault et al., 1970). Increases in lead in the more recent rings were related to increases in traffic density. However, the next study that was published (Szopa et al., 1973) found no correlation between lead levels in the annual growth rings of white oak and the closing date of a highway close to the sampled trees. Another study in the early 1970s (Ward et al., 1974) also used tree rings to document changes of lead levels in the petrol used since the Second World War. Yet another early investigation (Rolfe, 1974) examined the lead contents of three different tree species from Illinois in segments representing 10-year growth periods. This showed very marked differences between historical segments and current ones.

An early review of the potential of tree-ring analysis for monitoring heavy metal pollution patterns, possible pathways of metal uptake (via roots, foliar uptake, direct deposition on the stem surface), problems of transport of heavy metals from the uptake site to the tree ring, stability of heavy metals in a tree ring and possibilities for movement between tree rings concluded that many problems remain to be solved before the potential can really be estimated (Leep, 1975).

An investigation of the lead, copper and zinc content of tree rings and bark in 5 different tree species in Scotland concluded that tree rings cannot be used to reconstruct pollution history, but bark can be used to monitor the level of current atmospheric deposition of heavy metals on a regional scale (Barnes et al., 1976).

A very detailed study of the levels of twelve elements in hardwood trees using neutron activation analysis found a very pronounced difference in element levels between the outer rings (sapwood) and the inner rings (heartwood) of the samples (Tout et al., 1977). In addition there was clear evidence for element movement between the rings after deposition. The conclusion was that it is unlikely that tree rings can be used as a reliable record of historical element levels and pollution.

The analysis of Zn, Cu, Cd and Mn in spruce from Fichtelgebirge in Germany using 5-year segments (Schrimpf, 1980) displayed very clear profiles with increasing element contents since 1840 and 2 to 4 times higher concentrations for rings formed after 1950 compared to those formed up to 1840.

A study of the correlation between growth depression and increased iron and other metal contents in pine trees from the Great Smoky Mountains National Park, USA (Baes and McLaughlin, 1984), concluded "that multi-element analysis of tree rings would be useful in determining when changes in air pollution and acid rain began to occur and in geographically mapping their extent, thereby providing key evidence of their source or sources".

Lead concentrations in ring segments (at 5-year intervals) from hickory trees in two urban forests in Atlanta, USA (Ragsdale and Berish, 1988), showed that "changes in Pb concentrations in wood of urban hickory trees dramatically document the historical periods of early urbanisation, the addition of Pb to gasoline, and the recent period of restricted use of leaded gasoline".

The analysis of Pb, Cd, Zn, Cu, S, N and As in tree rings of spruce trees from Erzgebirge, Saxonia, Germany (Ilgen and Nebe, 1989) showed that in spite of possible lateral movements of some elements between tree rings, the method was well suited for historical monitoring of the emission history in that particular area.

To evaluate black spruce as a bioindicator of aluminium contamination around the Alcan aluminium refinery in Jonquiere, Quebec, Canada (Dion et al., 1993), 4 profiles (N, E, S and W) spaced at 5 to 100 km from the plant were sampled. Two-year sections of the tree rings were used for neutron activation analysis of aluminium. A clear spatial pattern of Al enrichment in the tree rings corresponding to the last 20 years could be observed, especially at sites directly exposed to environmental contamination along prevailing winds. It was not possible, however, to produce a clear historical record of the Al contamination when the Al contents measured were compared with production and process data from the smelter. The conclusion was that black spruce is not appropriate for chronological monitoring.

A huge number of additional publications exist, roughly 60% of the authors concluding that tree rings can be used for unravelling the pollution history of an area and 40% stating that this is not possible or is at least very difficult. It is quite surprising to see the great number of studies carried out with such totally contradictory results. However, a certain pattern seems to exist. Authors using softwood trees more often report successful studies than those using hardwood trees. In addition, authors using composite samples for 5- or even 10-year periods more often report a success than authors analysing single tree rings. A general drawback of the technique up to now has been the

difficult sample preparation procedure involving the separation of single tree rings or segments of tree rings before analysis either by AAS or neutron activation techniques. The laser ablation ICP-MS technique used in our study offers the great advantage of facilitating the study of element variations ring by ring - and even within rings - without any tedious sample preparation. Hoffmann et al. (1994) published some results on the possible use of this technique for tree ring analyses.

As the study area, we chose the surroundings of the nickel smelters at Monchegorsk and Nikel on the Kola Peninsula in Russia where the Geological Survey of Norway, in collaboration with the Geological Survey of Finland and the Central Kola Geological Expedition, Russia, is carrying out a major environmental geochemical mapping project over an area of 188,000 km<sup>2</sup> (Fig. 6.3.1)(see World Wide Web site <http://www.ngu.no/Kola>). Initial results show that the main contaminant elements, as detected in snow, moss and A<sub>0</sub>-horizon soil samples, are As, Co, Cr, Cu, Fe, Mo, Ni, S, Sb and V, with Cu and Ni reaching levels of several percent in soil within the immediate vicinity of the smelters. If any systematic and chronological relationship exists between local environmental contamination and contaminant concentrations in tree rings then we should see it in the tree-ring profiles from this extremely polluted area. We focused our initial investigations on tree rings from birch and pine because these species are both abundant and ubiquitous in the area.

## MATERIALS AND METHODS

Four pine (*Pinus silvestris*) and two birch trees (*Betula* sp.) from Monchegorsk in Russia and Skjellbekken on the Norwegian side of the border near Nikel in Russia (Table 6.3.1, Fig. 6.3.1) were cut down just prior to tree-ring analysis in March and August 1994. Slices of the trees were oven dried at 50 °C, and transverse sections of the slices were cut into approximately 5 cm long and 1 x 1 cm thick bars. The surfaces for the subsequent laser ablation ICP-MS (LA-ICP-MS) analyses were smoothed with abrasive paper which was checked for cross contamination of the elements investigated here.

A PlasmaQuad PQ 1 (VG Instruments, England) located at the Geological Institute (GPI), Kiel, was used in our study. The instrument has been upgraded with the High Performance Interface (VG Instruments) giving a higher and more uniform response across the mass range. A discrete dynode multiplier (ETP, Australia) was used during these measurements. Instrument configuration and operating conditions are summarized in Table 6.3.2. Data acquisition was performed in a rapid scanning mode with low dwell time (i.e. counting time per MCS channel) and large skipped mass regions (c.f. Tab. 6.3.2). This rapid repetitive scanning with 113 sweeps per run (60 s) accounts for the short-timed nature of the transient signals generated by the laser ablation process. The monitored analyte isotopes (Tab. 6.3.3) were selected after surveying for detectable trace elements in different wood samples by full mass range scans.

A VG LaserLab 1 prototype comprising a Spectron SL 401 Nd:YAG laser, a simple optical system, and a step-motor driven sample stage with airtight sample cell was coupled to the ICP-MS instrument (c.f. Tab. 6.3.2). Details of the LA-ICP-MS system are given elsewhere (Garbe-Schönberg and McMurtry, 1994). The LA-ICP-MS system was optimised using certified N.I.S.T. 610 and 612 glass beads. The response of the system was typically around 1 to 2\*10<sup>3</sup> counts per second per ppm La in the beads. The laser was operated in Q-switched mode at a low pulse repetition rate of 2 Hz. The Q-switched pulse energy was approximately 120 mJ. For measurements in the denser birch wood, however, 140 mJ pulses had to be used for sufficient material ablation. Facing very low concentrations of the contaminant metals in these wood samples each tree ring analysis was accomplished with 3 ablation steps parallel to the growth ring, and 45 pulses per step during a total acquisition time of 60 s. The consecutive laser pulses resulted in a

slowly prograding ablation of material with depth and a quite constant ion signal in the MS. The generated ablation holes in the wood were excellently reproducible in shape and diameter, with diameters of approx. 200  $\mu\text{m}$ . All results were normalized to the  $^{13}\text{C}$ -ion signal as the internal standard except for samples pine 1-2 and 2-2 which were normalized to  $^{26}\text{Mg}$ . Precision for three replicate analyses in a tree ring of birch was < 15 % for all analysed elements except Al, Mn, Sb, and Ba for which it was 20-40 %. Precision of 3 and 6 measurements in spring and late wood of pine, respectively, showed variation coefficients in the range of 10-50 %. Unfortunately, in most of the analysed wood slices concentrations of the major contaminant elements S, Co, Ni, As, Se, Sn, and Sb were within the limits of detection (i.e. < 10 ppm) of our LA-ICPMS system, and these results will not be discussed here.

All the concentration profile measurements derived from spring wood, which forms rings wide enough for analyses even in narrow rings near the centre of the stem. Nevertheless, spring wood is characterized by lower trace element concentrations than late wood. In the subsequent graphical presentations all analytical results from the tree profiles are given as mean normalized data.

## RESULTS

Copper is one of the predominant heavy metal contaminants in the Nikel and Monchegorsk area, reaching several percent in the topsoil and still being up to 140  $\mu\text{g ml}^{-1}$  in ammonium acetate leaches of the O-horizon (< 2mm). Figure 6.3.2 shows results for Cu (mean normalized) in pine and birch from the highly polluted Monchegorsk area and from Skjellbekken. The Skjellbekken area is far less polluted than the Monchegorsk area and contamination levels are not much in excess of background. Both pine and birch from Monchegorsk display similar distribution patterns of Cu, with elevated concentrations in rings which grew around 1960 and between 1965 and 1972. Both trees have very high Cu in their outermost ring(s). The smelter, however, was operating throughout the lifetime of these trees at about the same emission values. In contrast, a birch from the "remote" Skjellbekken area in Norway shows no significant peaks, but a more general trend towards elevated Cu concentrations with age. Just the opposite to this trend for Cu in birch can be seen in the rings of a 126-year-old pine tree from Skjellbekken (Fig. 6.3.2 a); Cu decreases between 1950 and 1980. Nevertheless, Cu concentrations increase significantly in the recent tree rings from 1980 until 1993. Elevated Cu concentrations can also be observed in rings from 1940 to 1950, and this seems to fit well with part of the industrial history of the area, since the smelter was built in 1930. But why does the concentration decrease whilst the production of the smelter remained constant? Moreover, this peak is low compared to the Cu enrichments in rings from the early years of this pine (1880 to 1900). The impression from these profiles is that individual time fluctuations may be the most important feature to be observed in tree rings, not increases (or decreases) due to certain events like the starting up of the smelter. These profiles, or parts of them, would be open to almost any interpretation desired.

Another interesting example is shown in Figure 6.3.3a where we see an apparently systematic distribution of Al in rings from two pines from Monchegorsk and one from Skjellbekken. An early aluminium maximum can be seen around 1890. After 1930 aluminium concentrations increase in all three trees until the beginning of the 1950s. After 1960, lower Al values seem to prevail. Again, the tendency for Al levels to increase in tree rings after 1930 seems to fit well with the pollution history in which the starting up of the smelter in Monchegorsk in 1930 is a milestone. Ore with a different composition began to be processed around 1970. The increasing uptake of Al in the wood could be interpreted as an early sign of a breakdown of the ecosystem and a release of toxic Al ions to the soils due to ever-increasing soil acidification. Thus, the method seems to work, but other samples from the Kola Project have shown that, in the surroundings of the smelters, pH values in

precipitation are especially high (basic) due to a high input of alkaline dust (Reimann et al. 1995, 1996). Thus, one starts to wonder about the earlier observations that fit so well into a desired story. Whether the irregular Al distribution observed in two young trees from Monchegorsk (Fig. 6.3.3b) merely reflects this situation of immobilized Al in soils around the smelter is open to speculation.

Finally, Figure 6.3.4a shows the Pb data from profiles across two pines from the heavily contaminated Monchegorsk area. From the late-1930s onwards, Pb increases steadily, peaking in tree rings from around 1970, after which a general decrease can be observed. This curve could be easily interpreted as reflecting the effects of the introduction of unleaded fuel in Western countries and, thus, the general decrease of lead levels in the atmosphere as reported in several studies. One problem with this interpretation is that leaded fuel is still being used in the area investigated, and even in increasing amounts since the opening of the border. A different Pb story is recorded in rings from younger trees in the Monchegorsk and Skjellbekken areas (Fig. 6.3.4b). A steady increase in Pb concentrations can be observed in both trees, even after 1970, but a drop occurred in rings grown in the mid-1980s.

Mg and Ba showed no significant trends in the investigated tree-ring profiles. The trace elements Co, Ni, As, Sb, Se and Sn could be detected in the wood, but concentrations were generally not within the detection limits of the system, thus preventing a quantitative interpretation. Interestingly, rings from the old pine at Skjellbekken (N-4) are characterized by significant tin concentrations, with a profile similar to that of Cu.

The spatial resolution of the LA-ICP-MS system allows separate analyses of spring wood and late wood within a single annual tree ring. Our results from analyses of pine indicate that all the analysed metals have significantly higher concentrations in late wood than in spring wood (Table 6.3.2). This observation has some consequences for the interpretation of integrative results of tree-ring analyses when clusters of several rings with different thicknesses are compared (e.g. an inverse correlation of growth rate with metal concentration could be the consequence of an increase in the relative proportion of late wood).

## CONCLUSIONS

Laser ablation ICP-MS is a powerful and comparatively fast method for studying relative element contents in and between tree rings. The resolution of the method is good enough to determine element variations even in just one ring representing one year's growth.

The results presented here from one of the most polluted areas in the world demonstrate that profiles can be "found" which confirm, for example, a pollution source hypothesis. However, in general, trees seem to behave rather individualistically in their heavy metal uptake, and airborne contamination is definitely not the most important factor influencing heavy metal contents as observed in tree rings.

There is, of course, the possibility that the Russian area is just too heavily polluted and the trees behave abnormally due to severe stress. The annual emission of SO<sub>2</sub> in this area is around 140,000 t. This in itself could reduce the element uptake considerably due to a general decline of physiological and biochemical processes. The radial growth of pine has decreased by up to 50% since the 1960s up to a distance of 30 km from the pollution source (Njöd and Kaupi, 1995), and root phytomass and length have also decreased since the beginning of the 1980s (Yarmishko and Yarmishko, 1993).



An additional point that can complicate the interpretation of element contents as observed in tree rings is the fact that roots are capable of changing their location (three-dimensional distribution) under heavy metal stress (Yarmishko and Yarmishko, 1993), starting to grow into deeper soil horizons and, hence, meeting quite different levels of elements (less heavy metals, but, for example, more exchangeable Al (Pereverzev, 1993).

Another important result of our study is that the element levels in rings from trees growing near background areas (Skjellbekken) are similar to those from trees in the immediate surroundings of the industrial plants. In addition, these trees may show temporal fluctuations in the concentration levels of metals that started long before the industrialization of the area. Already more than 100 years ago, metal levels in the wood were as high as after the smelters started production. We conclude that - although the idea is fascinating - tree rings cannot be used for studying the pollution history of any one area, but they may be useful for studying element uptake and life cycles of individual trees under environmental stress. This, however, is definitely not a task for geoscientists.

When results of tree ring studies using classical, preparative methods are being interpreted it is important to bear in mind that significant differences in element concentrations exist between the dense late wood and the less dense spring wood.

#### ACKNOWLEDGEMENT

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## TABLES

Table 6.3.1 Characterisation of the analysed trees.

Table 6.3.2 Instrument configuration and operating conditions of the Kiel LA-ICP-MS system (VG PlasmaQuad PQ1 + VG LaserLab 1) optimised for tree ring analysis.

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Table 6.3.4 Relative enrichment and depletion of elements in late wood of pine(in % #R-3 from Monchegorsk).

## FIGURES

Figure 6.3.1. Study area for the Kola Project, including the location of the most important emission sources (the nickel smelters in Nikel and Monchegorsk, the ore roasting plant at Zapoljarnij and numerous other industries, power plants and open pit mines). The analysed trees come from the surroundings of Monchegorsk and from N-Norway, 35 km SW of Nikel.

Figure 6.3.2. Distribution of copper (mean normalized data) in tree-ring profiles in a) old pine and young birch from Skjellbekken, and b) two young trees from the vicinity of the smelter at Monchegorsk.

Figure 6.3.3. Distribution of aluminium (mean normalized data) in tree rings of a) three old trees from Monchegorsk and Skjellbekken, and b) two young trees from the vicinity of the smelter at Monchegorsk.

Figure 6.3.4. Distribution of lead (mean normalized data) in tree rings of a) two old pines from the highly polluted Monchegorsk area which display a lead distribution that fits published data on bulk lead emissions reasonably well, and b) both the third pine tree from Monchegorsk (R-3) and the birch from the relatively little polluted Skjellbekken area near Nikel (birch 3-2) do not display this general trend but show rather individualistic patterns.

**Table 6.3.1.** Characterisation of the analysed trees

Sample ID	Tree species	Tree age [yrs]	Location	Pollution level
1-2	pine	120	Monchegorsk, luv of smelter	high
2-2	pine	95	Monchegorsk, dead forest	high
R-3	pine	34	Monchegorsk	high
R-4	birch	36	Monchegorsk	high
3-2	birch	35	Skjellbeken/ Nikel (Norway)	low
N-4	pine	126	Skjellbeken (Norway)	low

**Table 6.3.2:** Instrument configuration and operating conditions of the Kiel LA-ICP-MS system (VG PlasmaQuad PQ1 + VG LaserLab 1) optimised for tree ring analysis

<b>ICP</b>	
Incident power at 27 MHz	[kW]: 1.30
Reflected power	[W]: < 5
Coolant gas-flow	[L min <sup>-1</sup> Ar]: 13
Auxiliary gas-flow	[L min <sup>-1</sup> Ar]: 1.1
Carrier gas-flow (MFC)	[L min <sup>-1</sup> Ar]: 0.850
<b>INTERFACE:</b>	
Sampling depth	[mm]: 12
Sample cone (Ni) aperture	[mm]: 1.0
Micro-skimmer cone (Ni) aperture	[mm]: 0.75
<b>MASS ANALYSER:</b>	
Quadrupole type	12-12S
Mass resolution at 10% peak height	[m/z]: 0.8
<b>DATA ACQUISITION:</b>	
Detector type	AF 562 (ETP)
Detector mode	pulse counting
Acquisition mode	scanning
Mass range	[m/z]: 13 - 210
Skipped mass regions (additional)	[m/z]: 139-200
No. MCS channels per mass	20
Dwell time	[μs]: 160
No. sweeps	113
<b>LASER UNIT:</b>	
Laser type	Nd:YAG (SPECTRON SL 401)
Wavelength	[nm]: 1064
Laser mode	Q-switched
Pulse energy	[mJ]: 120-140
No. steps	3
Step increment	[mm]: 1
Pulses per step	45
Pulse repetition rate	[Hz]: 2

**Table 6.3.3: Compilation of monitored isotopes**

Element	m/z	Abundance [%]	Interferences, Remarks
C	13	1.11	Internal standard
Mg	26	11.0	CN, CC
Al	27	100	CNH
S	33	0.75	OOH
Mn	55	100	ArNH
Fe	56	91.5	ArO
Co	59	100	
Ni	60	26.2	
Cu	63	69.1	
Zn	66	27.8	
As	75	100	
Se	82	11.6	Kr (11.6% abundance)
Sr	88	82.6	
Sn	120	33.0	
Sb	121	57.3	
Ba	138	71.7	
Pb	208	52.4	

**Table 6.4.4: Relative enrichment and depletion of elements in late wood of pine (in %; tree # R-3 from Monchegorsk).**

Element	[%]
Magnesium	-13
Aluminium	52
Sulfur	equ.
Manganese	45
Iron	24
Cobalt	150
Nickel	340
Copper	280
Arsenic	130
Selenium	81
Tin	140
Antimony	230
Barium	-20
Lead	45

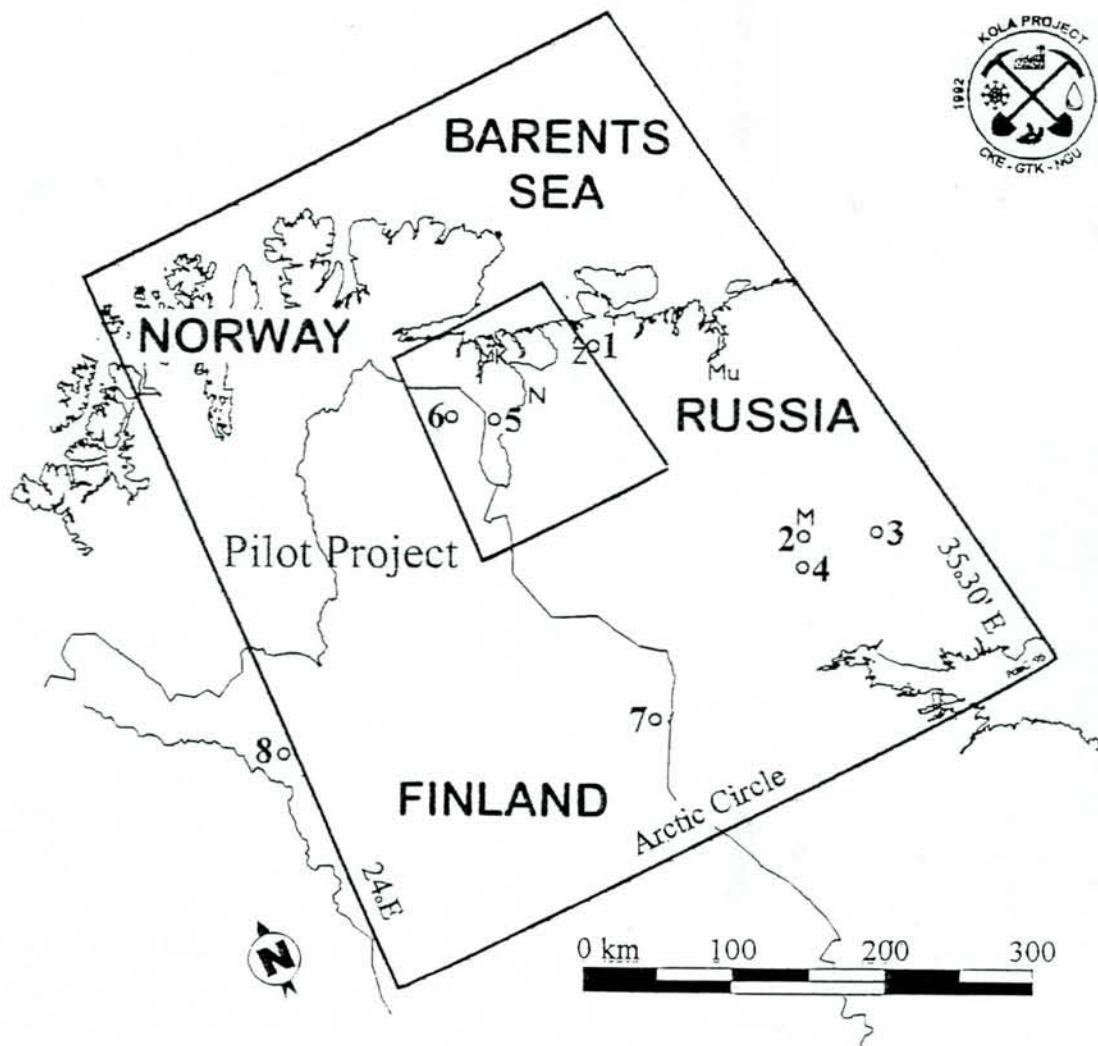


Figure 6.3.1. Study area for the Kola Project, including the location of the most important emission sources (the nickel smelters in Nikel and Monchegorsk, the ore roasting plant at Zapoljarnij and numerous other industries, power plants and open pit mines). The analysed trees come from the surroundings of Monchegorsk and from N-Norway, 35 km SW of Nikel.

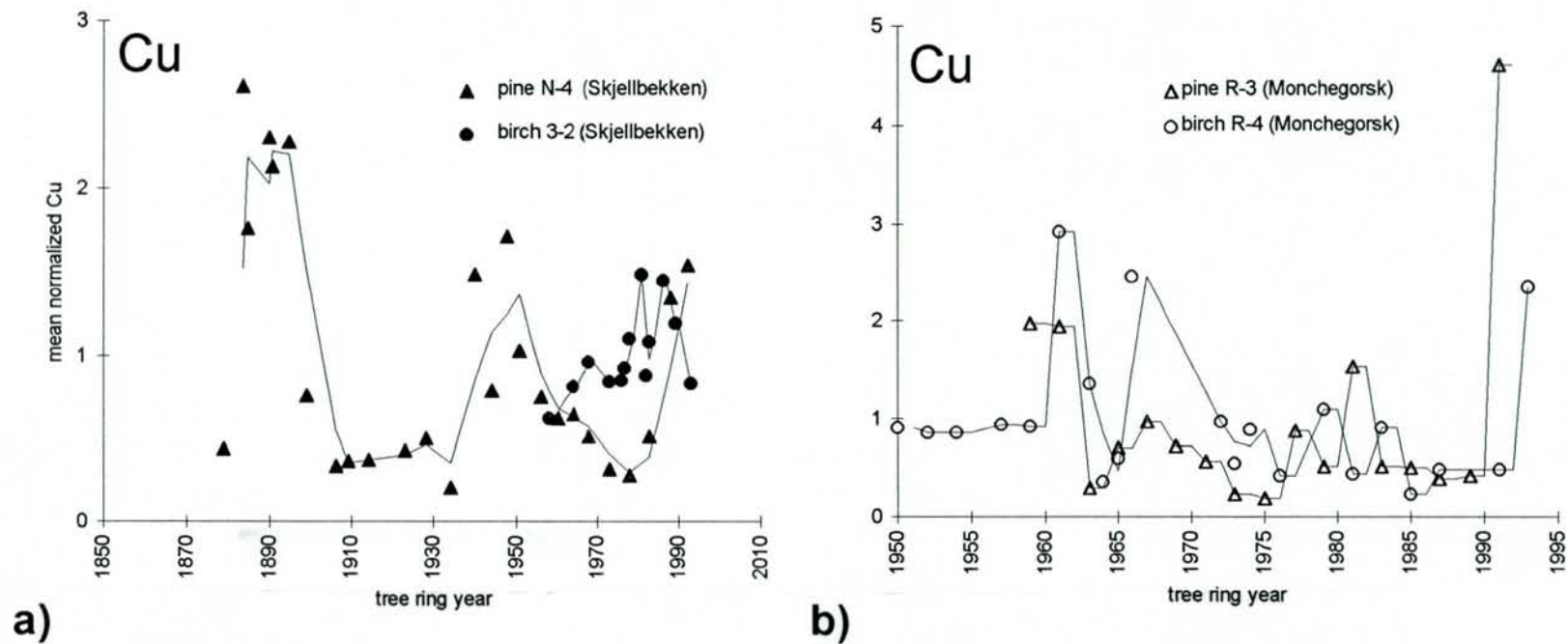


Figure 6.3.2. Distribution of copper (mean normalized data) in tree-ring profiles in a) old pine and young birch from Skjellbekken, and b) two young trees from the vicinity of the smelter at Monchegorsk.



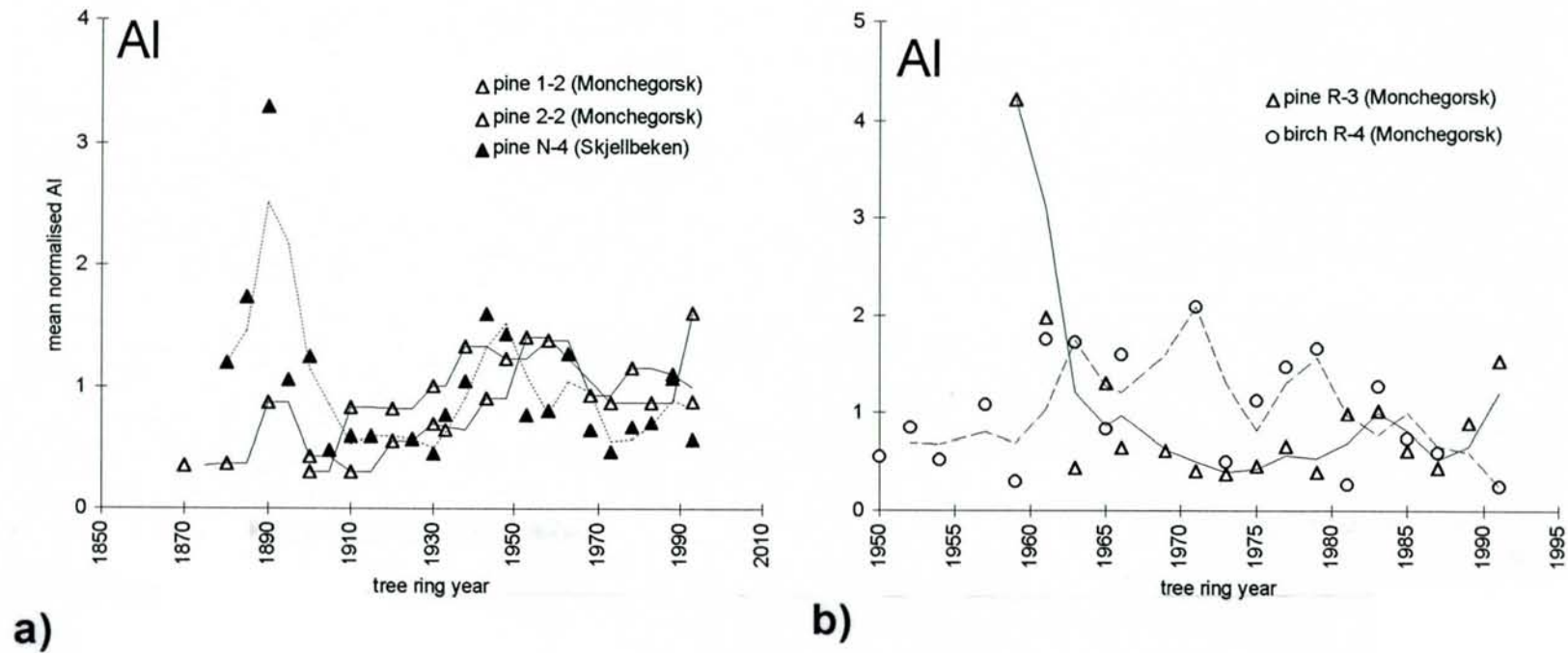


Figure 6.3.3. Distribution of aluminium (mean normalized data) in tree rings of a) three old trees from Monchegorsk and Skjellbekken, and b) two young trees from the vicinity of the smelter at Monchegorsk.

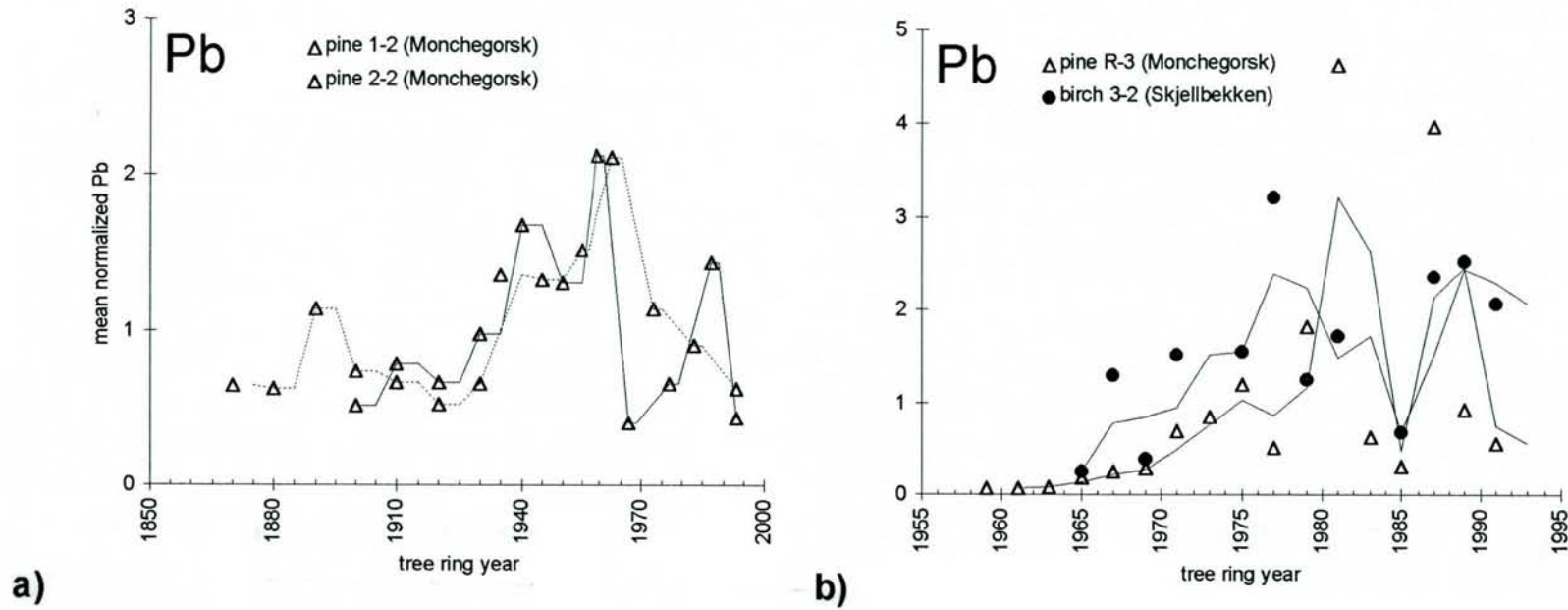


Figure 6.3.4. Distribution of lead (mean normalized data) in tree rings of a) two old pines from the highly polluted Monchegorsk area which display a lead distribution that fits published data on bulk lead emissions reasonably well, and b) both the third pine tree from Monchegorsk (R-3) and the birch from the relatively little polluted Skjellbekken area near Nickel (birch 3-2) do not display this general trend but show rather individualistic patterns.

## 7 TOPSOIL

### 7.1 Topsoil (0-5 cm) composition in eight Arctic catchments in northern Europe (Finland, Norway and Russia)

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#### ABSTRACT

Frozen topsoil samples (0-5 cm) were collected during March/April 1994 in eight Arctic catchments in northern Europe (4 in Russia, 3 in Finland, 1 in Norway), at different distances and wind directions from the emissions of the Russian nickel ore mining, roasting and smelting industry on the Kola Peninsula. Between 14 and 25 sites were sampled in catchment basins ranging in size from 12 to 35 km<sup>2</sup>.

Results show that close to the smelters in Monchegorsk and Nickel, topsoil is clearly enriched in Ag, As, Bi, Cd, Co, Cr, Cu, Fe, Ni, Pb, Sb, Se, Te and V. Cu and Ni median contents in topsoils collected close to Monchegorsk are about 600 times higher than those observed in background catchments. The effect of open-cast mining and waste dumps of alkaline rocks from the nephelinite industry near Kirovsk can be seen in the elevated contents of Al, Ba, K, La, Mn, Na, Sr, Ti, Y and Zn in topsoil collected in a nearby catchment. For many elements, variations observed in single catchments are as great as the total regional variation.

Several elements (e.g. Cd, Hg, Cu, Ni, Pb, S, Zn) show strong correlation between the organic content of the sample and the element content observed. It is thus necessary to determine the organic content of the samples and correct the element levels when using depth-related soil samples (here, the 0-5 cm topsoil layer) for regional mapping.

#### INTRODUCTION

The geological surveys of Finland (GTK) and Norway (NGU), and the Central Kola Expedition (CKE) in Russia, are carrying out a major geochemical mapping project (see World Wide Web site <http://www.ngu.no/Kola>) in a 188,000 km<sup>2</sup> area north of the Arctic Circle, comprising the entire area between 24° and 35.5° E north to the Barents Sea (Fig. 7.1.1). As part of this project, eight catchments (hereafter abbreviated as C1 - C8) widely distributed in this area (Fig. 7.1.1) were investigated in detail in 1994. Media sampled were: snow (meltwater and filter residue), rainwater, stream water, organic stream sediments, terrestrial moss, topsoil (0-5 cm), complete podzol profiles, Quaternary deposits and bedrock.

Some of the world's largest point sources of SO<sub>2</sub> emissions are located within the study area (Gunn et al., 1995), the nickel smelter at Nickel, the ore roasting plant at Zapoljarnij and the nickel smelter at Monchegorsk (Fig. 7.1.1) together accounting for about 300,000 t of SO<sub>2</sub>, 1900 t of Ni, 1100 t of Cu and 94 t of V<sub>2</sub>O<sub>5</sub> emissions yearly (Murmansk Regional Committee for Ecology and Natural Resources, see Reimann et al., 1996a).

The results of topsoil (0-5 cm) sampling carried out during March/April 1994 are presented here. Topsoil was collected during the winter together with snowpack samples (Äyräs et al.,

1996a) from the same sites. The samples were kept frozen at all times and were freeze-dried in the laboratory prior to analysis. This was done to prevent any loss of metals (e.g. Hg) or organic pollutants due to sampling or sample preparation. Other investigations from the surroundings of the Russian nickel industry (e.g. Chekushin et al., 1993, Reimann et al., 1996b) have shown that the combined content of Ni, Cu and Co can reach several weight percent in HNO<sub>3</sub> extractions of the O-horizon. Relatively little is known, however, about the local variation of element contents compared to the regional variation.

Very different approaches are used to document the content in soils of heavy metals deriving from anthropogenic contamination. Either the uppermost, organic soil horizon is sampled as determined by soil-genetical considerations, or a depth-defined sample (Blum et al., 1989) is taken (either 0-2 cm (e.g. ISO 10381: Guidance on the procedure for the sampling of urban and industrial sites with regard to soil contamination), or 0-3 cm, 0-5 cm, 0-15 cm or 0-25 cm (e.g. Darnley et al., 1995)). Both approaches have advantages and disadvantages. A depth-defined sample can be taken anywhere, independent of the existence of an organic layer or a certain soil type. Element levels obtained will be characteristic for, for example, "the uppermost 5 cm of soil" throughout the area. Problems may, however, arise due to the very different contents of mineral soil vs. organic material in the depth-related samples, as many heavy metals tend to accumulate in the organic fraction. Samples consisting only of organic soil may thus display considerably higher heavy metal contents than samples with a very thin organic layer and otherwise material from mineral soil horizons (in the survey area usually the E- or even the B-horizon of podzol profiles). Horizon-oriented sampling, on the other hand, will result in substantial differences in the thickness of the final samples taken - with anthropogenic input accumulating in the top few cm of the soil profile, this can result in difficulty in correcting differences between sites with very different thicknesses of the O-horizon. O-horizon samples were collected in a pilot project in 1992/93, when a subarea of 12,000 km<sup>2</sup> was mapped (Chekushin et al., 1993, Niskavaara et al., 1996, Reimann et al., 1996b). For the catchment study, it was decided to test the use of depth-defined sampling (0-5 cm topsoil layer) for regional mapping.

The aims of this study were (1) to establish an overview of heavy metal contents in topsoils (0-5 cm) collected in catchments located in different settings (geology, vegetation, distance from industry and from the ocean) throughout the main project area to guide the selection of proper analytical techniques for the main project, (2) to improve our understanding of element sources and cycling, (3) to guide the distinction of natural and anthropogenic element sources in the upper 5 cm of soils, (4) to study local vs. regional variability on a relatively detailed scale (1 sample site per 1 to 2 km<sup>2</sup>), to aid the interpretation of the results of the low density (1 sample site per 100-600 km<sup>2</sup>) regional mapping project, and (5) to decide whether horizon-related or depth-related sampling should be chosen for regional mapping.

The main characteristics of the eight catchments studied are summarised in Table 7.1.1.

## SAMPLING

Sampling of topsoil was carried out at the same time as snowpack was sampled. At all 5 subsample sites for a composite snowpack sample, holes were dug down to the ground using a plastic snow shovel. At the bottom, a 20 x 20 cm large soil sample was cut out of the frozen ground using a steel spade. Vegetation and large rock fragments were removed and the uppermost 5 cm were taken as the sample. The thickness of the organic layer was recorded. The final sample was a composite sample of the five subsamples per station. All the field teams

worked with exactly the same equipment. The samples were stored in polyethylene plastic bags, kept frozen at all times and delivered frozen to the GTK laboratory in Rovaniemi. More details on sampling are given in Äyräs and Reimann (1995).

## ANALYSIS

At the GTK laboratory, a subsample of the frozen topsoil was freeze-dried. This proved to be a rather difficult and time-consuming procedure. The dried subsamples were homogenised by milling in a domestic blender with blades made of uncontaminating material and sieved to pass through a <2 mm screen.

A 2 g subsample was digested for two hours at 90°C with 12 ml aqua regia (3:1 HCl and HNO<sub>3</sub>) (modified ISO standard draft 11466, GTK method 512). After centrifugation, the clear solution was analysed with ICP-AES for 30 elements (GTK method 512P) and with ICP-MS for 29 elements (GTK method 512M). In addition, Hg was determined with cold vapour AAS (GTK method 512H). Carbon, hydrogen and nitrogen were analysed with a CHN analyser (GTK method 820L).

A split of the samples was sent to the NGU laboratory in Trondheim, where loss on ignition was determined gravimetrically and pH and electrical conductivity were determined in a water extract. pH was additionally determined in a CaCl<sub>2</sub> extract.

For more details on analytical techniques and quality control procedures see Niskavaara (1995). The GTK laboratory is accredited according to ISO 9001 and ISO-Guide 25.

## RESULTS AND DISCUSSION

### *Element levels and variation*

Table 7.1.2 summarises the analytical results for each of the eight catchments separately (median, minimum and maximum values). The table shows the strong influence of anthropogenic emissions on the topsoil chemistry - Ag, As, Bi, Cd, Co, Cr, Cu, Fe, Hg, Mo, Ni, Pb, Sb, Se, Te and V show the highest median and the highest maximum contents in C2 (maximum content Fe; V in C5). The influence of a special lithology on the topsoil chemistry is best seen in C3 (Table 7.1.1), which is underlain by alkaline rocks and surrounded by waste dumps from the nephelinite industry in Apatity. Here Al, Ba, K, La, Li, Mn, Na, Si, Sr, Ti, Y and Zn show the highest median and maximum contents (maximum Mn in C4, and Ti and Zn in C1).

More detailed results, comparing local (within catchment) with regional (between catchments) element levels and variation are displayed graphically using boxplots (Velleman and Hoaglin, 1981) for the “emission-related” elements from Table 7.1.2 in Figure 7.1.2 and for the “lithological” elements, organic content (C+H+N) and pH in Figure 7.1.3. This graphical comparison of levels and variation can be used instead of an analysis of variance to judge the influence of regional vs. local variation (Reimann, 1989) in order to determine for which elements stable geochemical maps can be drawn. A stable map is defined as a map where after re-sampling and/or re-analysis of all samples the regional element distribution as displayed on the map does not change significantly (Garrett and Goss, 1980, Garrett et al. 1980).

In Figure 7.1.2, the data for many of the main contaminants had to be log-transformed prior to plotting due to the great variations in element contents between the catchments. Co, Cu, Ni

and Te show a typical pattern with element contents in  $C2 > C1 > C4 > C3 = C5 > C6, 7, 8$ . A similar pattern was observed for many other media sampled for the catchment study (e.g. snow (Äyräs et al., 1996a), rain (Reimann et al., 1996a) and stream water (Caritat et al., 1996) for the main pollutants). It is obvious that for many of the pollutants it should be rather easy to produce reliable regional maps based on one location per 300 km<sup>2</sup>, as the differences between catchments are much larger than the variation within catchments. Ni contents in C2 are, for example, 9 times higher than those in C4 just 25 km to the south, 130 times higher than in C3, about 50 km to the east and 560 times higher than in C7 close to the Finnish/Russian border, about 150 km southwest of Monchegorsk!

For a number of elements (Cd, Fe, Hg, Pb, S, V) it will, however, prove difficult to construct reliable, stable regional maps as the variation within catchments is nearly as great as, or even greater than (e.g. Fe in C5), the variation between catchments. The Fe and V contents in topsoils in particular show a rather strong influence of lithology on the results obtained - the additional anthropogenic inputs can easily be masked by local variations when only one sample is taken to represent a 30 km<sup>2</sup> catchment within a 300 km<sup>2</sup> area. If stable regional maps for these elements are to be produced using topsoils, it would definitely be necessary to take a rather large composite sample from several locations within the 300 km<sup>2</sup> area which the sample is intended to represent.

Figure 7.1.3 shows these boxplot comparisons for a selection of 14 additional elements that do not show a simple relationship between “contamination” (distance from smelter) and element levels. From studying element levels in snow (Äyräs et al., 1996) and rain (Reimann et al., 1996a) in the same 8 catchments, it is known that many of these elements are part of the emission spectrum of the Russian nickel industry. This is completely masked here by the differences in lithology/variations in natural element contents. In general, differences in element levels between the catchments are much smaller than for the majority of elements presented in Figure 7.1.2. The very special lithology of C3 is shown by the rather high contents of Al, Ba, K, La, Mn, Na, Sr, Y and Zn. For many of these elements we see in C3, at the same time, the highest variation in element content for the whole data set. It will not be possible to produce stable geochemical maps for these elements using topsoil samples.

The boxplot for the C+H+N content shows that the Finnish samples are generally richer in organic material. Samples from Russia and Norway show a substantial variation in organic content due to the relative proximity of the Barents Sea coast (climatic conditions) and the industrial deserts around the smelters. pH again shows a strong dependency on lithology (alkaline rock waste dumps?) with C3 being mainly composed of alkaline rocks displaying the highest pH levels and C6, underlain by granites, having the lowest pH values in the topsoils.

#### *Action levels*

For a number of metals, action levels are fixed by different authorities to distinguish between natural element contents and contaminated soils. These levels are based on the grain size fraction of <2 mm and aqua regia extraction, which was actually the reason for our choice of grain size fraction and dissolution technique. Geologists are aware that aqua regia extraction will not give total element contents in samples consisting of varying amounts of humus and mineral soil, due to differences in the solubility of the soil-forming minerals. Thus, aqua regia extraction introduces an undesirable source of variability, dependent on the unknown mineralogy of the sample, but is unfortunately accepted as the international standard for such investigations in the environmental sciences.

Table 7.1.3 compares the median element levels observed in the most contaminated catchment (C2), the maximum content for the whole data set and action levels as presently discussed in Norway (SFT = Statens forurensningstilsyn (State Pollution Control Authority)) and as used in Holland. It is interesting to note that, according to these action levels, one of Europe's most strongly heavy-metal polluted areas apparently has no contamination problems other than for Cu and Ni.

Figure 7.1.4 shows CDF diagrams for the whole data set and the 8 catchments separately for the major pollutants, Cu and Ni. It is clear that there is a range of concentrations, displayed in C6, 7 and 8, which reflect the natural background of the area, occasional data outliers (e.g. in C7) probably reflecting minor mineralisations. The sequence of catchments in these diagrams exactly follows the distance from the contamination source and we can conclude that almost all samples from C1, C2, C3, C4 and C5 have had considerable Cu and Ni added to their natural contents. Similar graphics can be drawn for all the other heavy metals. This demonstrates that defining fixed action levels for soils is a dangerous approach - environmental problems may be considerably underestimated when these levels are used, or considerable financial and practical problems may be imposed on authorities in areas with high natural heavy-metal levels. Action levels should thus generally be based on toxicological considerations and be carried out on a site-specific basis and not be merely based on statistics (it is interesting to note that the 50 percentile for both Cu and Ni in our data set is very close to the action level). Present action levels are not related to toxicology, contamination or natural element variations. The definition of land-use related action levels, as suggested by Eikmann and Kloke (1988), will not change this general picture as none of these approaches takes sufficient account of the large fluctuations in natural background, binding of the heavy metals and, thus, toxicology. The lower two diagrams in Figure 7.1.4 show very clearly the futility of defining fixed action levels for soils. Geochemical mapping and decisions based on the local/regional variations and anomalies as displayed in these maps may be alternatives.

#### *Influence of organic material on element levels*

One of the main questions when collecting depth-defined topsoil samples is how far the different proportions of organic material to mineral soil influence the analytical results. To be able to investigate this influence, carbon, nitrogen and hydrogen contents were determined for all samples and the sum of these three elements is taken to represent the total content of organic material in the sample. Table 7.1.2 and Figure 7.1.3 (C+H+N) show that there are considerable differences in the contents of these elements between the catchments, samples from the Finnish catchments in general displaying considerably higher contents than those from Russia and Norway.

The extent to which the element content depended on the observed C+H+N content was studied in XY diagrams, Figure 7.1.5 gives some selected examples. No influence of the content of organic material on measured element levels was observed for Ag, As, K, La, Li, Mg, Mn, Na, Si, Te and Y. Of these, Ag, As and Te are part of the emission of the smelter (compare C2 to the other catchments in Table 7.1.2). Otherwise, these elements can be characterised as a lithogenic suite of elements.

A negative correlation with increasing organic content is visible for Al, Cr, Fe, Sc, Ti and V. The more mineral soil in the samples, and thus the lower the organic content, the higher the contents of these elements. The degree of correlation is, of course, partly influenced by the different solubility of the soil-forming minerals in aqua regia.

A slightly positive correlation of element content with increasing organic material can be seen for the following elements: Bi (C2, C4), Ca (an artefact of different CaCO<sub>3</sub> contents?), Co (C2), Mo (C2), Sb (C2), Se (C2, C4) and Sr (C1, C3, C4, C5 - an artefact of the correlation of Sr and Ca?). It is interesting to note that there exists a correlation between organic content and element content for several of the “emission elements” (Bi, Co, Mo, Sb, Se) in the Russian catchments, but not in the background catchments. This may be exaggerated by the fact that the variation in the organic content of the top 5 cm of soils is especially high in the Russian catchments.

B, Ba, Cd, Cu, Ni (but not in Finland!), P, Pb (but negative in C6), S and Zn show rather strong positive correlation between the organic content and the measured element content of the samples. For some of these elements, a correction for organic content would definitely be necessary before preparing regional maps if topsoil is used as the medium.

S and P are accumulated as major nutrients in the organic layer. Their enrichment here has nothing to do with contamination. A vegetation-related enrichment might also be assumed for B, Ba and Zn, none of which is emitted in significant quantities. It is interesting to note that the definite major input of sulphur (Reimann et al., 1995a give a figure of 500 mg S/m<sup>2</sup> and year for C2 compared to 100 mg S/m<sup>2</sup> and year in the Finnish catchments) is not bound in the uppermost soil layers. It is partly found in the aquatic environment, where very high levels were observed in overbank sediments (Niskavaara et al. 1996).

We see no appreciable enrichment of P in C3, even though C3 is adjacent to one of the world’s largest open-cast apatite mines. This can only be explained by the fact that apatite dust was not dissolved by the aqua regia extraction used here, but would have required a stronger attack.

## CONCLUSIONS

Element levels for several heavy metals in C2, located close to the Monchegorsk nickel smelter, are found to be up to 600 times higher (Cu and Ni) than in Finnish background catchments. Ag, As, Bi, Cd, Co, Cr, Fe, Mo, Pb and Sb are 2-60 times enriched in the uppermost 5 cm of soil near Monchegorsk (Table 7.1.2). Boxplot comparisons of element levels observed in the eight catchments situated at different distances from industry are a good tool for distinguishing graphically between the influences of contamination and lithology. When the 0-5 cm layer of topsoil is used as the sampling medium only the above-mentioned heavy metals can be clearly recognised as being predominantly derived from pollution, and several of the major elements that are emitted are not revealed (Reimann, 1995a, b, 1996b).

Action levels, as presently used in many countries (e.g. Holland and Norway), are shown to have limited relevance to natural element levels or levels of contamination in one of Europe’s most strongly polluted areas. They should therefore generally be used with great caution and should not be used in legislation.

Nearly 50% of the elements analysed show equally large, or even larger, variations in element levels within single catchments when compared to all 8 catchments sampled. Boxplot comparisons, as used in Figures 7.1.2 and 7.1.3, are a valuable tool for deciding for which elements stable regional maps can be expected when low-density topsoil samples are used. They include many of the contaminants from the nickel industry (Ag, As, Bi, Co, Cr, Cu, Ni, Sb, Se and Te) as well as a few lithology-dominated elements (La, Sr and Y). To produce stable regional maps of the other elements, a different sampling medium or a different approach



to sampling would be necessary, compositing samples from several subareas. Even this approach may be inadequate for a number of elements where differences in levels between the catchments chosen are minimal, partly due to their very different nature. It will be interesting to test whether this problem can be overcome by producing “factor” maps, combining several elements in one factor, or by moving median mapping, compositing several sample sites statistically.

For the regional mapping project it was decided to use a combination of depth-related and horizon-related sampling: O-horizon samples, but only the uppermost 3 cm; when the organic layer was less than 3 cm thick only that layer was collected and the actual thickness was recorded in the field sheet.

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## TABLES

Table 7.1.1: Main characteristics of the 8 catchments investigated (for location within the study area, refer to Fig. 7.1.1)

Table 7.1.2: Median element contents and range (min-max) of all elements/parameters analysed for in C1 to C8. For catchment names, refer to Table 7.1.1, for location within the study area refer to Fig. 7.1.1. Highest median and maximum values are in bold print.

Table 7.1.3: Comparison of action levels as presently discussed in Norway (SFT: State Pollution Control Authority of Norway) and used in Holland (3 levels: «normal», further investigation necessary, immediate action necessary) to element contents observed in the project area. Under remarks, the catchments where samples which exceeded the SFT recommendations were taken are noted, the numbers in this column giving the number of samples above the action level in the different catchments.

## FIGURES

Fig. 7.1.1: Location of the study area for regional geochemical mapping (frame) and of the eight catchments discussed here (1: Zapoljarnij, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, and 8: Pallas).

Fig. 7.1.2: Boxplot comparison of element levels and variations of 16 elements showing the highest median content in C2 between all eight catchments.

Fig. 7.1.3: Boxplot comparison of 14 elements showing lithology as the dominant influence on the observed levels, and also the C+H+N content and pH in the eight catchments.

Fig. 7.1.4: CDF diagrams for Cu and Ni, showing the whole data set (N=174) in the upper diagram and the distribution in the 8 catchments (N see Table 7.1.2) in the lower diagram.

Fig. 7.1.5: XY diagrams showing the correlation between the organic content (C+H+N) and eight selected elements. Note the logarithmic scale used for Cu, Ni, Te and Ti.

No.	Name	Coordinates of catchment outlet	Size (km <sup>2</sup> )	Elevation (m a.s.l.)	Annual precip. (mm)*	Vegetation	Bedrock	Surface cover, peculiarities
RUSSIA								
C1	Zapoljarniy	69°27'01"N 31°03'49"E	19.02	25-373	454	birch forest tundra	gneiss	till, fluvio-glacial, outcrop
C2	Monchegorsk	67°50'30"N 32°54'48"E	22.38	128-507	391	technogenic desert, birch shrubs	dacite & andesite & tuffs, gabbro/norite	till, prone to erosion
C3	Kirovsk	67°32'50"N 33°48'55"E	20.01	240-1075	502	spruce forest, mountain tundra birch forest	nephelinite	till, diluvial/eluvial
C4	Kurka	67°41'25"N 32°50'14"E	20.49	152-466	502	north taiga spruce forest, birch; incipient deterioration	amphibolite, gneiss	till, fluvio-glacial
NORWAY								
C5	Skjellbekken	69°21'25"N 29°27'25"E	34.56	80-297	422	north taiga pine forest, birch	andesite, basalt & tuffs, 'black shale'	till, esker
FINLAND								
C6	Kirakka	69°35'12"N 28°51'46"E	11.86	110-200	386	north taiga pine forest	granite	outcrop, till, moraine ridge
C7	Naruska	67°21'44"N 29°22'05"E	20.16	263-490	513	north taiga spruce forest	gneiss	till, peat, outcrop
C8	Pallas	68°09'14"N 23°52'50"E	24.42	303-500	405	north taiga spruce forest	quartzite	till, peat

\* from the closest meteorological station (data from 1994)

**Table 7.1.1: Main characteristics of the 8 catchments investigated (for location within the study area, refer to Fig. 7.1.1)**

Table 7.1.2 Summary statistics, topsoil 0-5cm								
Name	Unit	C1	C2	C3	C4	C5	C6	C7
Det.Lim.	Method	N=22	N=25	N=23	N=25	N=25	N=14	N=20
Ag	mg/kg	0.17	<b>1.28</b>	0.05	0.37	0.24	0.08	0.27
0.01	512U	0.08-1.01	0.39- <b>3.61</b>	0.01-0.23	0.11-1.12	0.03-1.93	0.02-0.2	0.05-0.55
Al	mg/kg	2135	4930	<b>11600</b>	2090	2380	755	3680
10	512P	770-15000	2090-14800	2220- <b>31200</b>	890-6960	460-11300	340-5290	470-1700
As	mg/kg	1.4	<b>24</b>	1.4	1.3	0.8	0.4	0.4
0.2	512P+U	0.8-6.1	<b>11-60</b>	0.3-5.2	0.8-14	0.4-13.7	0.2-0.6	<0.2-1.7
B	mg/kg	4	4	5	4	5	5	4.5
3	512P	3-8	<3-9	<3-8	<3-9	<3-7	<3-7	<3-8
Ba	mg/kg	38	68	<b>113</b>	60	39	45	62
0.5	512P	15-100	22-240	12- <b>638</b>	21-224	17-167	17-70	21-214
Bi	mg/kg	0.09	<b>0.34</b>	0.14	0.21	0.09	0.04	0.14
0.02	512U	0.06-0.17	0.11- <b>0.76</b>	0.05-0.24	0.12-0.41	0.04-0.16	<0.02-0.15	0.07-0.19
C	%	20	31	15	24	27.5	<b>46</b>	37
1	820L	9-52	5-50	3-41	4-45	7- <b>53</b>	27-51	11-49
Ca	mg/kg	1800	1940	2650	1820	1950	<b>3435</b>	3040
3	512P	760-4030	760-7780	300-12900	940-630	510- <b>17000</b>	1090-5810	630-9530
Cd	mg/kg	0.31	<b>0.62</b>	0.29	0.35	0.24	0.24	0.28
0.01	512U	0.1-0.59	0.19- <b>1.64</b>	0.11-0.95	0.1-1.03	0.05-1.16	0.14-0.65	0.09-1.16
Co	mg/kg	15	<b>98</b>	3	14	3	0.6	1
0.5	512P	6-41	27- <b>242</b>	0.7-11	6-107	1-9	<0.5-4	<0.5-10
Cr	mg/kg	14	<b>42</b>	8	6	5	0.6	4
0.5	512P	5-45	24- <b>86</b>	2-19	2-57	1-20	<0.5-4	0.7-22
Cu	mg/kg	173	<b>2270</b>	16	141	20	4	4
0.5	512P	54-618	727- <b>6440</b>	3-32	44-441	7-91	3-10	3-42
Fe	mg/kg	7750	<b>16400</b>	6700	3780	4150	745	2840
10	512P	2620-29200	8150-25900	1770-13600	1610-34400	760- <b>75100</b>	260-6220	510-16800
H	%	3.2	4.3	2.6	3.7	4.1	<b>6.8</b>	6.0
0.1	820L	1.6-6.9	0.9-6.7	0.9-5.9	0.8-6.6	1.2- <b>7.2</b>	4.3-7.1	2.0-7.2
Hg	mg/kg	0.19	<b>0.28</b>	0.17	0.17	0.15	0.22	0.16
0.02	512H	0.05-0.47	0.07- <b>0.49</b>	0.03-0.44	0.03-0.3	0.07-0.29	0.2-0.3	0.08-0.25
K	mg/kg	500	300	<b>900</b>	600	800	450	600
200	512P	200-2100	<200-1000	<200- <b>3100</b>	300-1300	300-1100	300-700	200-1200
La	mg/kg	5.1	4.3	<b>38.2</b>	2.3	2.1	1.9	4.2
0.7	512P	1.3-10.7	2-12.3	5.1- <b>124</b>	1.2-22.2	<0.7-9.6	<0.7-8.3	0.8-291
Li	mg/kg	<0.7	<0.7	<b>2.6</b>	<0.7	<0.7	<0.7	<0.7
0.7	512P	<0.7-6.4	<0.7-5.5	<0.7- <b>8.4</b>	<0.7-4.2	<0.7-4.7	<0.7-0.8	<0.7-5.3
Mg	mg/kg	<b>1370</b>	1120	1130	480	920	815	740
5	512P	370-6350	610-3680	120-1870	250-3470	500- <b>6600</b>	370-1280	250-1930
Mn	mg/kg	105	97	<b>326</b>	169	151	22	47
0.5	512P	34-1170	25-389	4-1880	52- <b>10800</b>	52-4310	<15-111	<15-1130
Mo	mg/kg	<1	<b>4.7</b>	<1	<1	<1	<1	<1
1	512P	<1-2.5	1.3-13.5	<1-2.4	<1-3.2	<1- <b>15.8</b>	<1-12.8	<1-4
N	%	0.7	1.2	0.7	0.8	0.9	1.2	<b>1.3</b>
0.1	820L	0.4-1.6	0.3-2.2	0.2-1.6	0.2-1.6	0.3-1.7	0.9-1.5	0.5-2.5
Na	mg/kg	105	180	<b>650</b>	90	90	100	80
15	512P	80-210	90-450	40- <b>5770</b>	60-270	40-130	70-210	60-290
Ni	mg/kg	395	<b>2520</b>	19	298	27	2.5	4.5
1	512P	159-1250	504- <b>6360</b>	3-34	69-751	8-52	1-7	2-42
P	mg/kg	584	745	873	539	586	436	757
7	512P	288-989	246-1430	350-1980	240-896	219-995	349-848	385-1970
Pb	mg/kg	9	<b>26</b>	17	16	9	5	13
0.2	512U	6-15	8- <b>82</b>	6-43	4-30	6-14	3-11	5-52

S	mg/kg	729	1240	658	779	881	1205	1300
10	512P	351-1950	792-3440	257-1440	158-1850	277-2190	626-1680	507-2570
Sb	mg/kg	0.09	<b>0.79</b>	0.15	0.27	0.1	0.06	0.09
0.02	512U	0.05-0.21	<b>0.31-1.69</b>	0.04-0.29	0.08-0.58	0.06-0.31	0.02-0.12	0.02-0.26
Sc	mg/kg	0.6	<b>0.7</b>	<0.5	<0.5	<0.5	<0.5	<0.5
0.5	512P	<0.5-3.2	<b>&lt;0.5-1.9</b>	<0.5-0.8	<0.5-1.4	<0.5-4	all <0.5	<0.5-0.7
Se	mg/kg	0.44	<b>4.73</b>	0.43	0.96	0.32	0.28	0.36
0.02	512U	0.15-1.27	<b>1.38-9.02</b>	0.17-0.88	0.3-3.36	0.11-1.15	0.2-0.41	0.22-1.14
Si	mg/kg	220	250	<b>270</b>	220	240	165	260
10	512P	120-330	180-320	<b>190-580</b>	120-310	170-320	100-280	160-360
Sr	mg/kg	12	14	<b>268</b>	15	10	27	34
0.5	512P	6-30	5-62	<b>5-1230</b>	4-40	3-78	13-44	8-463
Te	mg/kg	0.08	<b>0.89</b>	0.02	0.09	0.02	<0.01	<0.01
0.01	512U	0.03-0.22	<b>0.33-2.15</b>	0.01-0.13	0.02-0.74	0.01-0.06	<0.01-0.04	<0.01-0.05
Th	mg/kg	<5	<5	<5	<5	<5	<5	<5
5	512P	all <5	all <5	<5-19	all <5	all <5	all <5	all <5
Ti	mg/kg	<b>863</b>	262	506	287	247	43	74
0.5	512P	<b>93-2690</b>	61-1030	126-1330	73-839	19-1100	8-226	13-656
V	mg/kg	28	<b>35</b>	15	13	13	1.4	4.3
0.5	512P	5-100	11-72	4-37	4-35	<b>2-214</b>	0.5-12	2-23
Y	mg/kg	0.9	1.5	<b>7.5</b>	1.0	0.6	<0.5	0.8
0.5	512P	<0.5-2.6	<b>0.7-4.4</b>	<b>0.8-33.5</b>	0.5-8.1	<0.5-3	<0.5-8.9	<0.5-57
Zn	mg/kg	29	43	<b>60</b>	33	33	37	21
0.5	512P	<b>10-189</b>	16-47	8-134	12-86	13-98	11-64	10-75
EC	mS/cm	90	103	69	96	119	125	106
	NGU-EL	53-167	65-253	23-159	30-203	41-197	96-163	47-174
pH CaCl2		2.9	2.8	3.6	3.0	3.1	2.6	3.1
	NGU-EL	2.6-3.4	2.3-3.7	2.2-4.9	2.7-4.9	2.5-5	2.1-3.3	2.7-3.9
Lol	%	37	67	37	52	57	89	78
0.1	NGU-GR	18-88	31-84	10-77	9-89	16-94	48-98	25-96

Table 7.1.3 Comparison of element contents in topsoil (0-5cm) with action levels

Element	Dutch soil levels			SFT-levels	Median C2	Maximum (catchment)	samples above SFT	remarks	Dutch		
	normal	invest.	action						normal	investig.	action
As	29	40	55	20	24	60 (C 2)	5%	only C2	5%	1%	none
Ba	200	410	625		68	638 (C 3)			5%	1%	1%
Cd	0.8	6	12	1	0.6	1.6 (C 2)	7%	6 C2, 1 C4, C5, C7	7%	none	
Co	20	130	240		98	242 (C 2)			25%	5%	1%
Cr	100	240	380	100	42	86 (C 2)	none		none		
Cu	30	110	190	100	2270	6440 (C 2)	38%	in C1, C2, C4	45%	38%	25%
Hg	0.3	5	10	1	0.3	0.5 (C 2)	none		8%	none	
Mo	10	100	200		4.7	13.5 (C 2)			none		
Ni	35	120	210	30	2520	6360 (C 2)	45%		45%	40%	35%
Pb	85	310	530	50	26	82 (C 2)	2%	2 C2, 1 C7	none		
Zn	140	380	720	150	43	189 (C 1)	2%	2 C3, 1 C1, 1 C2	1%	none	



Kola Project (CKE, GTK, NGU)  
Catchment Study 1994  
Catchment locations

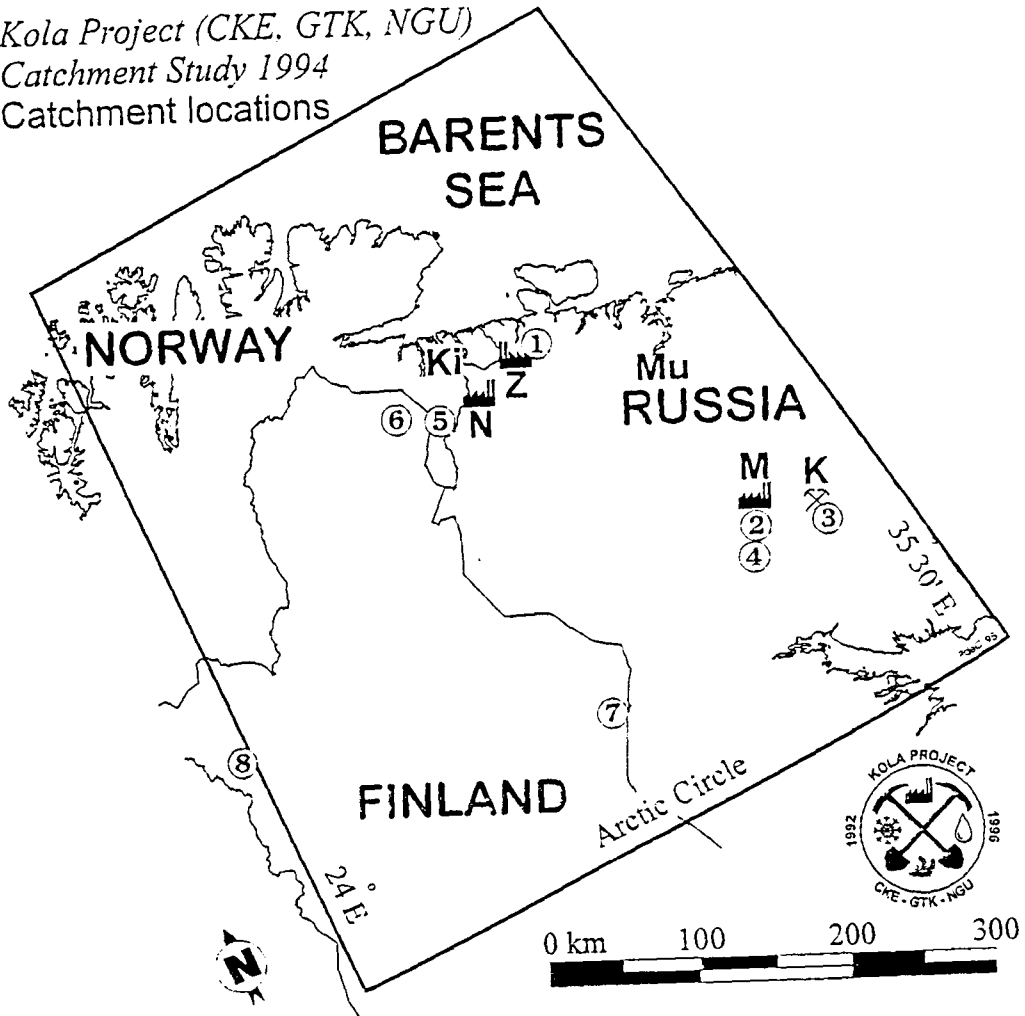
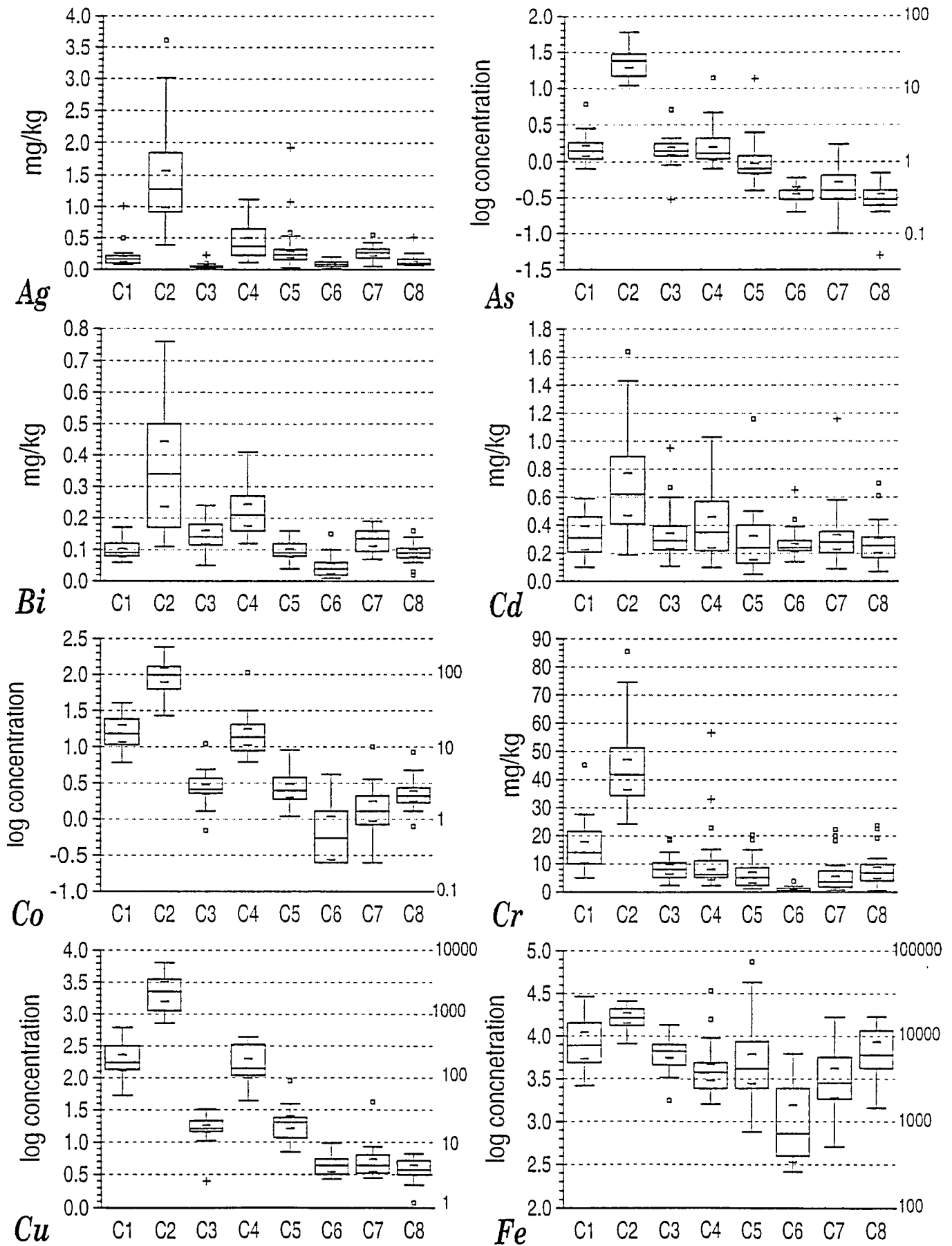


Fig. 7.1.1: Location of the study area for regional geochemical mapping (frame) and of the eight catchments discussed here (1: Zapoljarnij, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska, and 8: Pallas).

Fig. 7.1.2: Boxplot comparison of element levels and variations of 16 elements showing the highest median content in C2 between all eight catchments.



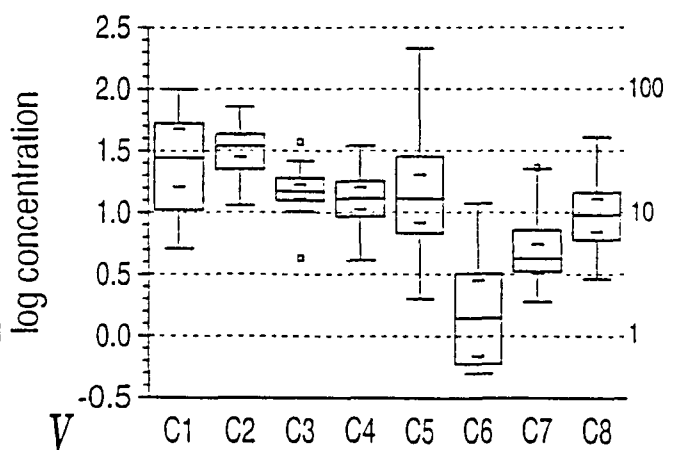
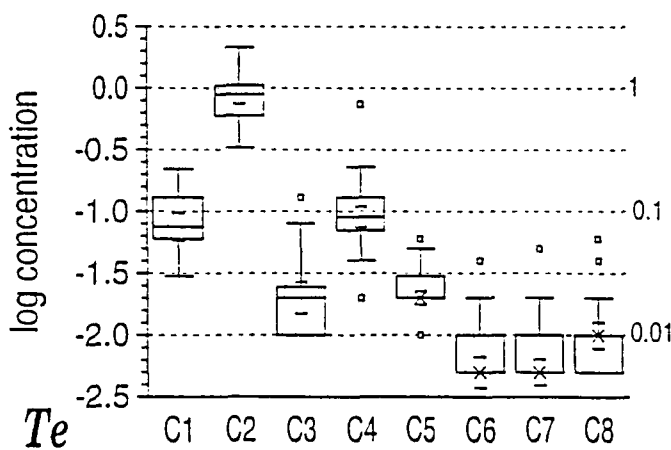
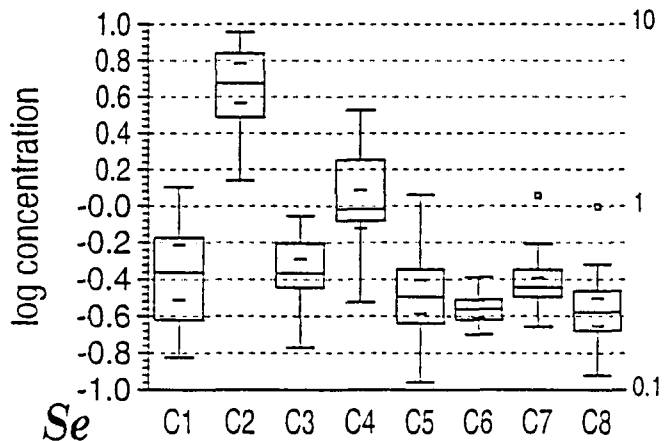
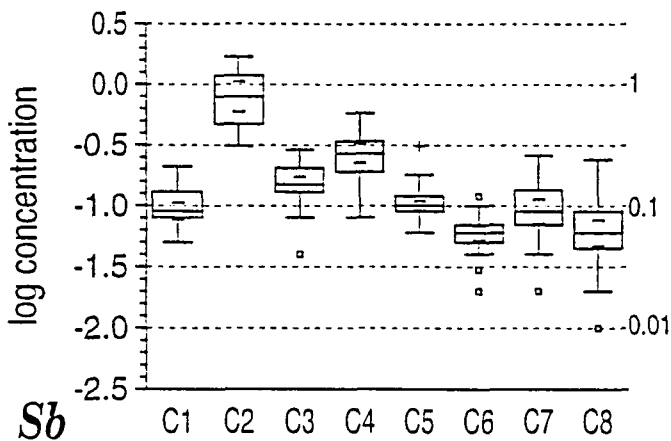
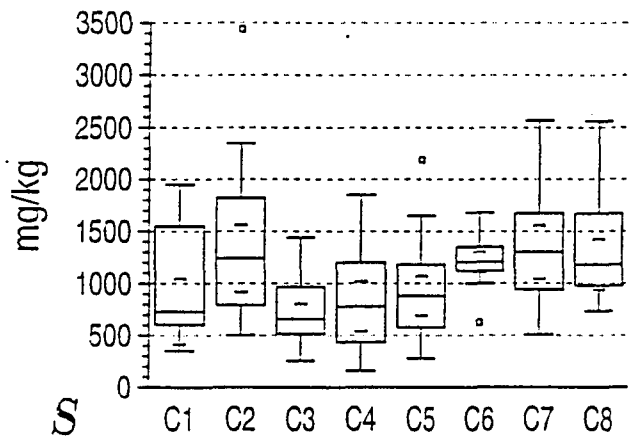
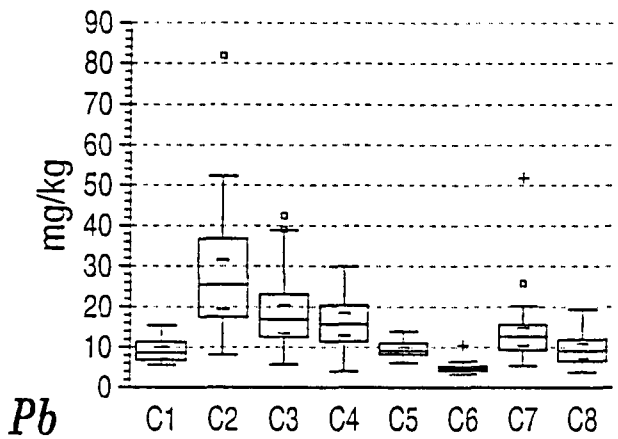
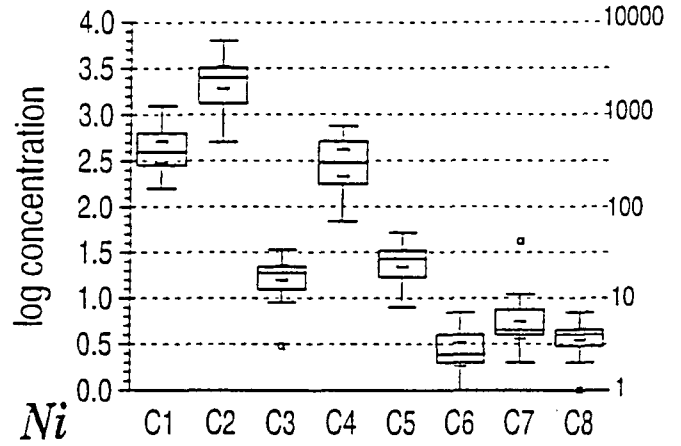
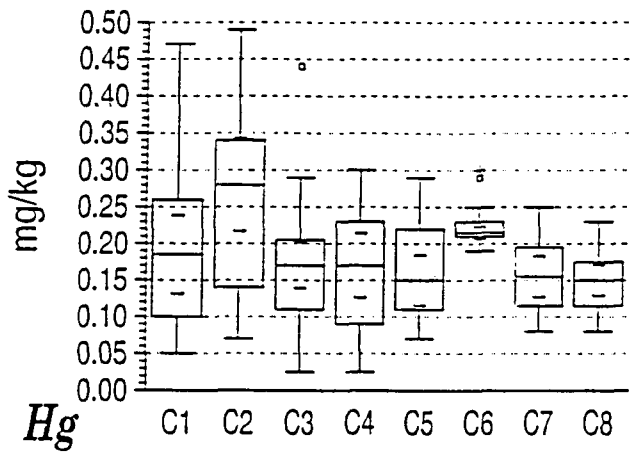
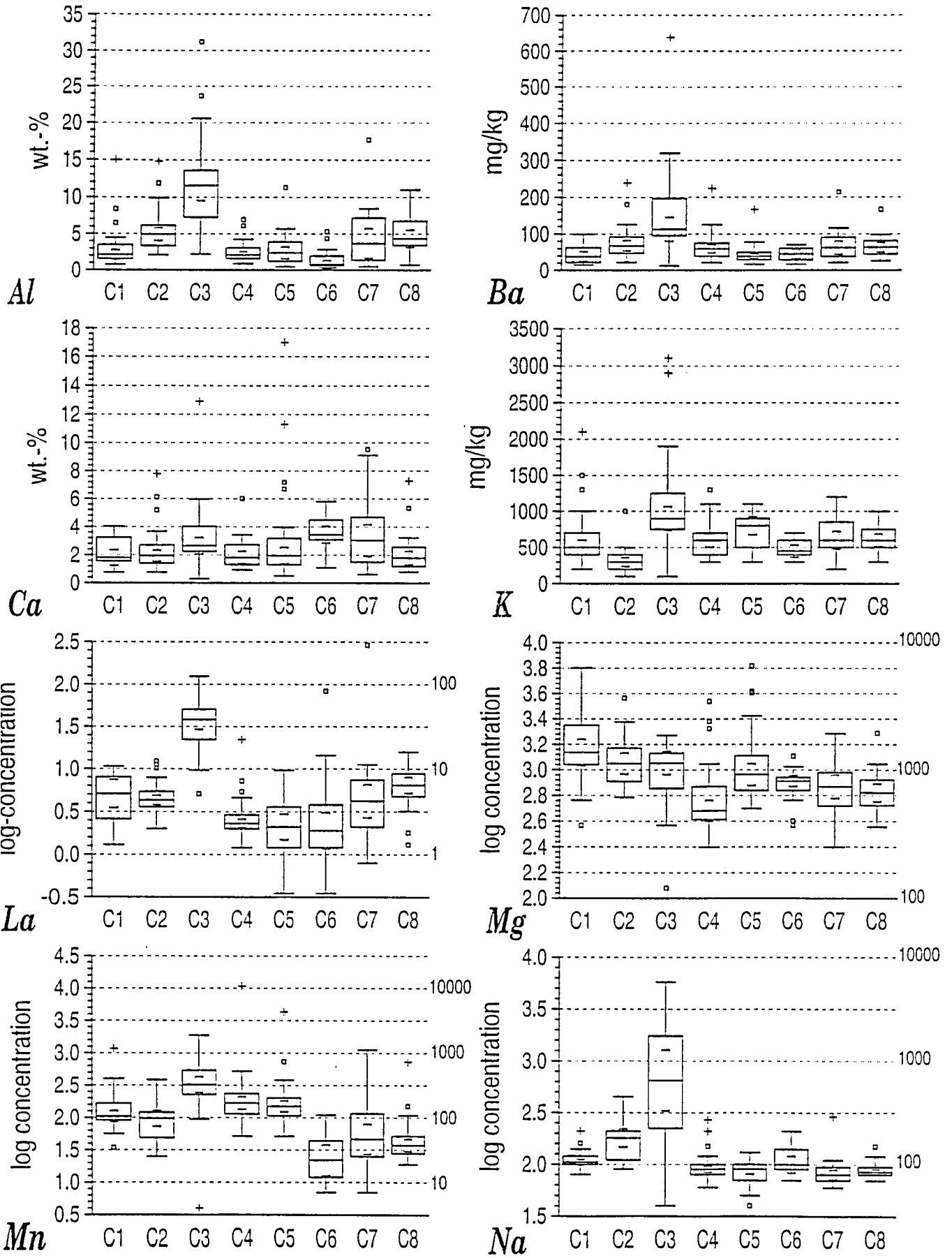
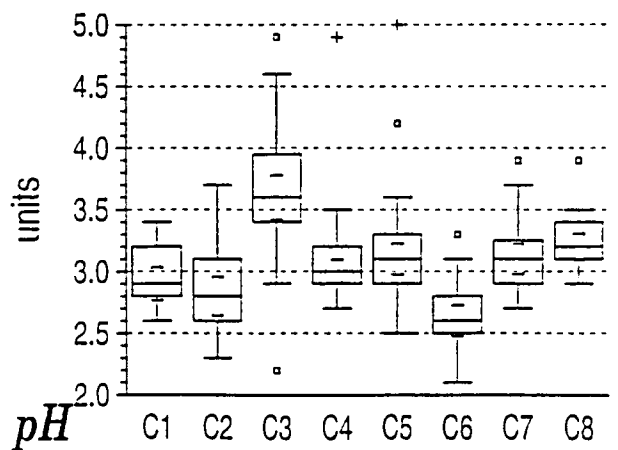
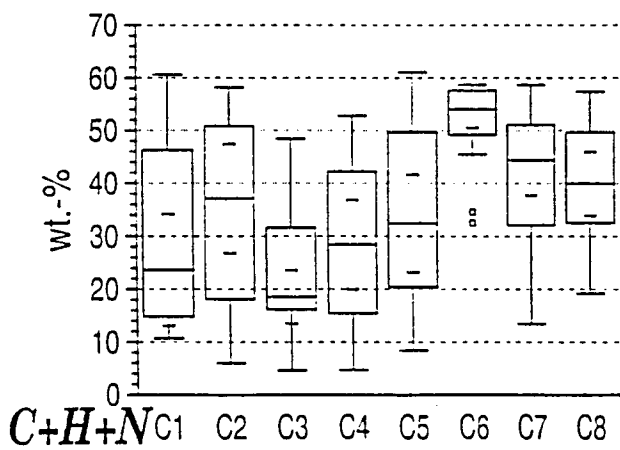
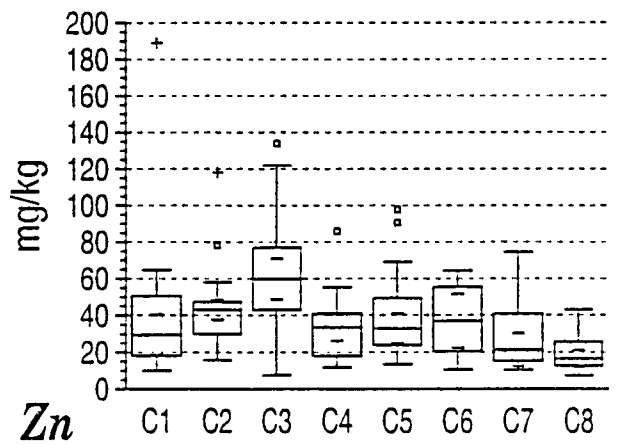
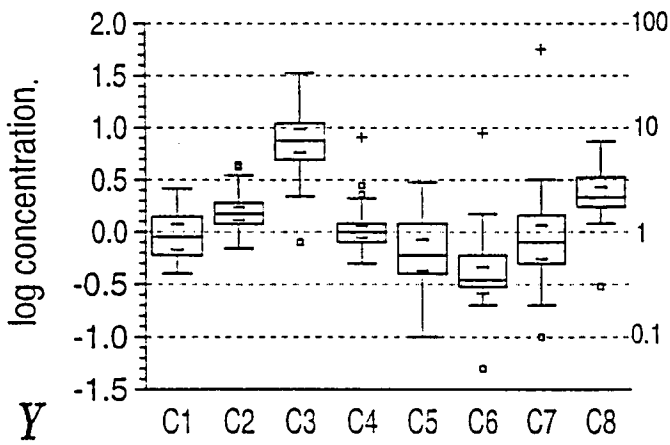
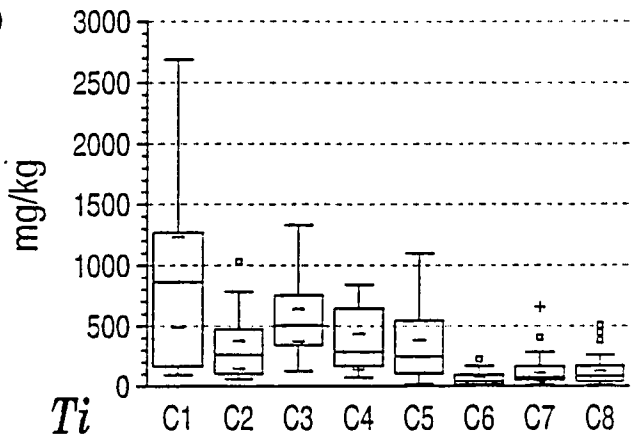
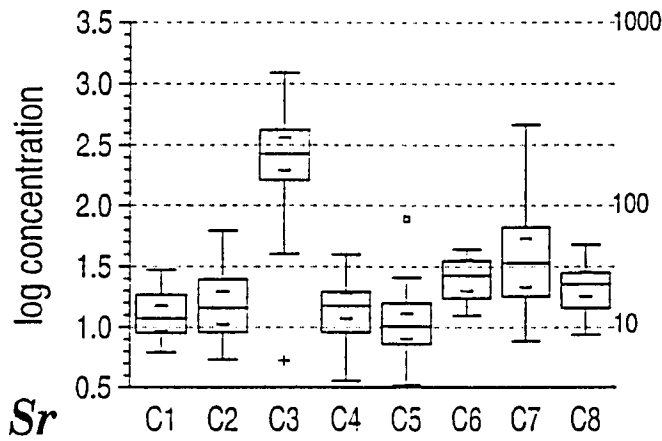
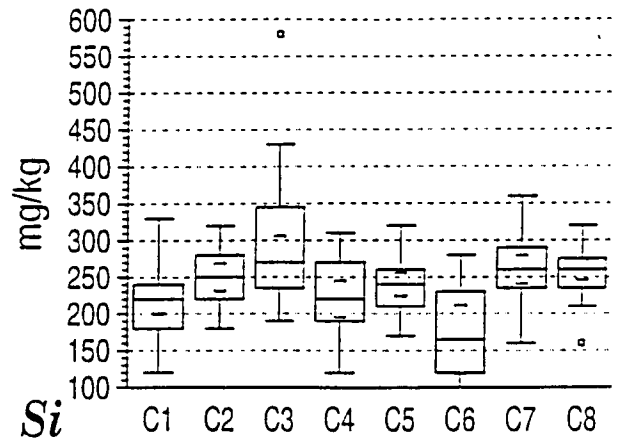
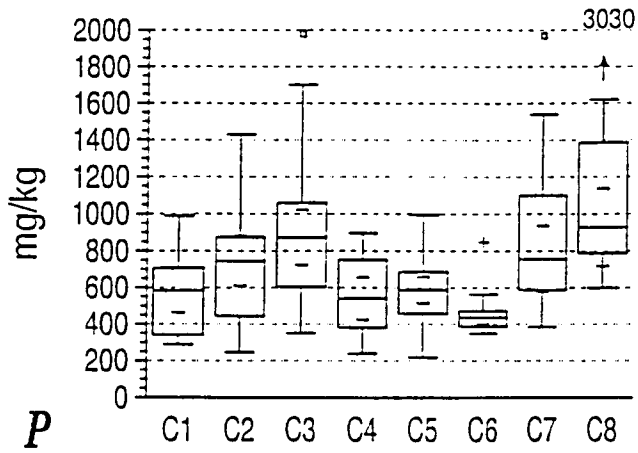


Fig. 7.1.3: Boxplot comparison of 14 elements showing lithology as the dominant influence on the observed levels, and also the C+H+N content and pH in the eight catchments.





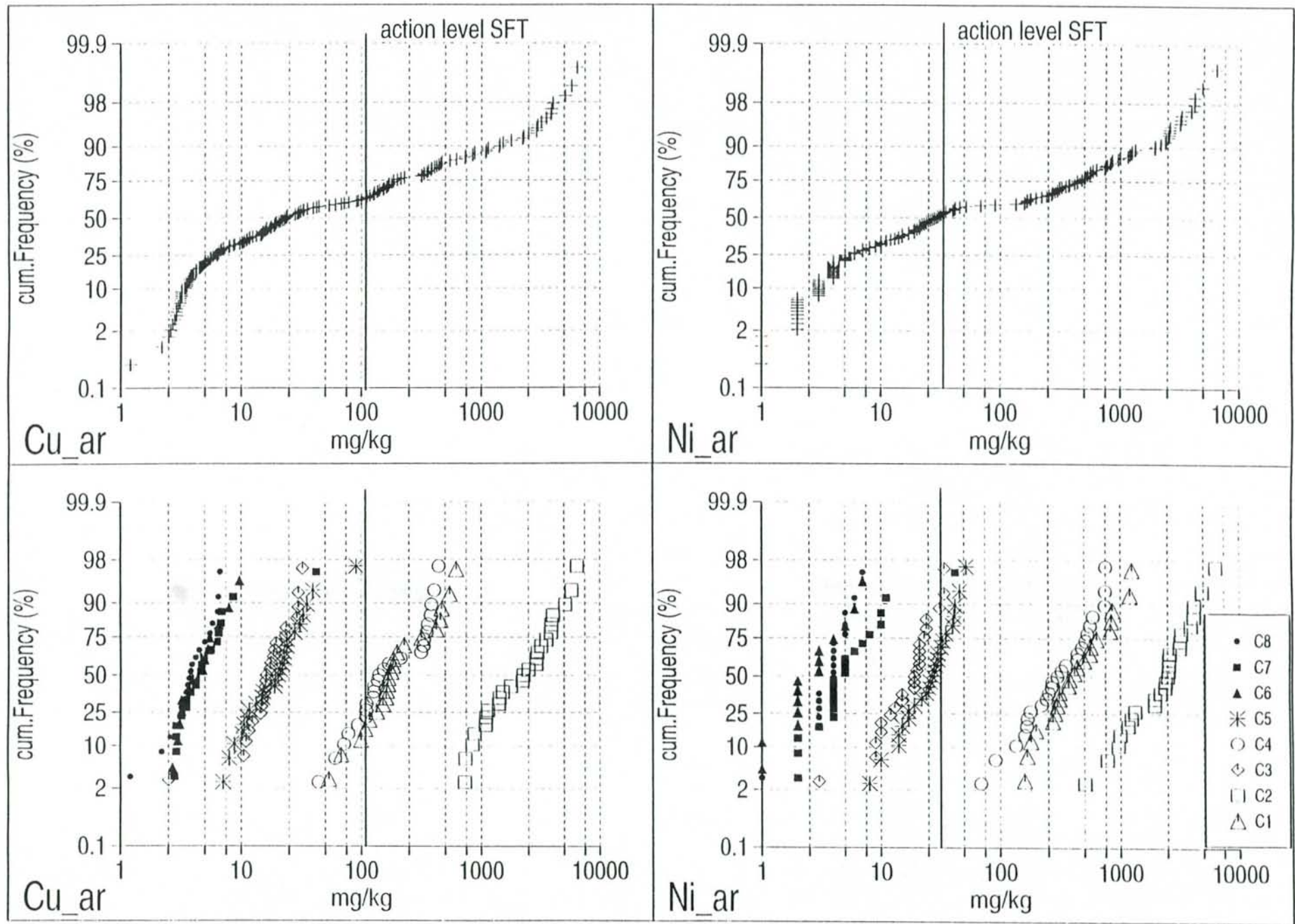
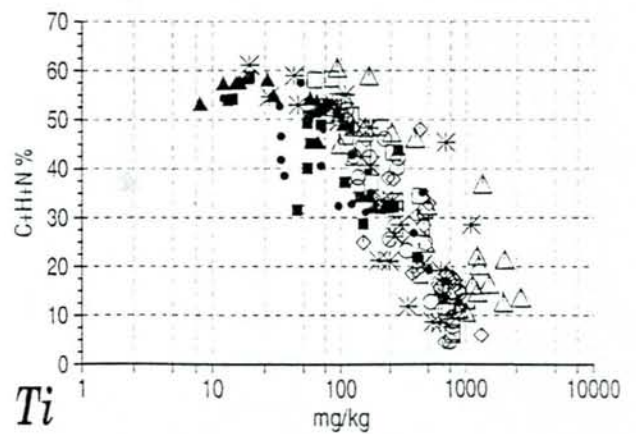
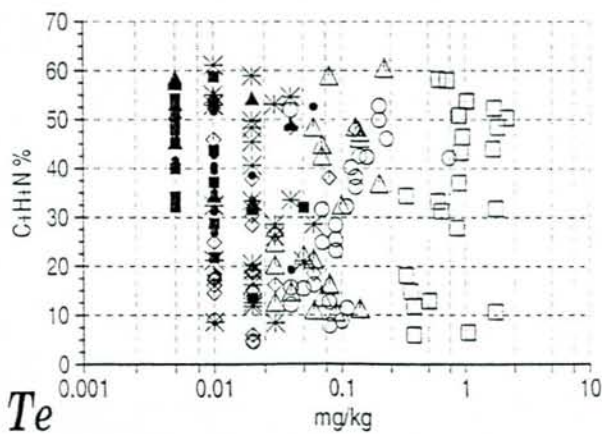
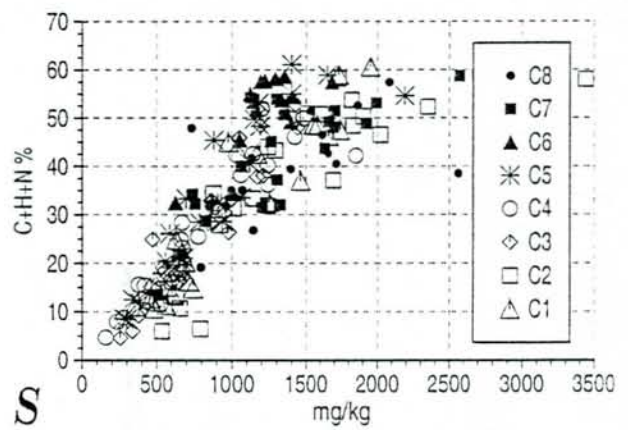
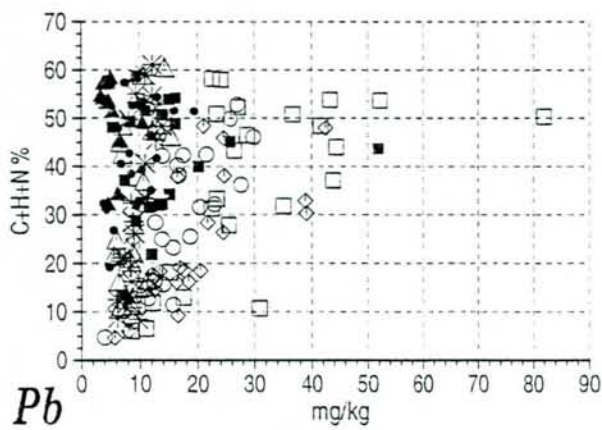
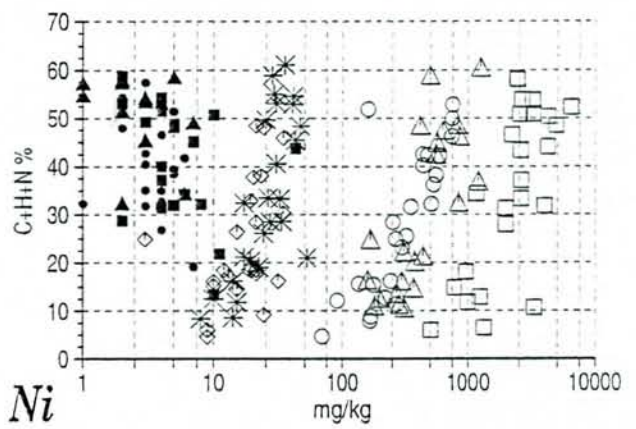
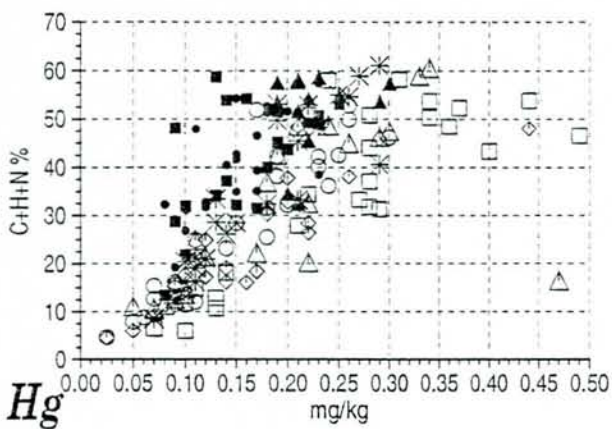
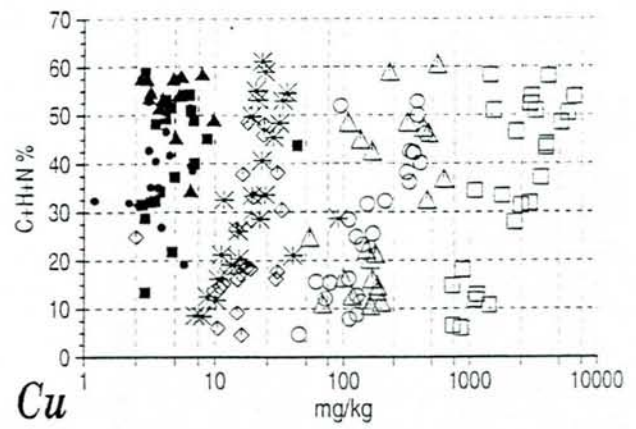
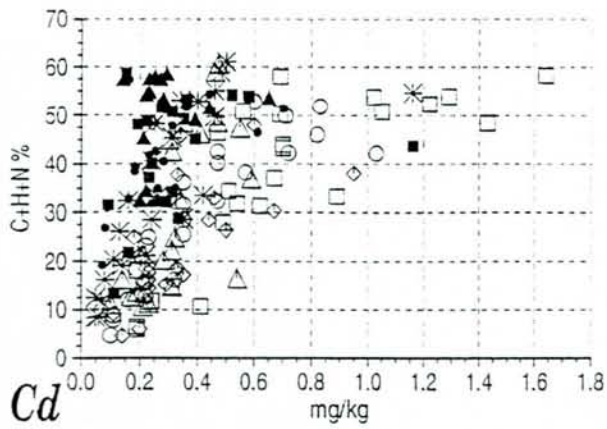


Fig. 7.1.4: CDF diagrams for Cu and Ni, showing the whole data set (N=174) in the upper diagram and the distribution in the 8 catchments (N see Table 7.1.2) in the lower diagram.



Fig. 7.1.5: XY diagrams showing the correlation between the organic content (C+H+N) and eight selected elements. Note the logarithmic scale used for Cu, Ni, Te and Ti.



## **7.2 Anthropogenic noble-metal enrichment of topsoil in the Monchegorsk area, Kola Peninsula, northwest Russia**

*Rognvald Boyd, Heikki Niskavaara, Esko Kontas, Victor Chekushin, Vladimir Pavlov, Morten Ofter and Clemens Reimann*

### **ABSTRACT**

Eight catchments, from 15-35 km<sup>2</sup> in area, have been studied within an ecogeochemical mapping programme in the western Kola Peninsula and contiguous parts of Finland and Norway. Three catchments, one NE of Zapolyarniy (1) and two, 5 and 25 km S of Monchegorsk (2 and 4) show high levels of deposition of heavy metals, especially Ni and Cu, and S, related to the metallurgical industry in these cities. 25 topsoil samples, from sites evenly distributed over catchment 2, have mean contents of Ni and Cu 1-2 orders of magnitude higher than the C-horizon samples, providing strong evidence for the anthropogenic origin of the heavy metals. The same samples show geometric mean total contents for the noble metals analysed of: 1.4 ppb Rh, 49.6 ppb Pt, 187.6 ppb Pd and 9.5 ppb Au. The pattern of concentration of the noble metals mirrors that found in published averages for ore from the Talnakh mineralizations in the Noril'sk province, though 1-2 orders of magnitude lower. This also clearly shows that the noble metal contents of the topsoil are anthropogenic, and suggests that they emanate from the plants in Monchegorsk at an early stage in treatment of the ore, probably as a minor component of Ni-Cu rich particles. The ratio Ni:Cu and PGE geochemistry reflect variations in the type of ore being processed at the plants close to the catchments.

### **INTRODUCTION**

#### *Project background*

Eight catchments, from 15-35 km<sup>2</sup> in area, have been studied as part of an ecogeochemical mapping programme in an area extending from 24°E to 35°30'E and S to the Arctic Circle in Finland and the S border of Murmansk region in Russia (Fig. 7.2.1)(see World Wide Web site: <http://www.ngu.no/Kola>). The main aims of the project have been to study the distribution of heavy metals at regional and local scales and to distinguish anthropogenic from natural concentrations. The main sources of heavy-metal pollution within the area are the nickel-copper processing plants at Nikel, Zapolyarniy and Monchegorsk. High levels of heavy-metal pollution in the area around Nikel and Zapolyarniy have been apparent and have been studied for a number of years, i.a. under the auspices of the joint Russian-Norwegian Commission on the Environment (Kismul et al., 1992). The project a part of which is reported here, involved ecogeochemical mapping over a much more extensive area than that covered by the commission and is the first to document the noble-metal content of soils in this area.

This paper presents data on the noble-metal content of topsoil samples from three of the catchments, those most heavily polluted, and will consider the geochemistry of the noble- and base-metals in the topsoil in relation to that of the ores being processed in the nearby plants. A brief description of the geochemistry of the ores and of the metallurgical industry will be given as background information for the results and discussion of them.



### *Geochemistry and processing of the Ni-Cu ores*

The metallurgical industry in the above-mentioned cities processes both local ore, from deposits in the Pechenga Zone, and ore from deposits in the Noril'sk province in Western Siberia. Though mineralizations had been reported earlier, the first major deposits in the Pechenga Zone were discovered in the 1920s. Production commenced during World War II and has continued up to the present. Several open-pit and underground mines are in operation. Annual production is estimated to be 30-35,000 tons Ni metal (Strishkov, 1989, quoted in Melezhik et al., 1994b). The Pechenga Zone is an early Proterozoic rift zone containing a thick sequence of picritic and tholeiitic volcanic rocks, minor intrusive bodies and intervening sedimentary units (Melezhik et al., 1994a). The ores, hosted in gabbro-wehrlite intrusions, include disseminated, breccia and massive ores (Melezhik et al., 1994b). Production is dominated by disseminated ore. The ores have a Ni/(Ni+Cu) ratio in excess of 0.5, most typically 0.6-0.7 (Melezhik et al., 1994b) and have relatively low total platinum metal (PGE) contents (of the order of 1 ppm in total sulphide) consisting of approximately equal amounts of platinum and palladium (and minor amounts of the other PGE) (Naldrett and Cabri, 1976)(Table 7.2.1).

Part of the production from deposits in the Noril'sk province in Western Siberia is transported by sea for processing on the Kola Peninsula. Deposits in the province have been mined since 1935 but transport of ore to the Kola Peninsula commenced in 1971-72. The province includes several groups of ore bodies in picritic-doleritic sills associated with the Triassic Siberian flood basalt province. Collectively the deposits form a Cu-Ni resource comparable to those of the Sudbury deposits in Canada (DeYoung et al., 1985) and are one of the two largest sources of platinum metals in the world (along with the Bushveld deposits in South Africa). The Noril'sk province contains a wide range of ore types, including large volumes of massive ore, also of various types. The massive ore typically has Ni/(Ni+Cu) ratios less than 0.5, in part less than 0.2: much of the ore is also very rich in platinum metals with grades reaching 200 ppm (Likhachev, 1992). Palladium is the dominant platinum metal (Smirnov, 1977, quoted in Naldrett, 1989) (Table 7.2.1). The type of Noril'sk ore processed at Nickel is thought to contain, on average, 2.35% Ni and 2.7% Cu in ore with 70% sulphides (Elkem Technology, 1993). The Noril'sk and Pechenga ores thus have compositions, both as regards major metals and PGE which are quite distinct, one from the other.

The major metallurgical plants in the Nickel-Zapolyarniy area, in addition to flotation plants, are a smelter in Nickel and a roasting plant in Zapolyarniy (Fig. 7.2.2). The smelter processes ore from Noril'sk, rich Pechenga ore, concentrate produced from local ore and pellets from the roasting plant (which processes local ore alone). Ni-Cu ore has been mined in the past in the Monchegorsk area but the metallurgical plants in the town now process pellets from Zapolyarniy, matte from Nickel and matte and ore from Noril'sk (Fig. 7.2.2). Nickel, copper and cobalt metal are refined, sulphuric acid is produced and a noble metal sludge is sent from Monchegorsk for further processing in Krasnoyarsk in western Siberia.

### *Catchment descriptions*

Catchment 1: Catchment 1 (Chekushin et al., 1995) lies 10 km NE of the Zapolyarniy plant: it is the drainage basin of a small western tributary of the R. Pechenga, with an area of 19 km<sup>2</sup> and an altitudinal range from 260 to 25 m.a.s.l. The vegetation type is Arctic forest tundra, visibly affected by pollution in the western part of the area. The bedrock within the catchment is dominated by gneisses and granites.

Catchment 2: Catchment 2 (Chekushin et al., 1995) lies 5 km S of the Monchegorsk plants: it is the upper part of the drainage basin of a river flowing into Lake Imandra. It has an area of 22.4 km<sup>2</sup> and an altitudinal range from 507 to 188 m.a.s.l. The vegetation type is northern taiga but most of the vegetation in the northern part of the catchment has died and much of the original soil cover has been removed by erosion. Mafic and ultramafic rocks form dominate the bedrock within the area: no mineralizations of significance are known within the catchment.

Catchment 4: Catchment 4 (Chekushin et al., 1995) lies 25 km SW of the Monchegorsk plants: it has an area of c. 20 km<sup>2</sup> and an altitudinal range from 460 to 150 m.a.s.l. The vegetation type is northern taiga but with clearly visible effects due to pollution (e.g. there is little moss remaining). mafic and ultramafic rocks form over half the bedrock of the catchment: no mineralizations are known within the area.

#### *Sampling procedure, preparation and analytical methods*

Sampling procedures are described in Äyräs and Reimann (1995), a description of the methods used for all media in the project. Sites were chosen to represent the type of superficial material and vegetation cover most representative of the catchment and were chosen to avoid steep slopes, active river and stream channels and beaches. Each sample is a composite of four subsamples taken at the corners of a 20 x 20 m square. The subsamples were 20 x 20 cm and min. 3 cm thick. Green vegetation, undecomposed needles and leaves and larger roots were removed from the samples. At least 1.5 kg was collected. All tools used (trowel and knives) were of stainless steel, free from paint. Samples were collected in late winter.

Samples were air-dried and homogenized by milling with a domestic blender with blades made of non-contaminating material and sieved to pass through a < 2 mm sieve. A 5g subsample was ashed in 500C and the ash digested with aqua regia. Gold, Pd, Pt and Rh were preconcentrated from the clear solutions with reductive coprecipitation using mercury as a collector. The elements were determined by graphite furnace-AAS (Kontas et al. 1990). Detection limit are for gold 0,1 ppb, for Pd and Rh 0,3ppb and for Pt 2 ppb. The noble metal results are compared with data for nickel and copper obtained by ICP-AES analysis after leaching in aqua regia (Reimann et al. submitted).

## RESULTS

Results obtained from catchment 2 are shown in Table 7.2.2. All the samples showed values above the detection limits for all four elements.

Twenty-four samples of topsoil from catchment 4 (23 km further south of Monchegorsk) showed values of Pd in the range 5 - 84 µg/Kg, and of Au in the range 0.2 - 5.4 µg/Kg. Geometric mean values were 22.1 and 1.8 µg/Kg Pd and Au respectively (Table 7.2.3). These samples were not analysed for Pt. Twenty-five samples of topsoil from catchment 1 (NE of Zapolyarniy) showed values of Pd up to 5 µg/Kg, with 11 samples below the detection limit of 2 µg/Kg. Values for Au were up to 5.3 µg/Kg, with one sample below the detection limit of 0.1 µg/Kg. Geometric means were 1.1 and 0.7 µg/Kg Pd and Au respectively (Table 7.2.3), assuming 1 µg/Kg for the samples falling below the detection limit for Pd.

Sixteen samples of Quaternary deposits, which can be regarded as equivalent to C-horizon soil, were sampled within catchment 2. With the exception of two samples with 13.7 and 25.8 ppb Pd respectively, all the values were in the low ppb area, with geometric means of 1.8 ppb Pt (taking six samples below the detection limit of 2 ppb as 1 ppb), 3.7 ppb Pd and 0.7 ppb Au.

Values found in 9 samples of Quaternary material from catchment 4 were comparable to the background levels in catchment 2, all values being in the low ppb range, with Pd generally exceeding Au and 6 samples falling below the detection limit for Pt. Geometric means for Pd and Au were 1,8 and 1 ppb respectively. Thirteen samples of Quaternary material from catchment 1 show low values for Pd and Au with geometric means of 1 and 1.1 ppb respectively. All except 2 samples had Pt values below the detection limit.

## DISCUSSION

Three clear factors suggest that the concentrations of the noble metals in the topsoil samples in catchment 2 have an anthropogenic origin:

- They are 1 - >2 orders of magnitude greater than the values found in (apparently) uncontaminated Quaternary material from the same catchment.
- A plot of the topsoil values on a logarithmic scale together with values from the Talnakh mineralisation in the Noril'sk province (Smirnov (1977) quoted in Naldrett (1989)) (Fig. 7.2.2) shows that the former have a distribution pattern which follows that of the Talnakh samples exceedingly closely though at a level 1-2 orders of magnitude lower.
- The values found in catchment 4 have noble-metal proportions generally similar to those in catchment 2, but at absolute levels which are reduced by a factor which could be anticipated in view of the generally lower level of heavy-metal contamination in the catchment in relation to catchment 2.

The ratios of Pd:Au in the Quaternary samples from these two catchments (Table 7.2.3) are compatible with a low level of anthropogenic contamination but the low concentrations and the lack of reliable data for Pt make such conclusions little more than speculation.

Catchment 1 had a level of deposition of Ni and Cu of 200-550 Kg/km<sup>2</sup> (the corresponding values for catchment 2 were 350-825 Kg/km<sup>2</sup>) in the winter half-year of 1993-94 (Chekushin et al. 1995 ++). This indicates a significant emission of base metals from the plant in Zapolyarniy. The low levels of concentration of Pd and Au in the topsoil samples from catchment 1 are compatible with the fact that the roasting plant in Zapolyarniy processes Pechenga ore with considerably lower contents of noble metals than Noril'sk ore. The data also indicate that any effect from the smelting of Noril'sk ore at Nikel further to the SW is negligible in catchment 1.

The noble- and base-metal values in Table 7.2.3 are, in one sense, not directly comparable, the former being based on total analyses and the latter on a partial solution by aqua regia. There is, however, good evidence (Ødegård 1995, Graff and Røste 1985) that those minerals not dissolved by aqua regia would not contain Ni in amounts which are significant in relation to the values in Table 7.2.3. The Ni :Pd ratio in the topsoil in catchment 2 is >10000:1: if we assume that the Noril'sk ore being processed at Monchegorsk has a noble metal geochemistry and Ni:Pd ratio similar to that published by Smirnov (1977) then the Ni:Pd ratio in the ore is of the order of 1000:1. This suggests that Ni (and probably Cu) are being emitted from certain of the plants at Monchegorsk without a significant component of noble metals. An alternative, though less probable explanation could be that the ore is significantly poorer in noble metals than assumed above.

The noble metal values found in catchment 2 are comparable to those found over known platinum metal mineralisations, e.g. in the Stillwater Complex in Montana (Conn, 1979), Lac Sheen and Lac Long - Lac Montauban in Quebec (Wood and Vlassopoulos, 1990) and Tulameen in British Columbia (Cook and Fletcher, 1994). Wood and Vlassopoulos (1990)

suggest that Pd is probably more mobile than Pt and Au in the surficial environment. There is no evidence for such mobility in the data from catchment 2.

The close similarity in the distribution pattern of noble metals in topsoil in catchment 2 and in typical Talnakh ore (Smirnov 1977) gives no indication that Pd is more easily leached than the other noble metals. The close correspondance between the geochemical pattern of the noble metals in the topsoil samples from catchment 2 and that of an assumed composition for Talnakh ore suggests that the noble metals are being deposited as trace minerals in particles of raw ore, i.e. at a very early stage in the processing of the ore, probably in the form of ore particles «blown out» from the smelter. Any effects from noble metal pollution from the other plants at Monchegorsk is completely overshadowed by this. The concentrations of noble metals, in addition to the base metals, being released from the plant in Monchegorsk (1,619 tons of Ni, 934 tons of Cu and 82 tons of Co in 1994 (Murmansk region CENR)), point to a potential for significant gains in metal value should it become possible to modernise the smelter in Monchegorsk.

#### ACKNOWLEDGEMENTS

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## TABLES

Table 7.2.1 Noble metal content of Pechenga and Talnakh ores. the former from Naldrett and Cabri and the latter from Smirnov

Table 7.2.2 Noble metal results from topsoil samples from catchment

Table 7.2.3 Geometric means of Pd, Au, Ni and Cu values from topsoil and Quaternary samples from the tree catchments.

## FIGURES

Fig. 7.2.1. Map of the project area showing the location of all the catchments, including numbers 1, 2 and 4 from which topsoil samples have been analysed for noble metals.

Fig. 7.2.2. Logarithmic plot of the minimum, geometric mean and maximum values for the twenty-five samples of topsoil from catchment 2 analysed for noble metals and the average of six samples of Talnakh ore (Smirnov 1977)

Table 1

Noble metal content of Pechenga and Talnakh (Noril'sk) ores (microgram/Kg). the former from Naldrett and Cabri (1976) and the latter from Smirnov (1977)

	Rh	Pt	Pd	Au
<b>PECHENGA AVE.*</b>	n.a.	600	500	200
<b>TALNAKH AVE.**</b>	2240	13700	36000	1600

Table 2

Noble metal results from topsoil samples from catchment 2 (microgram/Kg).

	Rh	Pt	Pd	Au
24301TS01	0,5	21,4	109	4,4
24302TS02	1,4	47,3	165	11,9
24303TS03	1,1	37,5	160	6,8
24304TS04	5	75,1	349	33,8
24305TS05	1,5	23,2	79	4,4
24306TS06	1	38,6	130	7,9
24307TS07	0,4	4,3	39	1,3
24308TS08	1,4	49,9	154	7,9
24309TS09	0,7	32	110	6,3
24310TS10	2,6	71,2	265	12,1
24311TS11	1,2	28,6	84	5
24312TS12	1,3	42,6	170	4,7
24313TS13	3,7	167	682	26,1
24314TS14	13,2	466	1760	99,5
24315TS15	1,4	68,1	216	11
24316TS16	1,4	56,9	187	14,8
24317TS17	1,4	37,3	113	6,9
24318TS18	1	51,3	221	11,1
24319TS19	2,4	100	295	19,6
24320TS20	1	43,2	153	7,7
24321TS21	0,9	28,5	102	4,7
24322TS22	3,1	53,4	407	7,8
24323TS23	0,9	25,2	76	4,2
24324TS24	0,5	120	370	20,5
24325TS25	1,7	137	413	21,6
<b>GEOM. MEAN</b>	1,4	49,6	187,6	9,5
<b>MAX.</b>	13,2	466	1760	99,5
<b>MIN.</b>	0,4	4,3	39	1,3

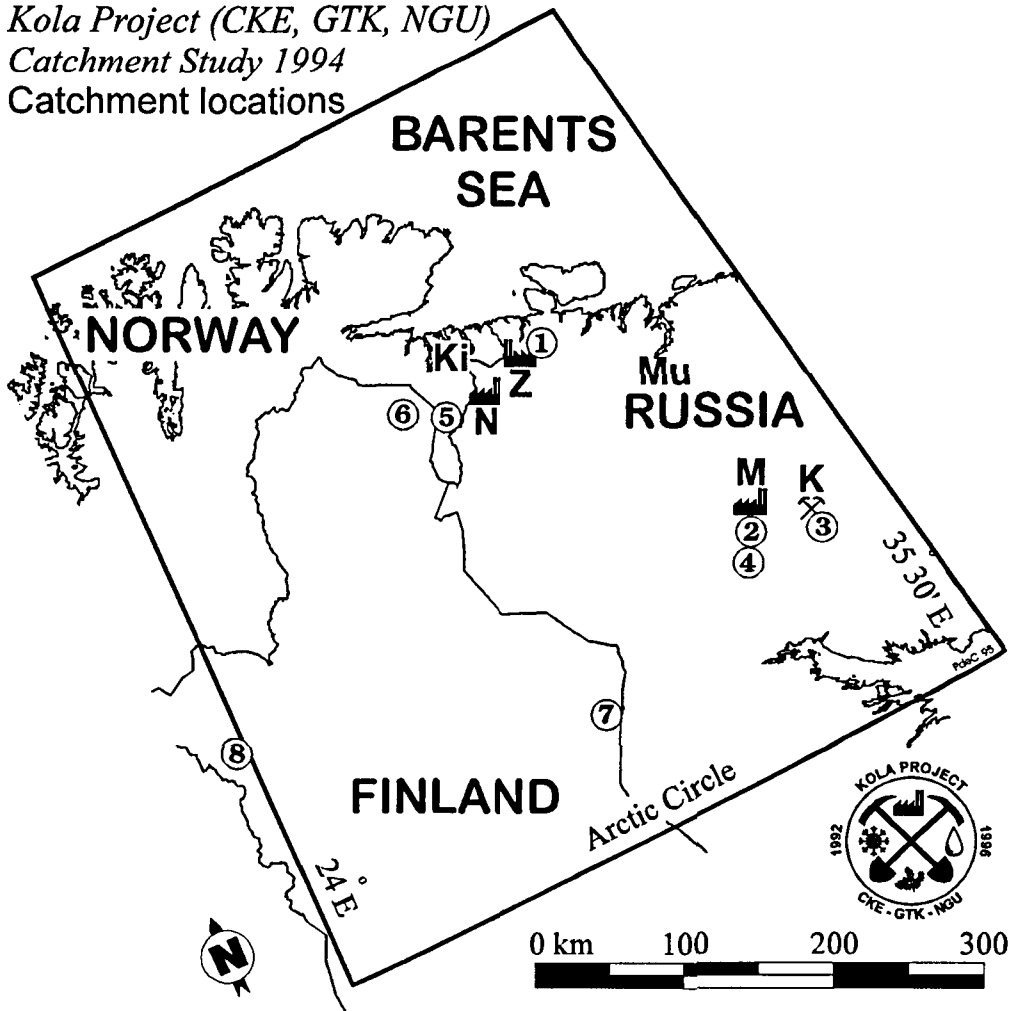
Table 3

Geometric means of Pd, Au, Ni and Cu values from topsoil and Quaternary samples from the three catchments (mg/Kg). The numbers in brackets are the numbers of samples of each type. Data for Ni and Cu are from Reimann et al. submitted).

	TOPSOIL				QUATERNARY			
	Pd	Au	Ni	Cu	Pd	Au	Ni	Cu
Catchment 1 (25/13)	0,0011	0,0007	417	193	0,001	0,0011	31	60
Catchment 2 (25/16)	0,1876	0,0095	2176	2108	0,0037	0,0007	104	72
Catchment 4 (25/7)	0,0221	0,0018	298	169	0,0018	0,001	45	71



*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
Catchment locations



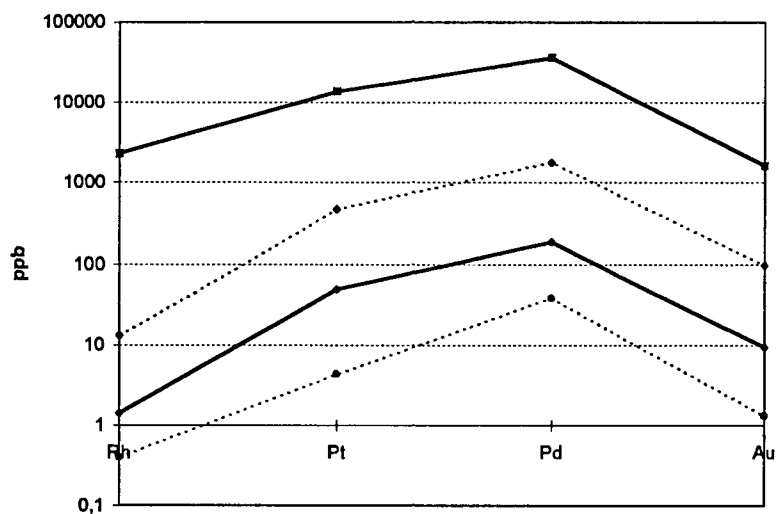


Fig. 7.2.2. Logarithmic plot of the minimum, geometric mean and maximum values for the twenty-five samples of topsoil from catchment 2 analysed for noble metals and the average of six samples of Talnakh ore (Smirnov 1977)

### 7.3. Mapping of Radioactivity in Topsoil (0-5 cm) and Reindeer Lichens in Parts of the Barents Region

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#### INTRODUCTION

The Geological Surveys of Finland (GTK) and Norway (NGU) and the Central Kola Expedition (CKE) are carrying out a major collaborative regional geochemical mapping project in a 188,000 km<sup>2</sup> area north of the Arctic Circle in these three countries (Fig. 7.3.1). The aims are (1) to establish regional geochemical maps of the distribution of heavy metals and radionuclides within the study area, (2) to build up a soil sample bank for use in future studies and (3) to assess the environmental impact of the heavy metal industry and nuclear activity in this ecologically vulnerable area.

During the summer of 1995, a variety of media, including moss, reindeer lichen, organic topsoil (humus of maximum 3 cm thickness) and topsoil 0-5 cm were collected. These sample media were selected as they are good indicators of the atmospheric deposition of elements in the survey area. Complete podzol profiles were also sampled at all locations. The lower part of soil pits (C-horizon) reflect the geological background and the B- and C-horizons will be analysed for the same 30 elements as the moss and organic topsoil samples. A total of ~ 640 localities were sampled, giving a mean density of 1 sample per 300 km<sup>2</sup>. Due to heavy grazing by reindeer in the Norwegian and Finnish parts of the study area and adverse climatic conditions, reindeer lichen samples could not be taken at all the visited localities.

#### RESULTS AND DISCUSSION

##### *Pilot project 1992*

As a preparation for this mapping project, a pilot study was carried out in the area of the nickel mining, roasting and smelting activity near Nikel and Zapoljarniy, close to the Norwegian and Finnish borders (Fig. 7.3.1). A variety of sample media were collected including snow, stream water, terrestrial moss, A<sub>0</sub>-horizon-soil, C-horizon-soils, stream sediment and overbank sediment. The samples were collected from 15 sampling stations within each of the three countries.

The <sup>134</sup>Cs and <sup>137</sup>Cs content of the uppermost 5 cm of the topsoil was determined during this pilot project. The results showed a high local variability. Representative sampling of topsoils in

the regional project thus require rather large composite samples. The maximum observed level for  $^{137}\text{Cs}$  was  $2.6 \text{ kBq/m}^2$  and there were no indications of pollution by contemporary sources.

#### *Catchment studies 1994*

A detailed study of eight small catchments situated at different distances from major industrial centres within the project area was carried out as part of the main project during 1994 (Fig. 7.3.1). The objective of this study was to better understand the processes occurring within catchment areas and to identify inter-relationships between different media. A total of four catchments were studied in Russia, three in Finland and one in Norway. During 1994, the chemical composition of the following media was studied: precipitation (snow and rainwater), vegetation (terrestrial moss, ground lichen and crowberry), soil (humus, «topsoil 0-5 cm» and horizons in podzol profiles), stream water, organic stream sediment, Quaternary deposits, bedrock and in some catchments, groundwater.

#### *Radiocesium in ground lichen*

For each medium more than 30 elements were analysed. For three of the Russian catchments (catchments 1, 3 and 4),  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations were determined in ground lichen (*Cladonia* sp. and *Cetraria* sp.) using HPGe analysis at the Finnish Centre for Radiation and Nuclear Safety (STUK). These results are presented as boxplot comparison in Fig. 7.3.2. Lichens are long-living plants that obtain their nutrients mainly directly from the air and are therefore suitable as bio-indicators of atmospheric fallout. Reindeer lichen is also well known as the main source of uptake of radioisotopes such as  $^{137}\text{Cs}$  in reindeer (Rissanen and Rahola, 1990).

The observed  $^{137}\text{Cs}$  concentrations calculated to May 1995 vary from 30 to 180 Bq/kg dry weight and are probably a result of both the atmospheric nuclear bomb tests in the 50's and 60's, and of the more recent fallout from the Chernobyl accident in 1986. Only one high  $^{137}\text{Cs}$  concentration (550 Bq/kg) was observed in a *Cladonia sulphurina* lichen sample (a species not consumed by reindeer) sampled from catchment 4, situated 25 km SW from Monchegorsk.

The  $^{134}\text{Cs}$  concentrations levels for May 1995 vary from less than detection limit to 10 Bq/kg. These concentrations are very low due to the short half-life of  $^{134}\text{Cs}$  (2 years). The Chernobyl accident appears to have had a relatively modest effect on the Kola Peninsula, and these results presented here are lower than the radiocesium content in reindeer lichens from both Finnish Lapland and Norway (National Institute of Radiation Hygiene, 1986). We did not observe increased radiocesium levels related to the underground use of civil nuclear explosives in the shutdown mines surrounding catchment 3, situated 3 km SE from Kirovsk.

#### *Natural occurring thorium and uranium*

The total content of Th and U was analysed for several of the collected media by ICP-MS and ICP-AES (Fig. 7.3.3). The majority of the sampled media reflecting atmospheric fallout (moss, lichen, snow, rainwater and partly topsoil) display concentrations below the detection limits for both Th (d. e.  $< 0.2 - 5 \text{ mg/kg}$ ) and U (d. e.  $< 0.2 \text{ mg/kg}$ ), with the exception of Th in catchment 3 near Kirovsk. As a result of the geology in catchments 6 and 8 in Finland, the U contents seen in organic stream sediments are extremely high (maximum of 510 mg/kg). These findings are partly in agreement with the contents in stream moss, stream organic matter and till reported in these areas from the geochemical atlas of Northern Fennoscandia (Bølviken *et al.* 1986).

## REGIONAL MAPPING (1995)

Topsoil and reindeer lichen sampled during summer 1995 will be analysed for a variety of radioactive isotopes by the Norwegian Radiation Protection Authority. The resulting data will be used to produce regional maps of radioactivity for the study area. The topsoil samples (consisting of the uppermost 5 cm of the soil after the removal of any living surface vegetation) were collected using a stainless steel auger designed for this purpose. The fixed depth of the topsoil samples resulted in the collection of a varying proportion of mineral soil in the samples. Because the pilot project had highlighted the problem of very high local variability, a minimum of ten subsamples were collected at each locality using the auger. A detailed description of the field methods used is given in Äyräs & Reimann (1995).

The composite samples are dried and homogenised in the laboratory. Approximately 100 g. of each sample is then placed in a 0.3 litre plastic container and sent for high purity germanium semi-conductor (HPGe) analysis for a variety of radioisotopes to the Norwegian Radiation Protection Authority. Both naturally occurring isotopes (e.g. isotopes from the Th- and U-decay series) and anthropogenic isotopes including  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  are determined by this gamma-spectrometric determination. Similar analyses are carried out for the lichen samples and a comparative study of the radioactivity in topsoil and reindeer lichen will be undertaken. At selected 'hotspots', radioisotope concentrations will be determined throughout complete soil profiles to better unravel the sources and the pollution history.

Due to financial restrictions, topsoil samples and reindeer lichen presently will be only analysed for radioactive isotopes. For the other sampled media, the Geological Survey of Finland will carry out analysis for more than 30 elements, including Th and U, by ICP-MS, ICP-AES and AAS techniques. The first regional maps will be ready in the course of 1996.

## CONCLUSIONS

A high variation of the natural occurrence of Th and U due to differing bedrock lithologies was observed in the catchment study. Surface concentrations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were generally low at our selected sites in the project area, but highly variable on the small scale. Thus the sample size and number of subsamples for topsoil from each site had to be increased for the production of maps of radionuclides in topsoil. Regional maps will be ready in 1996 as part of a joint ecogeochemical mapping project covering 188,000 km<sup>2</sup> in the northern part of Norway, Finland and Russia. Both natural occurring and anthropogenically spread radio-isotopes will be documented on an average sample density of 1 sample per 300 km<sup>2</sup> of the most populated and industrialised part of the Barents region.

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## FIGURES

Figure 7.3.1 Location of the studied catchments

Figure 7.3.2. Boxplot diagram showing the variation of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in Bq/kg (HPGe-determination) and As, Cr, Ni and Sr in mg/kg (ICP-MS analysis) in ground lichen from three of the Russian catchments.  $n = 12 - 14$  per catchment.

Figure 7.3.3. Boxplot diagram showing the variation of Th and U in mg/kg and  $\mu\text{g/l}$  (ppb) analysed by ICP-MS and ICP-AES in various media sampled from all eight investigated catchments. # indicates the number of samples in each catchment.

Kola Project (CKE, GTK, NGU)  
Catchment Study 1994  
Catchment locations

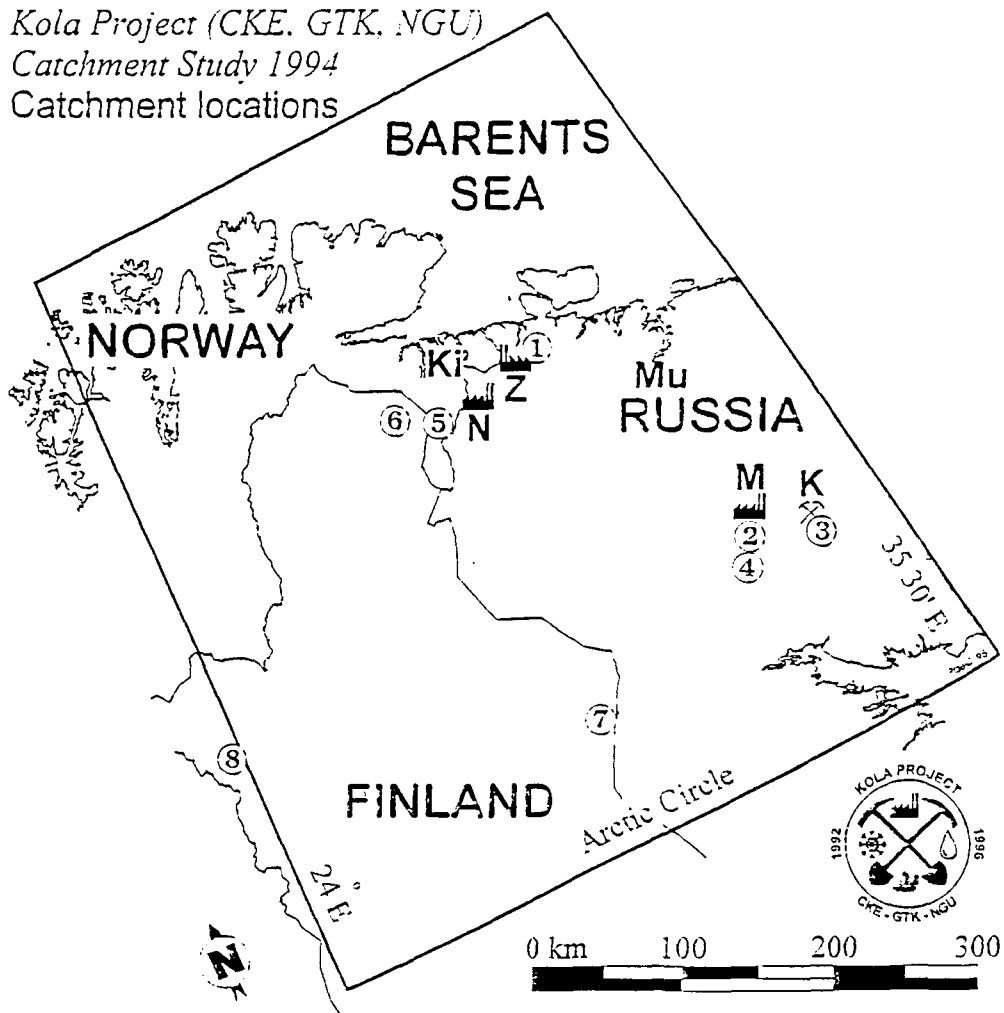
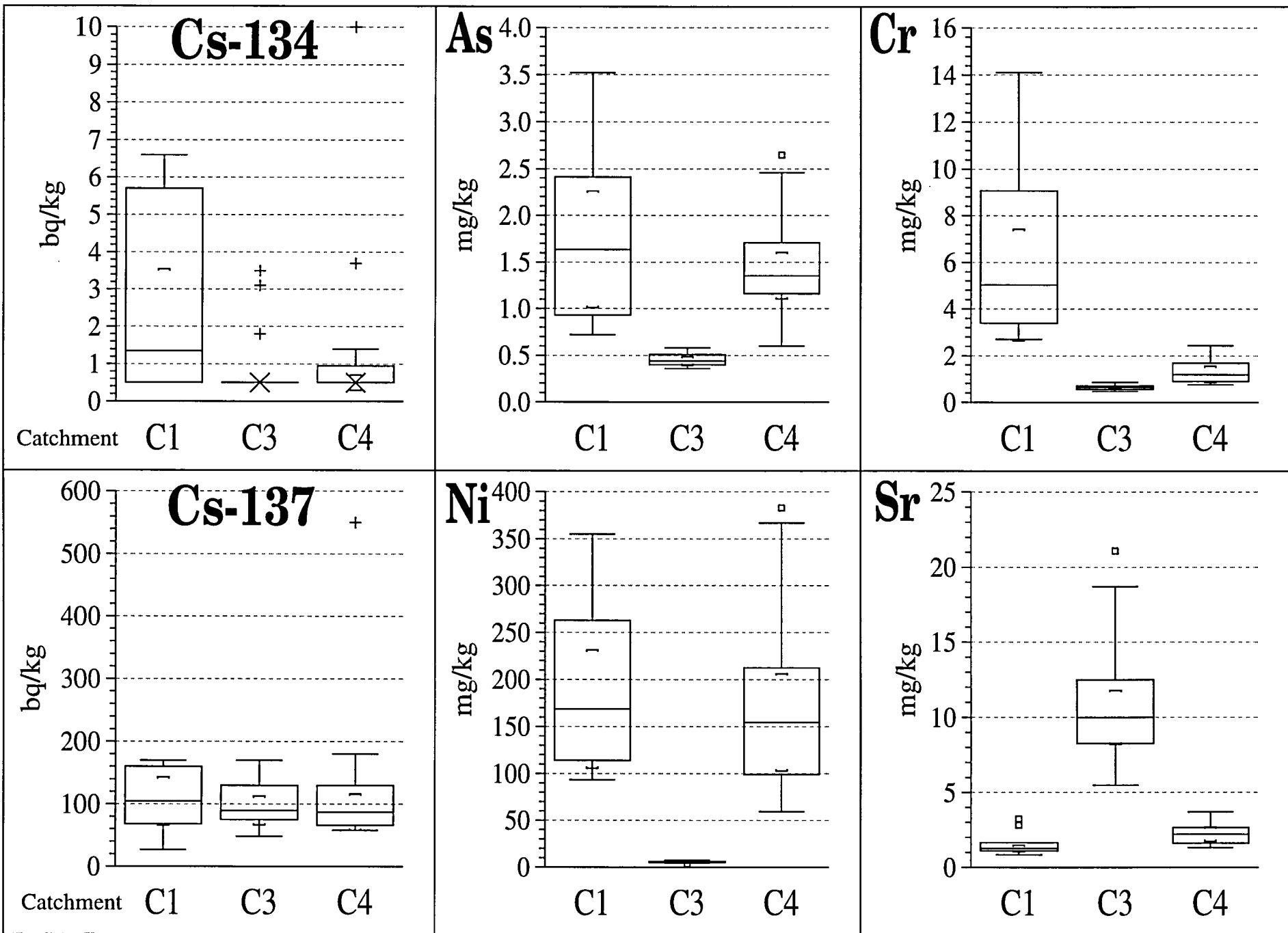


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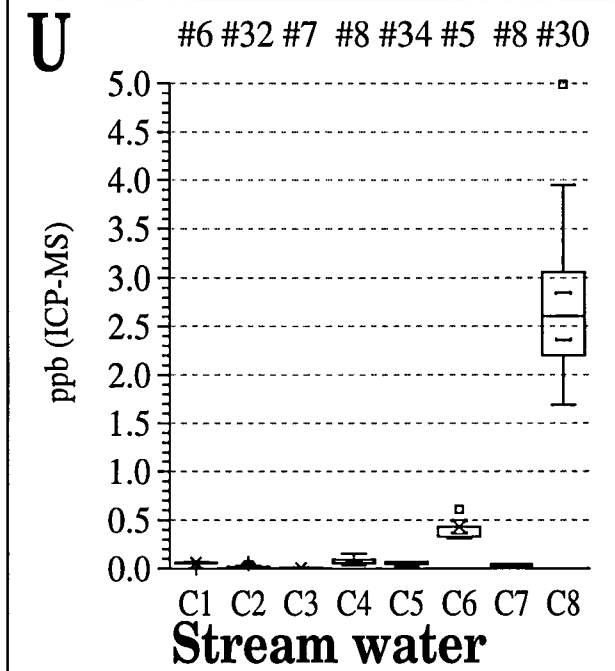
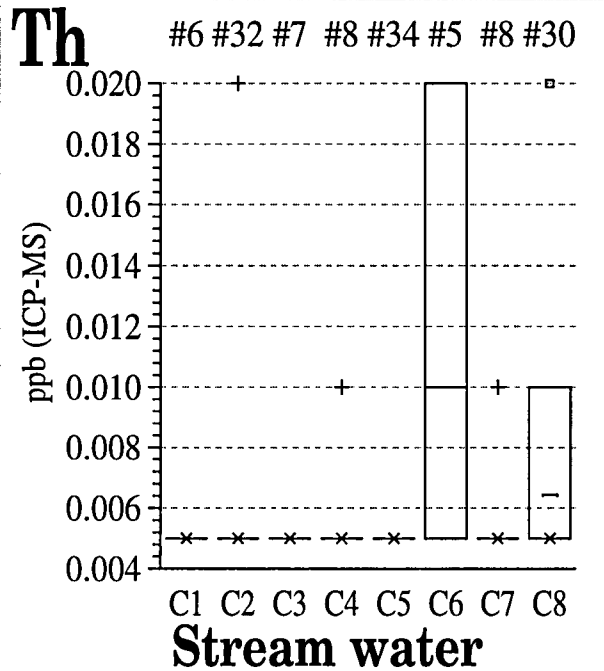
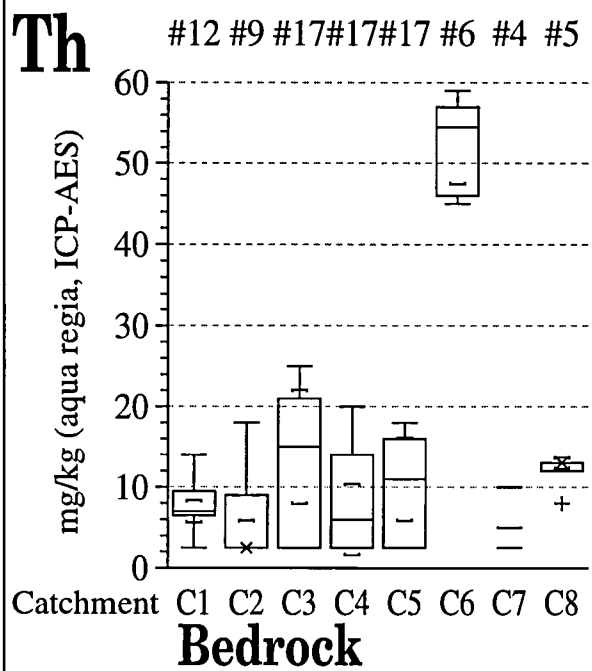
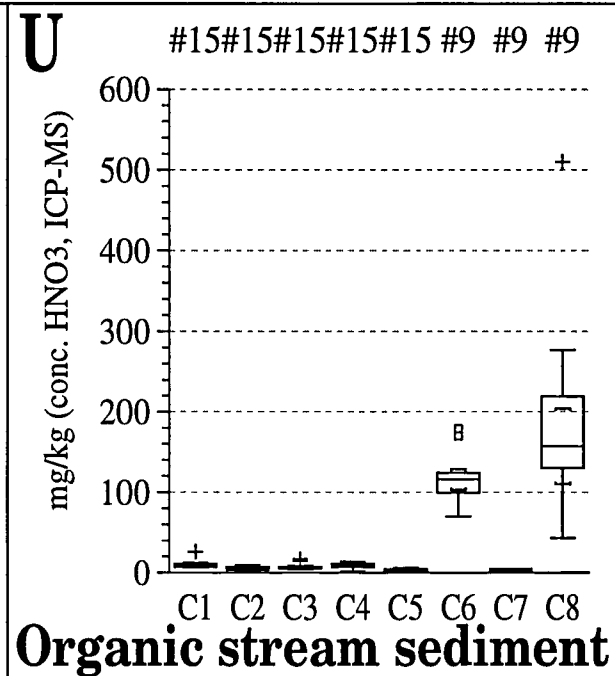
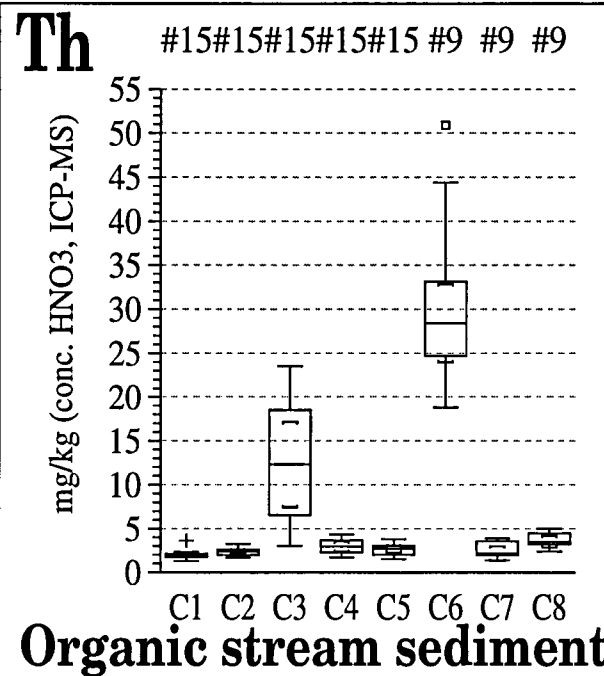
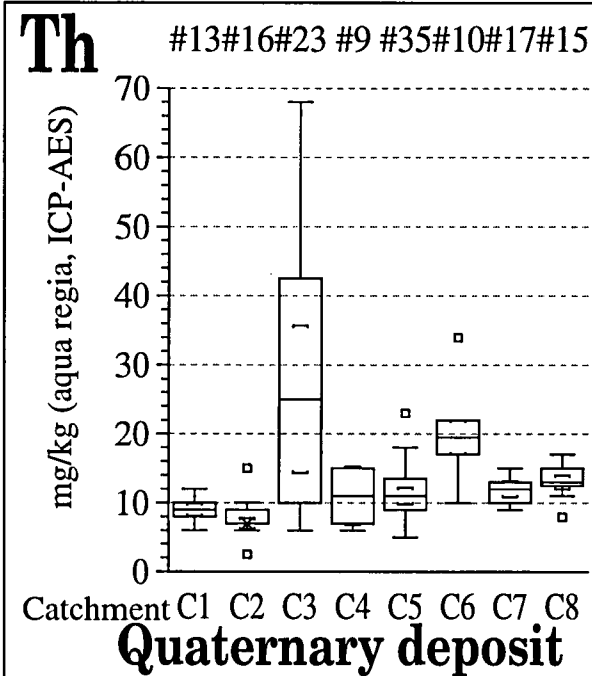


Figure 7.3.3. Boxplot diagram showing the variation of Th and U in mg/kg and µg/l (ppb) analysed by ICP-MS and ICP-AES in various media sampled from all eight investigated catchments. # indicates the number of samples in each catchment.

## 7.4 Seasonal variability of total and easily leachable element contents in topsoils (0-5 cm) from eight catchments in the European Arctic (Finland, Norway and Russia)

*Heikki Niskavaara, Clemens Reimann, Victor Chekushin and Galina Kashulina*

### ABSTRACT

Frozen topsoil samples (0-5 cm) were collected during March/April 1994 in eight Arctic catchments in northern Europe (4 in Russia, 3 in Finland, 1 in Norway), at different distances and wind directions from the emissions of the Russian nickel ore mining, roasting and smelting industry on the Kola Peninsula. Between 14 and 25 sites were sampled in catchment basins ranging in size from 12 to 35 km<sup>2</sup>. Sampling was repeated in spring right after snow melt, in summer and in the autumn to study seasonal variability and the fate of elements at the time of snow melt.

The < 2 mm fraction of air dried topsoils was analysed for total (aqua regia extraction) and easily leachable (in 1M ammonium acetate, buffered at pH 4.5) element contents using ICP-AES and GAAS for up to 35 elements. Results for selected elements are presented here.

Soil organic matter can be shown to be the controlling factor determining element contents and fate. In catchments close to the Russian nickel industry the topsoils have low carbon and nitrogen contents. Carbon and most elements reach maximum contents in winter time, lowest contents are observed in mid summer. Soil organic matter and elements associated with it are thus leached out of the soils during snow melt. During summer this process continues. Elements will be enriched in surface waters, in the lower layers of podzol profiles or reach the groundwater.

The use of the two leaches described provides a simple method to study the behaviour and pathways of elements in the topsoils during the arctic year. Using the proportions of easily leachable to total concentrations, a good estimation of the status of the topsoil in the study area can be given.

### INTRODUCTION

The Geological Surveys of Finland (GTK) and Norway (NGU) and the Central Kola Expedition (CKE) in Russia are carrying out a major environmental geochemical project (see World Wide Web site <http://www.ngu.no/Kola>) in a 188,000 km<sup>2</sup> area north of the Arctic Circle, comprising the entire area between 24° and 35.5° E north to the Barents Sea (Fig. 7.4.1). The Kola project consists of three phases: a pilot project in the border area of the three countries (see Niskavaara et al. 1996, Reimann et al. 1996a, in prep. a), a catchment study in 1994 (see Äyräs et al. 1995, in prep. a, Caritat et al. in prep., Reimann et al. 1996 b, in prep. b) and the regional mapping of the whole area in 1995 (see Äyräs et al., in prep. b, Reimann et al., in prep. c). In the catchment study eight catchments (hereafter abbreviated as C1 - C8) widely distributed in this area (Fig. 7.4.1) were investigated in detail in 1994. Media sampled were: snow (meltwater and filter residue), rainwater, stream water, organic stream sediments, terrestrial moss, topsoil (0-5 cm), complete podzol profiles, quaternary deposits and bedrock.

Some of the world's largest point sources of SO<sub>2</sub> emissions are located within the study area (Gunn et al., 1995), the nickel smelter at Nikel, the ore roasting plant at Zapoljarnij and the nickel smelter at Monchegorsk (Fig. 7.4.1) together accounting for about 300,000 t of SO<sub>2</sub>, 1900 t of Ni, 1100 t of Cu and 94 t of V<sub>2</sub>O<sub>5</sub> emissions yearly (Murmansk regional committee for ecology and natural resources - see Reimann et al., 1996b). During the last fifty years these emissions have resulted in an

extensive contamination of the soil and the whole terrestrial environment providing a man-made testing area to study the effects of metal and sulphur contamination to different compartments of terrestrial environment.

Soils act both as a major sink for pollutant elements and as principal source of metals entering the food chain. The topmost few cm of the soil profile is a particularly sensitive substrate in the terrestrial environment. It receives atmospheric deposition of heavy metals and sulphur and is prone to the eroding effects of rain, wind and frost. Depending on the vegetation type, climatic conditions, soil formation conditions, the extend of anthropogenic effects etc. the topsoil contains varying amounts of organic and inorganic material. Soil organic matter is one of the major factors controlling the physical and chemical properties of soils including metal binding capacity, buffering capacity and water holding capacity. Other major physicochemical factors effecting retention of metals are the amount of clay minerals and secondary hydroxy -and oxide precipitates. The metal binding properties may hence vary substantially depending on the soil composition and genesis. As a rough presentation of complexation stabilities of elements with natural organic substances the following sequence has been presented (Merian, 1991):

$Cr > Fe > Al > Pb >> Cu > Ni > Co >> Cd > Zn >> Mn = Ca = Mg$

Budgets of heavy metals in the upper forest soils are normally negative for Cd and Zn (losses of elements are bigger than the input to the soil) and positive for elements like Fe, Cu, Pb and Hg (Borg and Johansson, 1989). Acidic deposition, however, will increase the leaching of more strongly bound elements. In strongly polluted areas near industrial centres the physicochemical properties of topsoil are severely damaged. Effects are changes in the total budget of contaminants and nutrients and in the available nutrient contents. In arctic areas the topsoil is frozen for at least six month of the year. In spring the still partly frozen soil is affected by sudden, intensive flushing of acidic snow melt water percolating through the uppermost soil layers. In contaminated areas these melt waters contain even higher amounts of acidifying components in addition to large amounts of heavy metals accumulated with the whole winters deposition.

Aqua regia digestion (ISO standard 11466) has been widely accepted to give a good estimate of the maximum potential pool of soluble elements in soils in environmental sciences. The residual part of the elements which are not released by aqua regia digestion are mostly bound to silicate lattices and are considered unimportant in estimating the element mobilities and behaviour. The mobility of elements in soils depends, however, strongly on their specific chemical forms and the ways of binding. The biological and ecological effects of the elements as well as their behaviour and pathways in the terrestrial environment can be assumed to be more related to alterations in the soluble and mobile fractions than to changes in the total concentrations. Hence it is important to determine in addition to total concentrations which part of this total pool of elements is mobile under particular conditions. A variety of single weak leaches (Pickering, 1981) or sequential extraction schemes (e.g. Tessier et al. 1979, Chao, 1983, Ure et al 1993) have been widely used to estimate the easily leachable, mobile, exchangeable or plant available proportion of the element contents. Most of these leaches, however, will only give a very rough idea of the speciation of elements in soils and results strongly depend on the specific leach chosen, elements analysed and purely on technical details, such as extraction time, volume of the extractant, temperature etc. Thus the results are often very difficult to interpret and almost never comparable between different laboratories and may cause misleading interpretations.

Instead of speciation a new term, operationally defined “forms” of elements, has been created. When the purpose of the study is not to fully characterise the soil sample, but to determine soluble

fractions of elements in particular conditions a relatively simple extraction scheme may be sufficient. A single leach with 1M ammonium acetate buffered with acetic acid to various pH's (regularly 7.0 and 4.65), depending on the substrates (soil) pH, is a frequently used single extraction procedure. Using ammonium acetate the base cations in the soil samples are released from exchangeable sites and therefore only relatively little effected by pH. In contrast, Fe and Al are supposed to be released mainly from precipitated compounds such as hydrated oxides and hydroxides and are thus strongly affected by pH. Most of the heavy metals react also strongly to the pH of the solution used for extraction (Andersson, 1976). By buffering this solution to pH 4,5, which roughly is the pH of the snow melt water and the soils in the study area, an operationally defined fraction of elements as an estimate of the effects of snow melt water on soils, can be determined.

During the Pilot project a comparison of total and easily leachable element contents in humus (O-horizon) was carried out (Niskavaara et al. 1996, Reimann et al. in prep. a). The results revealed the importance of the determination of mobile fractions of the elements in these badly contaminated areas to assess their mobility and pathways. The influence of the snowmelting period on the whole terrestrial environment in the Arctic is another important question to be addressed. It was thus decided to study the fate of heavy metals in the uppermost soil layer in more detail during the catchment study.

Copper and nickel were observed to behave differently in humus in the surroundings of the Cu/Ni - smelter in Nikel. Copper was strongly bound to organic matter whereas nickel occurred in easily leachable forms explaining the high concentrations of Ni found in stream waters of the study area. Adamo et al (1996) have carried out a study in another severely contaminated Cu/Ni -smelter area: Sudbury, Canada. They studied the chemical and mineralogical forms of Cu and Ni using sequential extraction and scanning electron microscopy and energy dispersive X-ray analysis. They noticed the same distribution of Cu and Ni as Niskavaara et al. (1996) but their conclusions were opposite: Cu occurred in more mobile forms than Ni. This is in contradiction to widely accepted concepts (Alloway, 1990; Merian, 1991). However the distance from and the nature of the emission source (feed material, mining activity/smelter/roasting plant) will probably effect the occurrence of the elements in the emission spectrum ( wet/dry deposition, particle size, mineralogy) and differences in element behaviour in different surroundings will be the result. On the Kola Peninsula, Russia, all these variables can be studied in detail by a proper selection of the study areas. Then different "fingerprints" of the emissions can be defined by comparing element associations and mobilities even when the total concentrations do not differ from each other.

The aims of this study were (1) to establish an overview of the seasonal variation of heavy metal contents in topsoils (0-5 cm) collected in 8 catchments located in different settings (distance to industry and the ocean, geology, vegetation) throughout the main project area (2) to improve our understanding of element mobilities and fate at the time of snow melt in arctic terrain and (3) to compare total element contents with easily available element contents.

The results of topsoil (0-5 cm) sampling carried out during March/April, April/June, July and September of 1994 for selected elements are presented here. Total element levels for about 35 elements based on winter sampling and suitability of topsoil samples for regional geochemical mapping are discussed in Reimann et al. (1996c).

## MATERIAL AND METHODS

### *Sampling sites*

Samples were taken in eight catchments (numbered from C1 to C8) representing different distances to pollution sources and the sea, different geology and different vegetation types (Tab. 7.4.1). C1 is situated close to the nickel ore roasting plant at Zapoljarnij, C2 is adjacent to the nickel smelter in Monchegorsk, C3 is adjacent to the big open pit apatite quarry in Kirovsk with its very special bedrock lithology. C4 is about 30 km to the south of Monchegorsk. C5 is situated in Norway about 35 km to the SW (off wind) of the smelter in Nikel, C6, 7 and 8 are all in Finland and represent as pristine background areas as one can find in Europe. They are characterised by different geological environments.

Podzols are the prevailing soil type in these areas. The thickness of the uppermost organic layer (O-horizon) can vary between 1 and 10 cm. Thus the uppermost 5 cm of soil can contain different proportions of the O-horizon and the underlying mineral soil horizons (E-horizon or even B-horizon). To understand the following discussions it is important to notice, that all horizons of podzol profiles are characterised by having different chemical and physical properties, including binding and buffering of heavy metals and nutrients and water holding capacities. The proportions of the different horizons present in the samples will thus have a major influence on the observed chemical composition and these differences are responsible for most of the “noise” we see in the data discussed here. In C2, the ecosystem is severely damaged, the soils are already strongly eroded and the organic and even the eluvial layers lack in places.

### SAMPLING

The first collection of topsoil samples was carried out at the same time as snowpack sampling (Äyräs and Reimann, 1995). Up to 25 sample sites were distributed evenly throughout the studied catchment. At each sample site, 5 subsample sites were selected for a composite snowpack sample and holes were dug down to the ground using a plastic snow shovel. At the bottom a 20 x 20 cm large sample was cut out of the frozen ground using a steel spade. Vegetation and big rock fragments were removed from the sample and the uppermost five cm were then taken as the sample. The thickness of the organic layer was recorded. The final sample was a composite sample of the five subsamples per station. The samples were kept frozen at all times and delivered frozen to GTK's laboratory in Rovaniemi.

The second sampling took place as close after snowmelt as practically possible (access to the field areas). All winter sites were revisited and a new set of samples, following the sample procedures as during winter time, was collected from each sample site. In summer (July) and autumn (September) half of all sample sites, spread evenly over the catchment, were again revisited and samples were taken following the same procedure as before.

All field teams worked with exactly the same equipment. Polyethylene plastic bags were used for storage of the samples.

### *Sample preparation and analysis*

The samples were air dried at room temperature right after arrival at GTK's laboratory in Rovaniemi. For the winter samples the effects of air drying versus freeze drying were tested on a portion of the samples. No prominent differences were noticed in the concentrations of total or easily leachable elements. Observed small differences (<5%) could be explained by differences in

residual moisture. The dried samples were homogenised by milling with a domestic blender with blades made of uncontaminating material and sieved to pass through a <2 mm nylon screen.

A 2 g subsample was mixed with 12 ml aqua regia (1:3 HCl and HNO<sub>3</sub>), left standing over night then digested for 2 hours at 90 °C and diluted with water to 60 ml and centrifuged to obtain a clear solution (modified ISO standard 11466).

6 g of sample were shaken in a horizontal shaker for 2 hours with 30 ml of 1M ammonium acetate solution buffered at pH 4.5, centrifuged and filtered through a 0.45 µm membrane filter to obtain a clear solution and acidified with suprapure nitric acid.

Both solutions were analysed with an inductively coupled plasma atomic emission spectrometer (ICP-AES) for up to 31 elements. A graphite furnace atomic absorption spectrometer (GFAAS) was used to analyse for Ag, As and Cd by direct determination, for Bi, Sb, Se, and Te after preconcentration using reductive coprecipitation (Niskavaara and Kontas, 1991) and for mercury using cold vapour-AAS from the aqua regia digests of the winter and spring samples only. Carbon, hydrogen and nitrogen were analysed with a CHN analyser (Leco).

All analysis were carried out at GTK's laboratories. About 10% of all samples were analysed in duplicate. The reference sample BCR CRM277 was used as a quality control sample to assess the reproducibility of the total digestion analysis. For more details on analytical techniques and quality control procedures see Niskavaara (1995). The GTK laboratory is accredited according to EN 45001 and the ISO-Guide 25.

#### *Data analysis*

All graphics were produced using the DAS program (Dutter et al., 1992), based on exploratory data analysis (EDA) methods (Tukey, 1977, Velleman and Hoaglin, 1981). Kürzl (1988), Reimann et al. (1988), Rock (1988) and O'Connor and Reimann (1993) give an introduction to the advantages of using exploratory data analysis methods when dealing with geochemical data.

The boxplot as used here is an useful graphic originating from exploratory data analysis (EDA) (Tukey, 1977). It provides a graphical data summary, relying solely on the inherent data structure and not on any assumptions about the normality of the data distribution. It basically divides the distribution of the results into quartiles, firstly by finding the median (displayed as a line in the box), and then doing the same for each of the remaining halves. These upper and lower points or "hinges" define the central box which thus in itself again contains 50% of all data. The width of the box is called the hingespread (HS) and can be used as a robust equivalent to the standard deviation. "Whiskers" are then drawn from the ends of the box, each extending 1.5 times the width of the box towards the maximum and the minimum (taken back to the last real data point). Any values outside of these whiskers are defined as data outliers. The notches (square brackets), placed at  $1.58 \times (HS / \sqrt{n})$  on either site of the median, are a test of significance of medians from different populations presented in the boxes (for more information see Tukey, 1977 or Velleman and Hoaglin, 1981). Important information about the dataset, e.g. median, quartiles, skewness of the distribution, the existence of data outliers and the significance of observed differences can all be extracted at one glance from this simple graphic.

## RESULTS

The variation of element contents between sample sites within one catchment and between the seasons is large - figures 7.4.2-6 shows this for some selected elements in the contaminated catchments C1, C2, C4 and in the background catchment C8. The largest variations can be observed for the most polluted catchments and for the samples taken in the winter season. These large variations in element contents can be explained by topographical, vegetational etc. differences between the sampling sites within one catchment. Especially in the catchments close to the smelters the differences in distance to the emission sources and the different proportions of the O, E and B-horizon material in the depth related samples will additionally increase variations. The gradient of deposition for almost all elements follows exponential curves (Chekushin et al. in prep., Reimann et al., 1996d) and can thus be very steep adding a further source of large variations.

Strong positive correlations with the organic content (C+H+N) have been observed for total contents of many elements (S, Hg, Cd, P, B, Pb, Ca, Zn and Se) (Reimann et al. 1996c). A negative correlation with the organic content can be observed for Al, Cr, Li, Sc, Ti, Y and V, reflecting the minerogenic part of the topsoil. For the main contaminants Cu and Ni no correlation with organic content can be observed for the data from all catchments but strong correlations exist in single catchments (Reimann et al. 1996c).

For the easily leachable proportion of element contents a positive correlation with carbon is best seen for Cd, Mg, Ca, Na, K, Zn, P, S and Sr. Only Fe and Al showed marked negative correlations here.

It may thus have been justified to correct (normalise) all data against (C+H+N). However, to obtain information of the total element pool and the flow of material and elements within the catchments it was decided to use plain uncorrected data for this study. It must be mentioned that for regional mapping purposes and reliable comparisons between the catchments the interpretations could certainly be improved by normalising all element contents against carbon when using topsoil samples.

The concentration levels of many elements are exceptionally high in the most polluted catchments C1, C2 and C4. For example, the easily leachable concentrations of Cu and Ni in C2 are at the same level as total concentrations of these elements in the Sudbury area (Adamo et al 1996). This shows how severely polluted the study area is.

### *Carbon, nitrogen and hydrogen*

Large seasonal differences in C, H and N-contents can be observed in the Russian catchments C1, C2 and C4 (Fig. 7.4.2). High concentrations (e.g. median >20% C) in the winter samples drop considerably at the time of snow melting in spring and reach their minimum in mid-summer (<10% C). This seasonal dependency does not exist in the Finnish background catchments where the carbon contents are consistently 30 - 40% (see C8 as an example in Fig. 7.4.2). Our interpretation is that the organic matter is only loosely bound in the Russian catchments where vegetation is severely damaged. During snow melt it is thus eroded and flushed into the aquatic environment. During summer it can additionally be leached to lower levels of the podzol profiles. In all catchments the lowest C, H, N-contents can be observed in the summer samples while an increase occurs towards the fall (fresh supply of litter).



### *Nickel*

The total concentrations of Ni in the topsoil of C1, C2 and C4 are strongly increased when compared to the Finnish background catchments (Fig. 7.4.3,4), while they are only slightly increased in C3 and C5 (Reimann et al., 1996c).

No predominant seasonal variations can be observed in the total nickel concentrations in C5, C6, C7 and C8 (for C8 see Fig. 7.4.3). In C3, a slight decrease from a winter maximum towards spring and summer followed by an increase in the fall can be observed. In C1, C2 and C4 winter concentrations are up to four times higher than the concentrations measured during the rest of the year (Fig. 7.4.3). Note that the scales used for constructing the boxplots differ significantly between the catchments. As for C, H, N, the lowest concentrations are observed for the summer samples. C2, where vegetation is nearly completely damaged, shows the largest differences between winter and summer samples.

The seasonal variations of easily leachable Ni in C1, C2 and C4 closely follow the patterns described above for the total contents. C1 and C2 show the largest differences with a large decrease of available Ni from winter to spring.

The easily leachable proportion of nickel (calculated as percentage of ammonium acetate leachable contents of the total contents) is surprisingly high in C1 and C5 (Fig. 7.4.4). This result is similar to the regional results obtained in the surroundings of Nikel and Zapoljarnij during the pilot project (Niskavaara et al., 1996), where 20-30% of the total Ni-contents in the O-horizon were observed in an easily leachable form. Although the total concentrations in C2 are up to 6 times higher than in C1, the easily leachable concentrations are roughly the same. In C4 the easily leachable Ni-contents in the topsoil are comparatively higher than in C2, revealing that close to the smelter a large proportion of the nickel must be deposited in particulate form, while at a distance of only 30 km (C4) the soluble input begins to prevail. The same effect was noticed by Äyräs et al. (1996b), comparing particulate vs. water soluble deposition using snowpack samples. They found that about 70% of the deposited Ni is in particulate form in C2. In other catchments the easily leachable portion of nickel in the topsoils varies between 5 - 20% of the total concentrations (Fig. 7.4.4).

### *Copper*

As for Ni, total concentrations of Cu in C1, C2 and C4 are strongly increased compared to the low concentrations observed in the Finnish catchments C6, C7 and C8 (Fig. 7.4.4, 5). C3 and C5 take an intermediate position with regards to the Cu contents in the topsoils. Note that again for constructing the boxplots in Fig. 5 different scales had to be used for each catchment.

No seasonal variations are noticed in C5, C6, C7 and C8 (Fig 7.4.5). In the strongly polluted catchments C1, C2 and C4 the seasonal variations of the Cu-contents are following the same patterns as observed for Ni - maximum concentrations in winter and minimum levels in summer. This seasonal effect is biggest in C1 and C2 - the Cu-concentrations measured in the summer samples are only about 40% of those observed in the winter samples.

The seasonal variation of the easily leachable Cu-contents follows the trends observed for the total concentrations, an exception being C1 and C2 where minimum concentrations are already observed in spring (Fig. 7.4.5). Considering the big variation in easily leachable concentrations and the high maximum values, particularly in C2, the drop in concentrations is dramatic in spring. Even the variation diminishes prominently.

In C2 exceptionally high concentrations of easily leachable Cu are noticed (median 440 ppm; max. 1800 ppm) (Fig. 7.4.4). In addition the highest proportion of easily leachable Cu is noticed in C2 (Fig. 7.4.4, 8). The same was observed for Cu in podzol profile samples from C2 where high easily leachable concentrations of Cu were found in humus (about 20%), in the eluvial layer (about 45%) and in the B1 layer (about 15%) but not in the lower podzol layers (Räisänen et al. 1996). This is directly related to airborne deposition. Äyräs et al. (1996b) report that about 85% of the Cu in snow samples from C2 occurs in water soluble forms. The catchments C1 and C4 show also a high proportion of easily leachable Cu, while in the other catchments less than 10% of the total Cu-contents are available.

### *Sulphur*

Results obtained for sulphur are rather surprising at the first glance. The highest concentrations of sulphur are not found in the Russian catchments, receiving the highest S-deposition, but in the Finnish catchments (C8 in Fig. 7.4.6). This can be explained by the strong positive correlation of sulphur and the C+H+N-content of the samples (Reimann et al. 1996c). A strong seasonal variation can, however, be observed for the total S-contents in the topsoils of the polluted catchments with highest values observed in winter and minimum values occurring in summer.

The seasonal variation of easily leachable S follows the patterns displayed by the total contents in C2 and C4. In C1 where the lowest easily leachable S-concentrations can be observed during the spring.

Easily leachable S-contents are exceptionally high in C2 (up to >20%) and considerably lower (<15%) in all other catchments (Fig. 7.4.8). Räisänen et al (1996) observed in C2 that in a strongly eroded soil profile all of the S occurred in the mobile form in the B-horizon.

Sulphur is not retained and enriched in the topsoil. Sulphur originating from atmospheric deposition is probably percolating directly through the topsoil towards deeper soil layers (B-layer) and the groundwater or is immediately washed out via surface flow into the streams draining the catchments. The dominant factor regulating the sulphur content of the topsoils is the amount of organic material and thus the background catchments show the highest S-concentrations in the topsoils. When the sulphur results are normalised against (C+H+N) clear differences between the catchments and within one catchment become visible (Reimann et al. 1996c). Biologically bound nutrient sulphur is partly washed out to the streams together with the soil organic matter where it concentrates in the sediments (Niskavaara et al., 1996). Humus is thus not a good indicator of airborne S-deposition because of the very low contrast between natural levels and additional, anthropogenic input even in areas with very high deposition values (Niskavaara et al. 1996).

### *Base cations (Ca, Mg, K, Na)*

Total concentrations for these elements do not show any marked differences between the catchments with the exception of C3, which is dominated by alkaline rocks (Reimann et al., 1996c). The easily leachable concentrations of the base cations show, however, by far the lowest concentrations in C1 to C4 in Russia, topsoils from C2 showing the minimum contents (Mg see Fig. 7.7.4, 7).

In terms of seasonal variations there exists a decrease in total and easily leachable element contents from winter through spring towards the summer in C1, C2 and C4. Values are increasing again in the autumn (Fig. 7.4.7).

The easily leachable proportion of base cations in C6, C7 and C8 is in the range of 50-90% (Fig. 7.4.8). In the contaminated catchments this proportion is generally <60%. This depletion is most prominent for Na and Mg being. This proves that the pool of exchangeable base cations is severely depleted due to anthropogenic contamination (compare Reimann et al. in prep. a). The proportion of easily leachable vs. total element content can thus be taken as a measure of soil damage. Using this approach catchments C3 and C5 shows also signs of Ca and Na depletion indicating some damage to the pool of exchangeable base cations.

The cation exchange capacity (CEC), calculated as a sum of milliequivalent concentrations of ammonium acetate leachable Ca, Mg, Na, K and Al, ranges from 30 meq/kg to 400 meq/kg, being lowest in C2 and C4 (median around 50 meq/kg in summer).

#### *Aluminium and iron*

The total concentrations of Al are highest in catchments C3 and C2 (Fig. 7.4.4). The exceptional geological setting in C3 explains these really high concentrations, which are also seen in the easily leachable concentrations. The high aqua regia soluble concentrations in C2 can be explained either by geology or they show that the eroding effect of acidic deposition has broken the structures of aluminosilicates so that they are more accessible to the dissolving effect of aqua regia. For iron the highest concentrations are found in C2 reflecting the same process (Reimann et al, 1996c).

In the background catchments C6, C7 and C8 the easily leachable proportion of Fe is steadily increasing with increasing total Fe-content. Al shows a similar trend. This correlation does not exist in the other catchments. In C8 a high proportion of Al and Fe occur in easily leachable forms (Fig 7.4.8). For Al, however, the most dominant feature is the strong deviation that the values from C3 show to all other catchments due to its exceptional geology (Fig. 7.4.4). Seasonal variations are not observed for these two elements in any of the catchments or in the two different extractions used.

## DISCUSSION

A strong positive correlation exists between many of the emission related elements as well as the nutrient elements and the content of organic matter (C+H+N) of the studied topsoil samples. The content of organic matter is thus the most dominant factor controlling concentration and retention of these elements in the topsoils. The concentration of most of these elements is highest in winter and lowest in July. This is explained by the fact that during late autumn and winter the debris of the dying vegetation is accumulated on the topsoil, increasing the contents of all these components. In addition the capacity of the soil to retain water is higher at this time of the year than during the summer.

During the snow melt episode in spring a considerable amount of organic matter is washed out of the most contaminated catchments. Due to the high level of pollution, vegetation and the uppermost soil layer are extensively damaged and the mechanisms to tie the newly developed humus to the soil layer are not functioning. Elements in easily leachable form and those bound to the organic matter are thus leached out as surface flow during snow melt. Stream sediments and lake sediments will act as the primary sinks for these elements. The loss of organic matter and associated elements continues throughout the summer. In addition the nutrient elements liberated during weathering and erosion are effectively removed from the topsoil. During summer the dominating mechanism is probably the leaching of these components to lower soil layers (E- and B-horizon) and to the groundwater. Wind and rain erosion of the damaged upper soil layer can also increase this loss.

In pristine areas where a real humus layer and a healthy vegetation exist this washout of organic matter is not so prominent. A general increase in element contents is again observed in the fall, when the organic content of the soil is slowly increasing due to decaying vegetation. The observed differences between autumn and winter samples are, however, still surprisingly high - concentrations in autumn are only half of the winter concentrations for some pollutants. During the winter season, when the topsoil is frozen, it is well protected from further erosion and demobilisation of elements. Elements percolating through the snow cover during occasional melt episodes may further accumulate in the topsoil layer during this period.

The leaching and transport mechanisms of heavy metals differ from each other. The transport of Cu to surface water and lower soil layers happens mostly in forms bound to organic substances (compare Borg and Johansson, 1989) while Ni (and Cd, Mn, Zn, Pb, Co) represents an element which is typically transported as acid-mobilised easily leachable compound.

The base cations are typically occupying the exchange sites of bulk soil and exist in high proportions in easily leachable forms in the background catchments. The easily leachable portion of base cations (particularly Na and Mg) has decreased considerably in C1, C2, C3 and C4. Also C5 shows signs of depletion of the pool of exchangeable base cations. This indicates damages in the buffering system of the topsoil in these catchments.

An interesting difference in the behaviour of some typical pollutants is noticed in two most damaged catchments C1 and C2. In C1, close to the nickel ore roasting plant at Zapoljarnij, an exceptionally high amount of the total Ni-concentration occurs in an easily leachable form, while Cu and S are more strongly bound. In C2, close to the smelters in Monchegorsk, Ni is rather well bound, probably as insoluble particulate emission products (oxide or sulphides), and a high proportion of the total Cu- and S-contents occur in an easily leachable form. The Cu-deposition in this catchment is so high that the capacity of the limited amount of organic matter to bind Cu is exceeded. Thus a substantial part of Cu is retained in soluble form in the winter samples. This soluble part of Cu is, in addition to the part of Cu bound to the organic matter, to a large extent lost during the snow melt period. For this reason we observe an exceptionally large drop of total and particularly easily leachable Cu-concentrations during the snowmelt period.

## CONCLUSIONS

A simple and straight forward method using two different extractions: (1) an ISO-standardised aqua regia digestion (to determine the so called total element contents) and (2) a 1M ammonium acetate leach buffered to pH 4,5 - provides valuable information on element pools, mobilities and pathways in the surrounding of the Cu/Ni-industry on the Kola Peninsula, Russia. The total concentrations give an overview of the level of pollution. High total element concentrations can, however, be produced by natural, geogenic factors (e.g. mineralisation) as well. Hence additional information, as obtained by a weak leach, is needed to estimate the fate of elements in the terrestrial environment.

Using proportions of easily leachable concentrations to total concentrations provides a promising and quick tool to differentiate "healthy" and damaged soils. Although large variations in easily leachable element proportions exist between catchments and seasons and even within a single catchment a clear sequence of leachability of elements in the ammonium acetate extraction can be established: Cd > K > Mg > Na > Ca = Sr > Mn > Zn > Ba = Pb > Co > As > Ni > P, S, Al > Cr, Fe, Cu, V.

Clear deviations from this sequence can be observed in the polluted catchments, depending on the characteristics of the deposition and the physicochemical status of the topsoil. These differences are especially large for the base cations (particularly Mg and Na), Cu, Ni and S.

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## TABLES

Table 7.4.1: Main characteristics of the 8 catchments investigated (for location within the study area refer to Fig. 7.4.1)

## FIGURES

Fig. 7.4.1: Location of the study area for regional geochemical mapping (frame) and of the eight catchments discussed herein (1: Zapoljarnij, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska and 8: Pallas).

Fig. 7.4.2: Boxplot comparison of concentration levels and seasonal variations of carbon and nitrogen in catchments C1, C2, C4 and C8

Fig. 7.4.3: Boxplot comparison of concentration levels and seasonal variations of total and easily leachable concentrations of nickel in catchments C1, C2, C4 and C8

Fig. 7.4.4: XY-diagrams showing the total element contents of Cu, Ni, Mg and Al plotted against the proportion (%) of easily available element contents in the 8 catchments studied.

Fig. 7.4.5: Boxplot comparison of concentration levels and seasonal variations of total and easily leachable concentrations of copper in catchments C1, C2, C4 and C8

Fig. 7.4.6: Boxplot comparison of concentration levels and seasonal variations of total and easily leachable concentrations of sulphur in catchments C1, C2, C4 and C8

Fig. 7.4.7: Boxplot comparison of concentration levels and seasonal variations of total and easily leachable concentrations of magnesium in catchments C1, C2, C4 and C8

Fig. 7.4.8: Proportion (%) of easily leachable concentration from total concentrations of selected elements in catchments C1 - C8.



No.	Name	Coordinates of catchment outlet	Size (km <sup>2</sup> )	Elevation (m a.s.l.)	Annual precip. (mm)*	Vegetation	Bedrock	Surface cover, peculiarities
RUSSIA								
C1	Zapoljarniy	69°27'01"N 31°03'49"E	19.02	25-373	454	birch forest tundra	gneiss	till, fluvioglacial, outcrop
C2	Monchegorsk	67°50'30"N 32°54'48"E	22.38	128-507	391	technogenic desert, birch shrubs	dacite & andesite & tuffs, gabbro/norite	till, prone to erosion
C3	Kirovsk	67°32'50"N 33°48'55"E	20.01	240-1075	502	spruce forest, mountain tundra birch forest	nephelinite	till, diluvial/eluvial
C4	Kurka	67°41'25"N 32°50'14"E	20.49	152-466	502	north taiga spruce forest, birch; incipient deterioration	amphibolite, gneiss	till, fluvio-glacial
NORWAY								
C5	Skjellbekken	69°21'25"N 29°27'25"E	34.56	80-297	422	north taiga pine forest, birch	andesite, basalt & tuffs, 'black shale'	till, esker
FINLAND								
C6	Kirakka	69°35'12"N 28°51'46"E	11.86	110-200	386	north taiga pine forest	granite	outcrop, till, moraine ridge
C7	Naruska	67°21'44"N 29°22'05"E	20.16	263-490	513	north taiga spruce forest	gneiss	till, peat, outcrop
C8	Pallas	68°09'14"N 23°52'50"E	24.42	303-500	405	north taiga spruce forest	quartzite	till, peat

\* from the closest meteorological station (data from 1994)

Table 7.4.1: Main characteristics of the 8 catchments investigated (for location within the study area refer to Fig. 7.4.1)

*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
Catchment locations

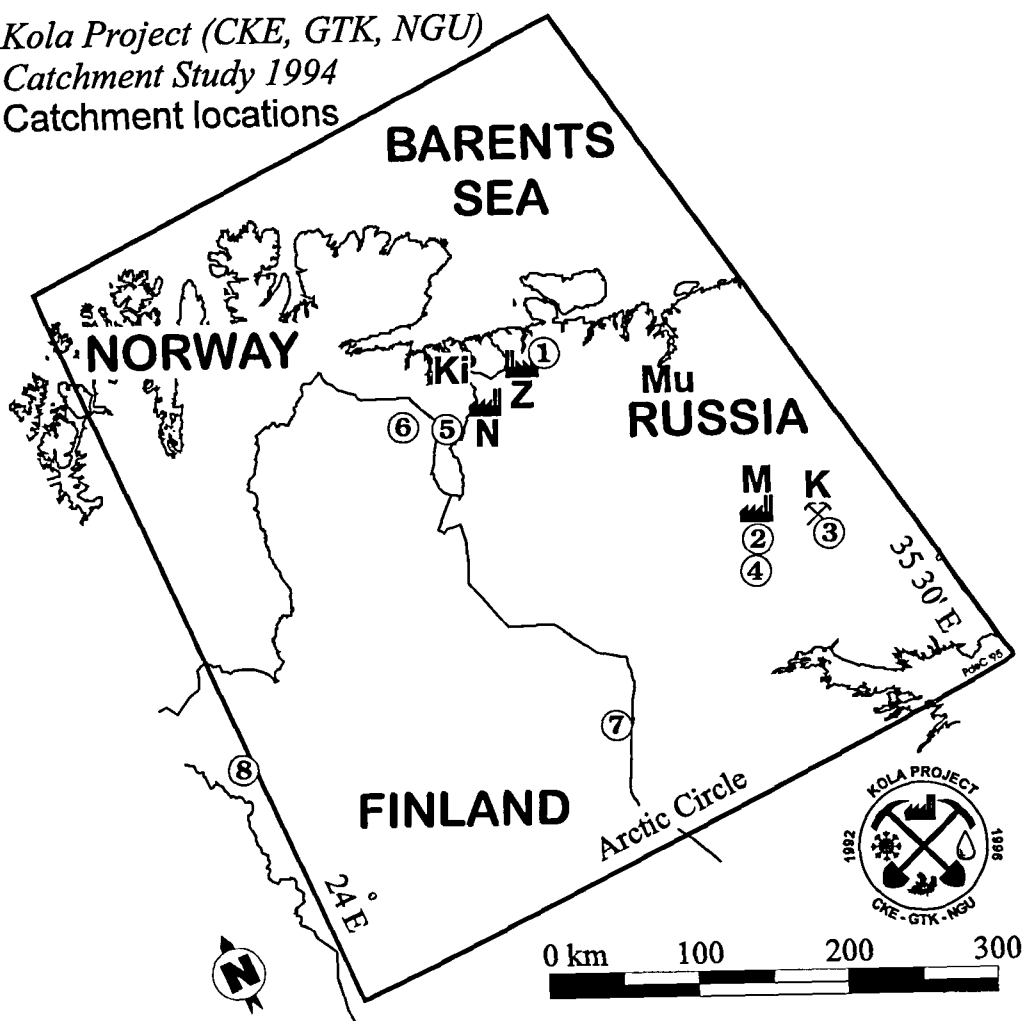


Fig. 7.4.1: Location of the study area for regional geochemical mapping (frame) and of the eight catchments discussed herein (1: Zapoljarnij, 2: Monchegorsk, 3: Kirovsk, 4: Kurka, 5: Skjellbekken, 6: Kirakka, 7: Naruska and 8: Pallas).

Fig. 7.4.2: Boxplot comparison of concentration levels and seasonal variations of carbon and nitrogen in catchments C1, C2, C4 and C8

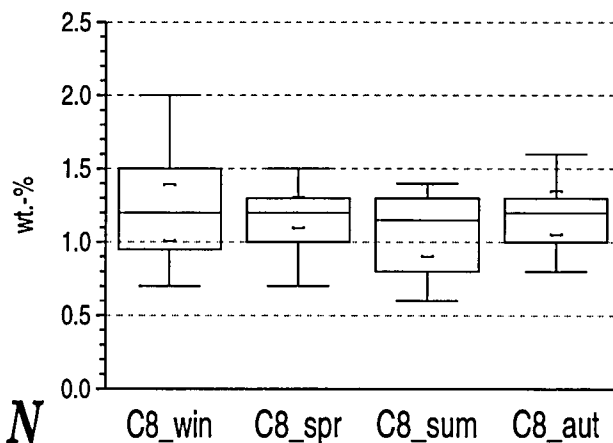
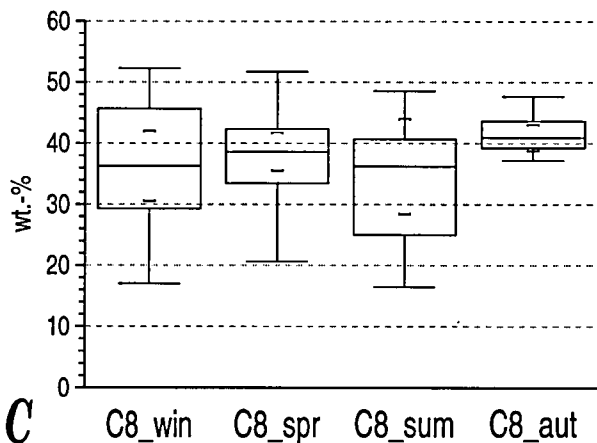
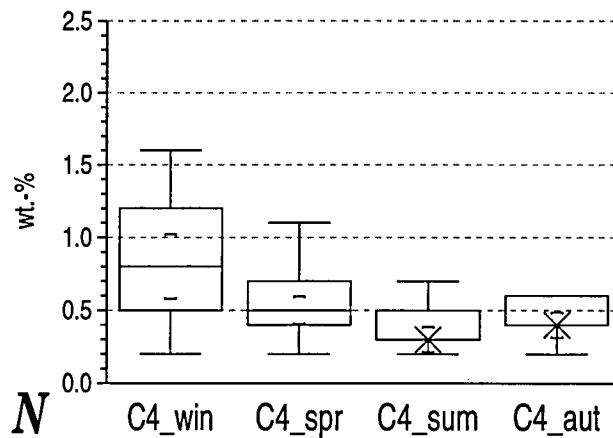
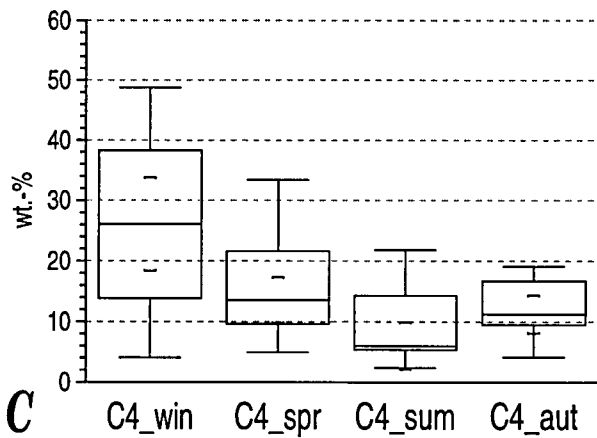
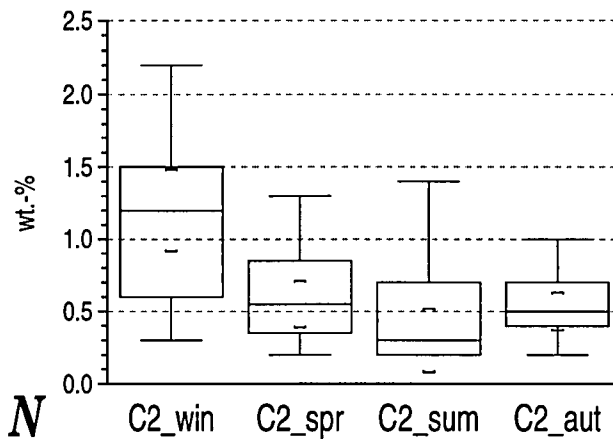
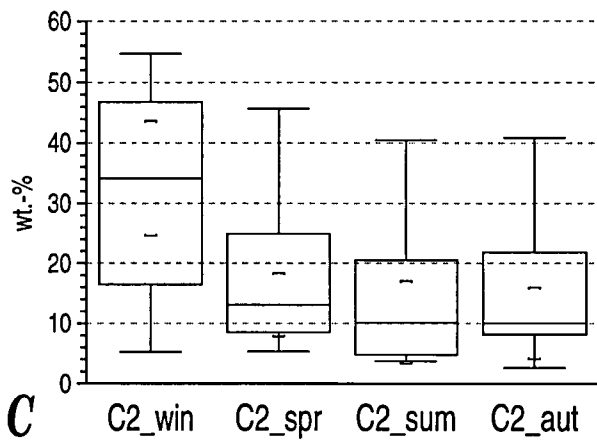
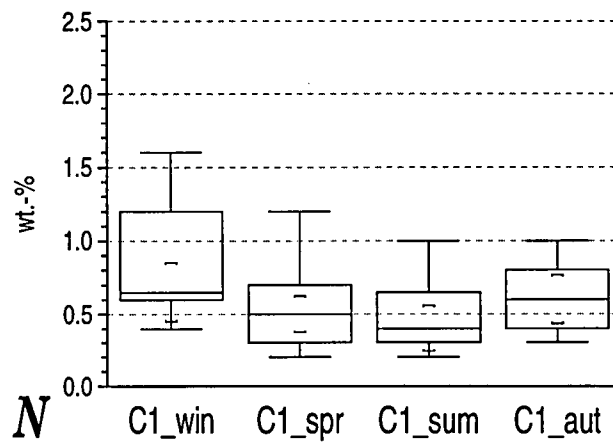
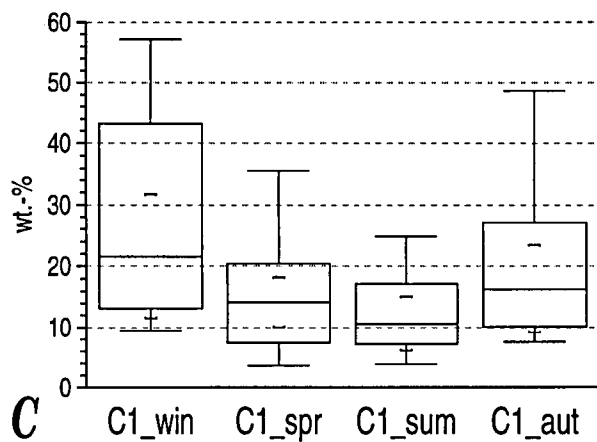


Fig. 7.4.3: Boxplot comparison of concentration levels and seasonal variations of total and easily leachable concentrations of nickel in catchments C1, C2, C4 and C8

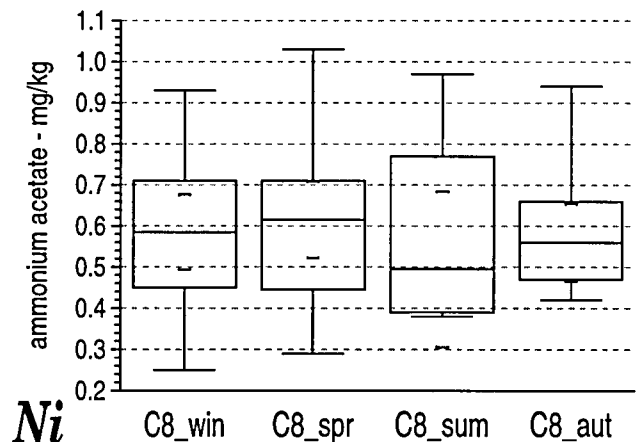
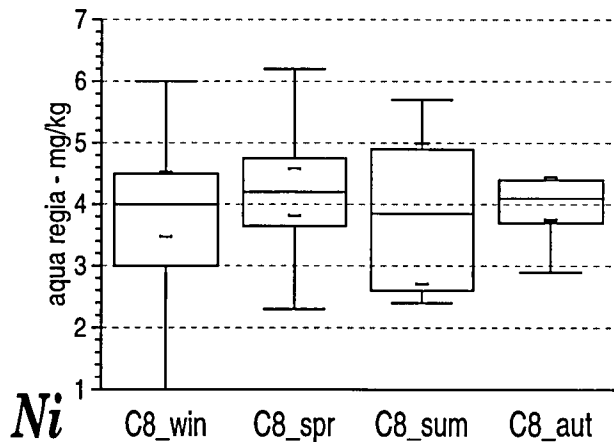
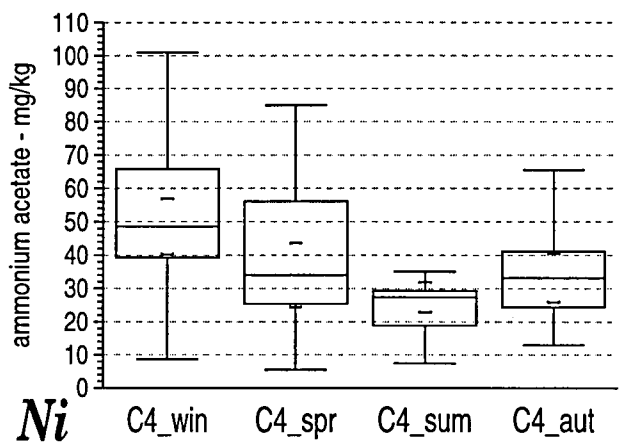
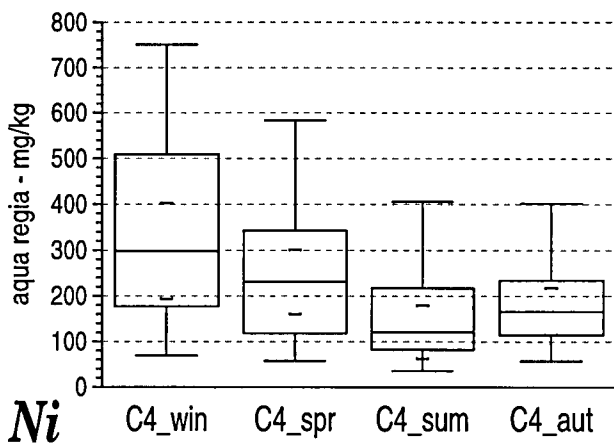
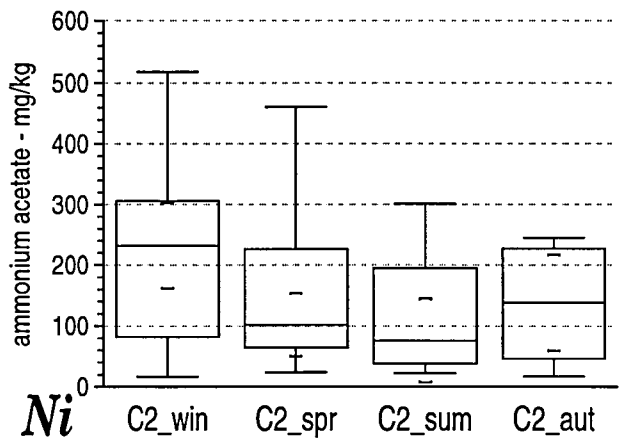
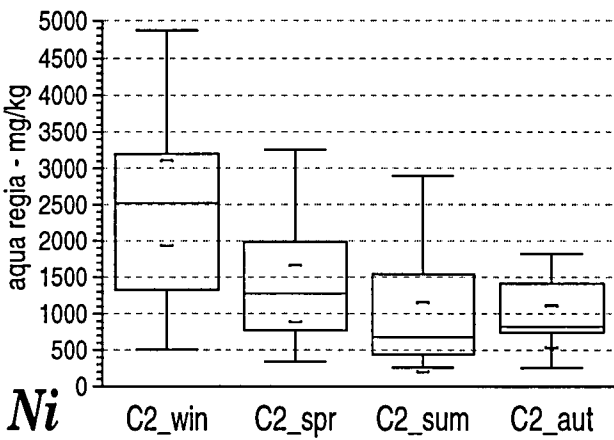
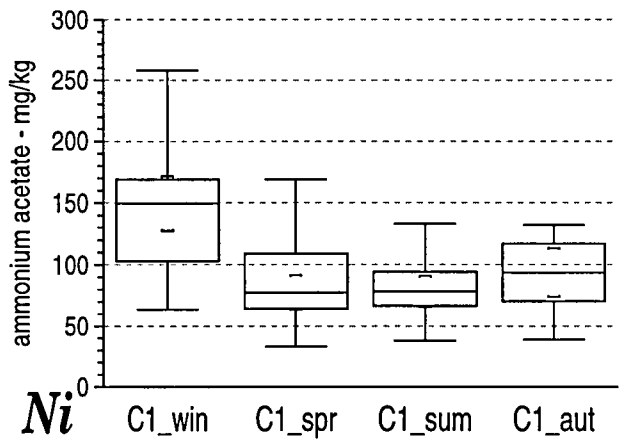
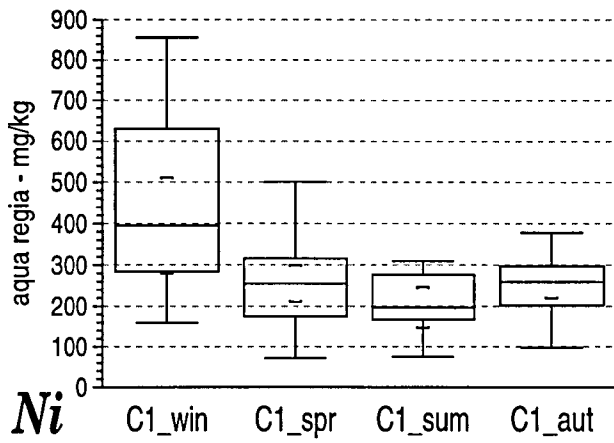


Fig. 7.4.4: XY-diagrams showing the total element contents of Cu, Ni, Mg and Al plotted against the proportion (%) of easily available element contents in the 8 catchments studied.

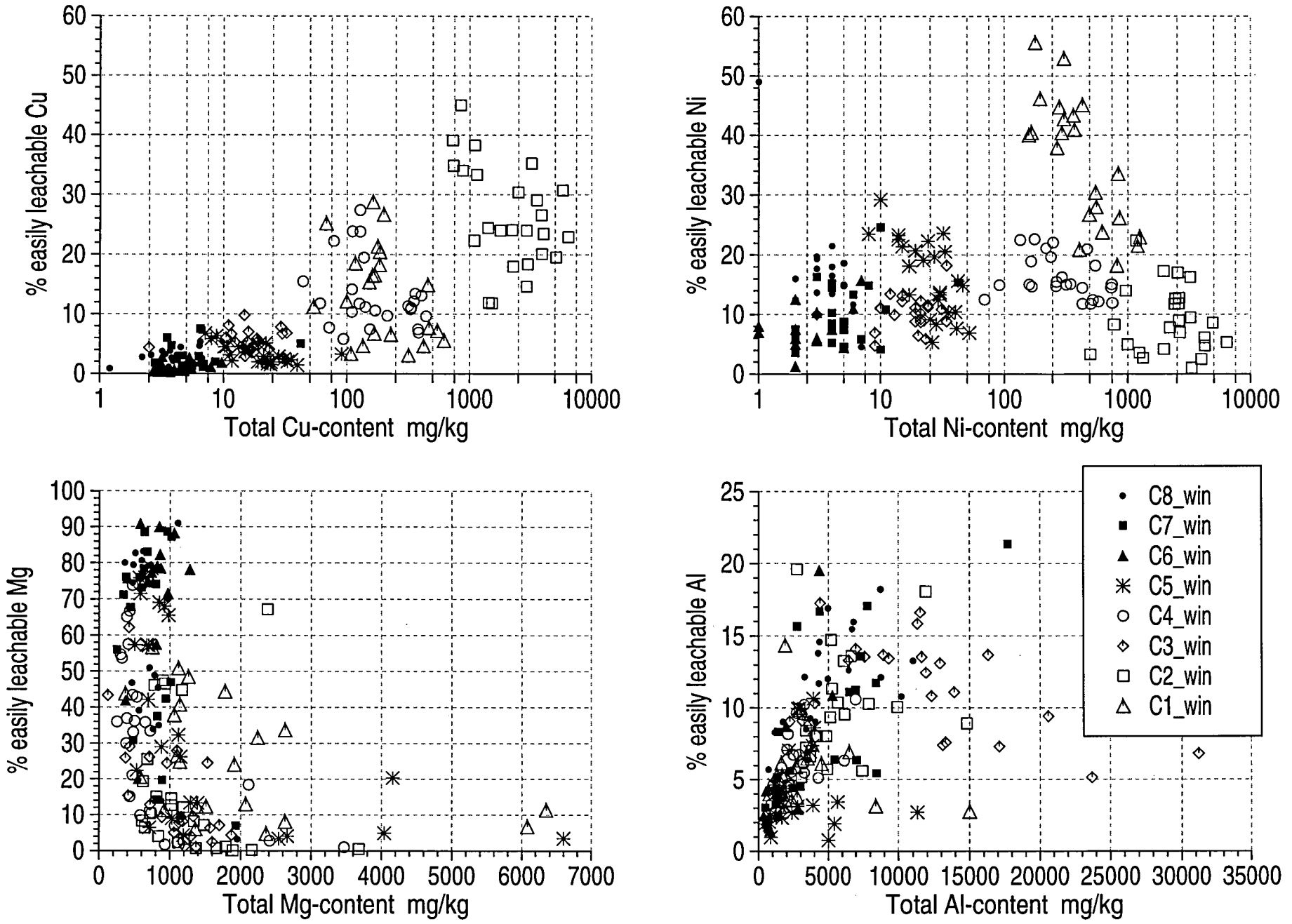


Fig. 7.4.5: Boxplot comparison of concentration levels and seasonal variations of total and easily leachable concentrations of copper in catchments C1, C2, C4 and C8

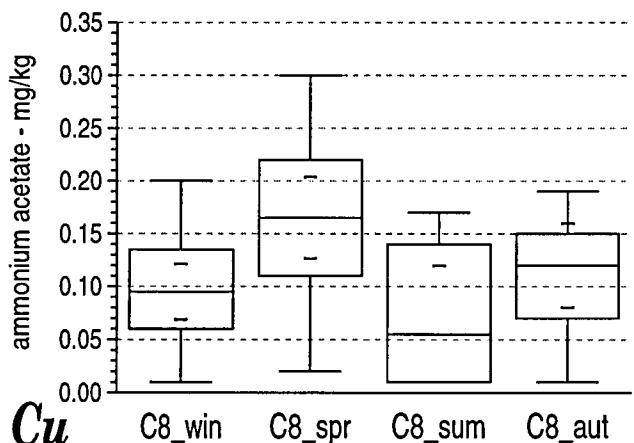
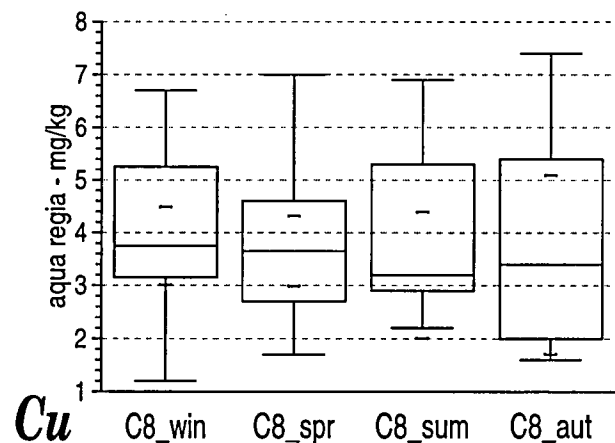
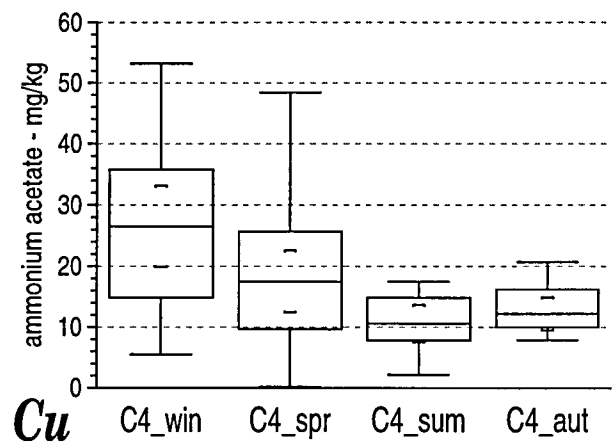
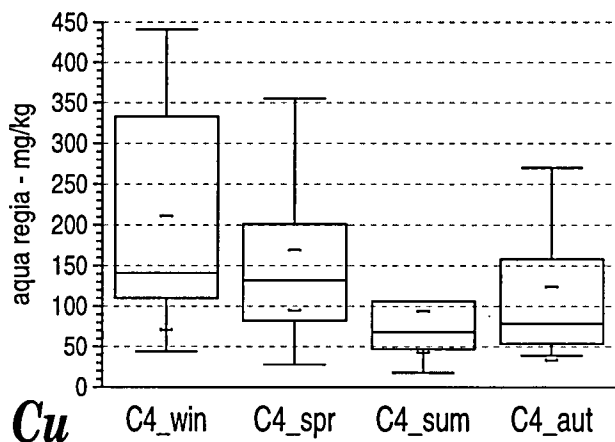
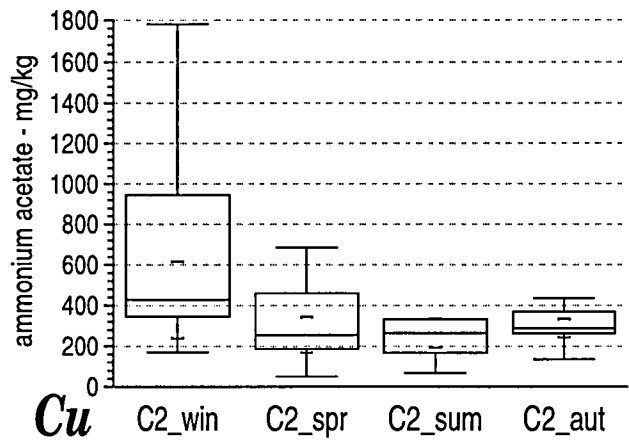
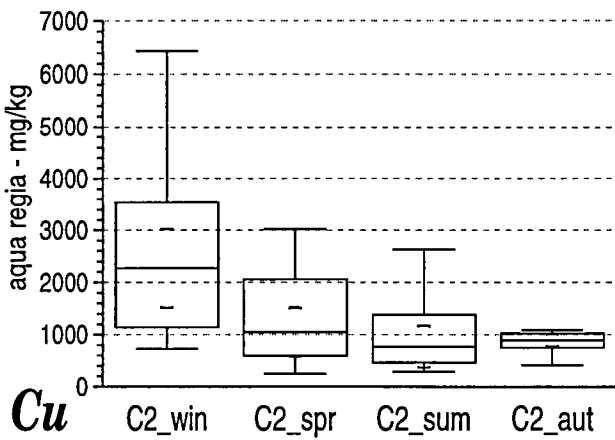
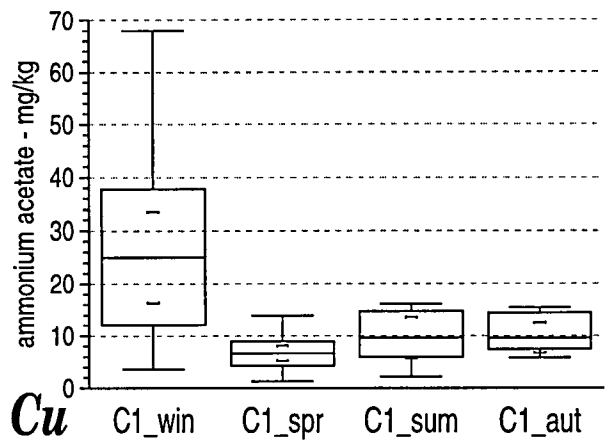
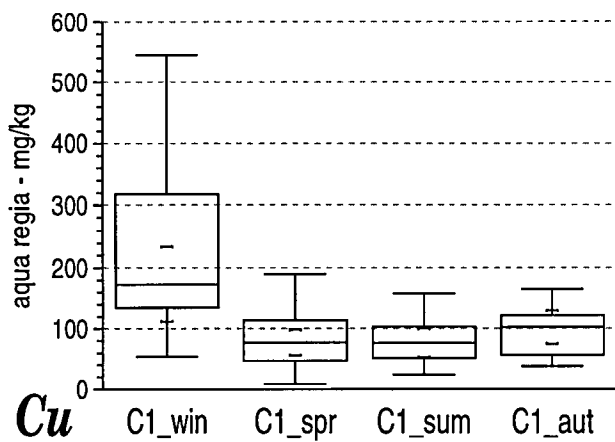


Fig. 7.4.6: Boxplot comparison of concentration levels and seasonal variations of total and easily leachable concentrations of sulphur in catchments C1, C2, C4 and C8

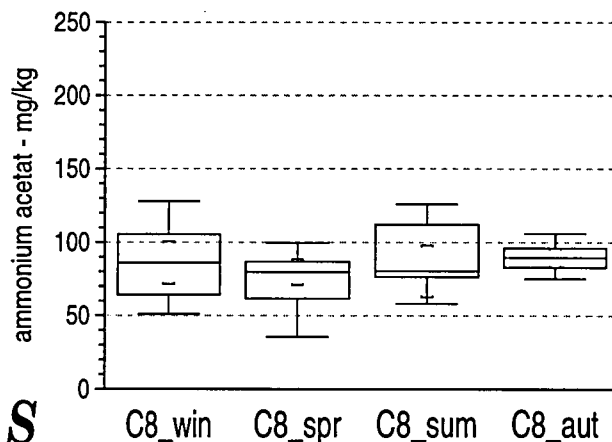
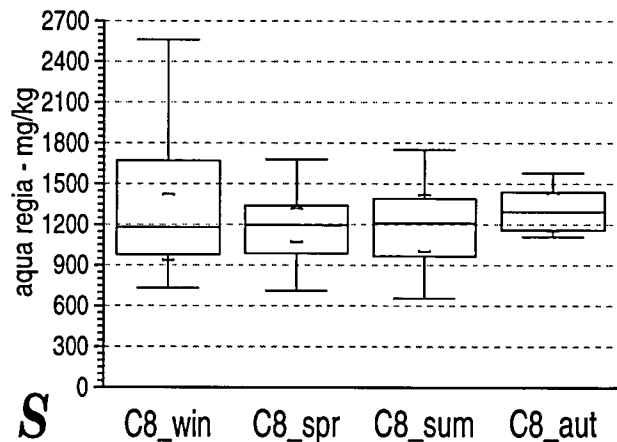
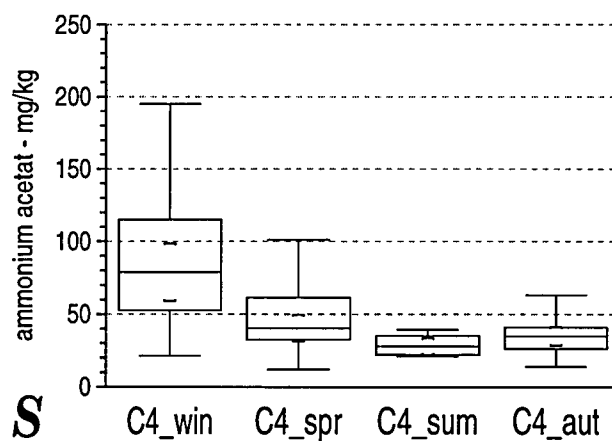
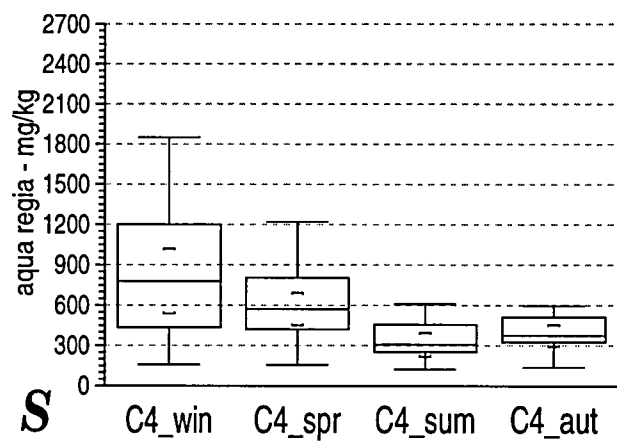
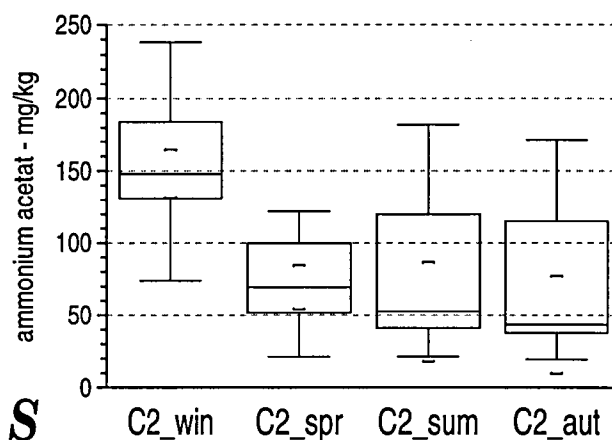
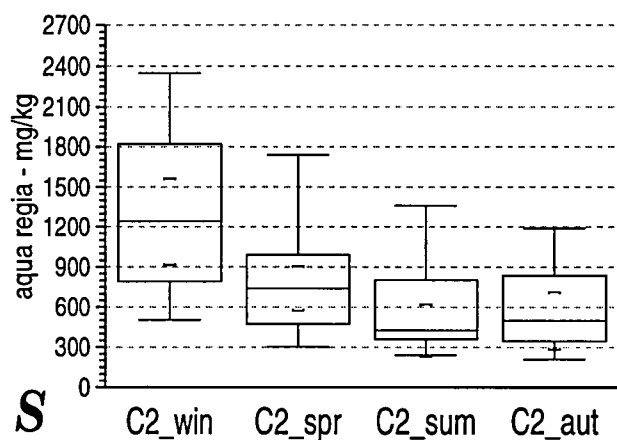
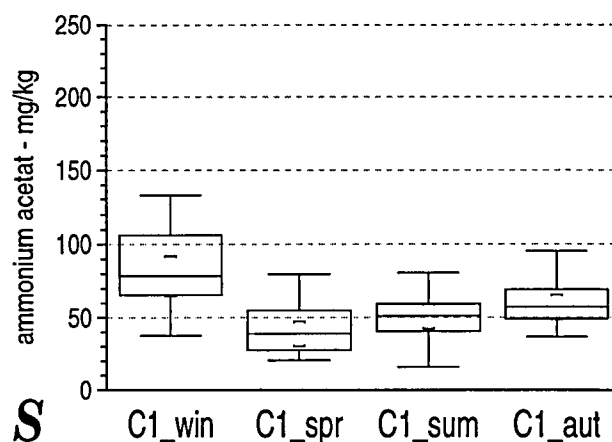
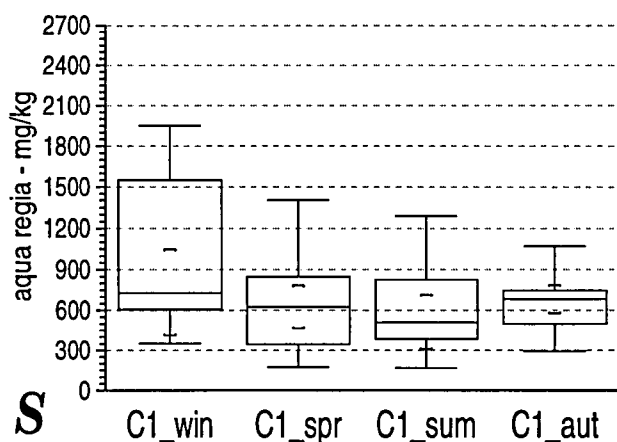


Fig. 7.4.7: Boxplot comparison of concentration levels and seasonal variations of total and easily leachable concentrations of magnesium in catchments C1, C2, C4 and C8

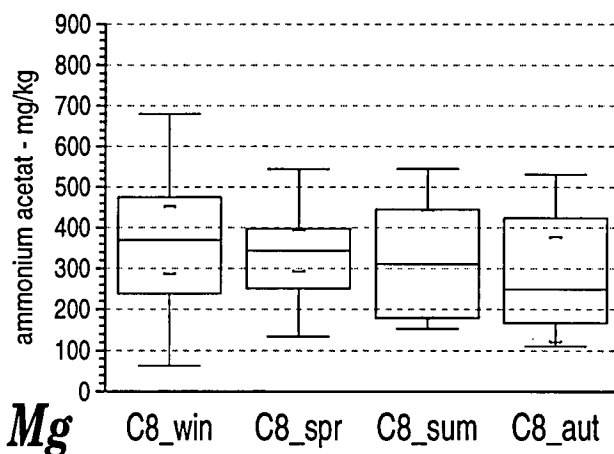
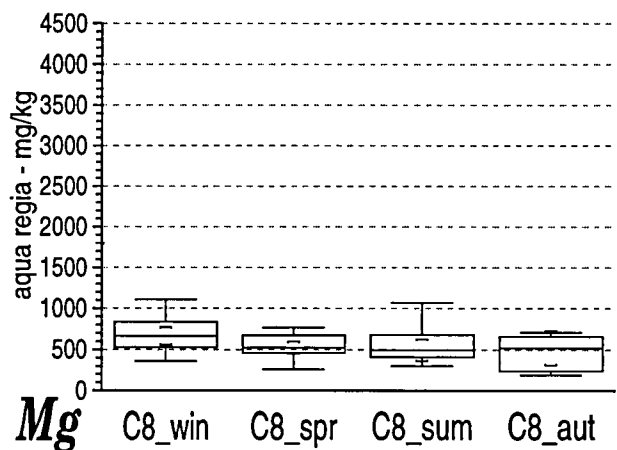
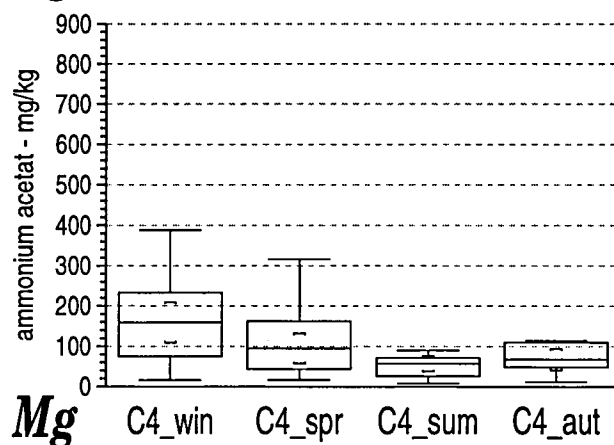
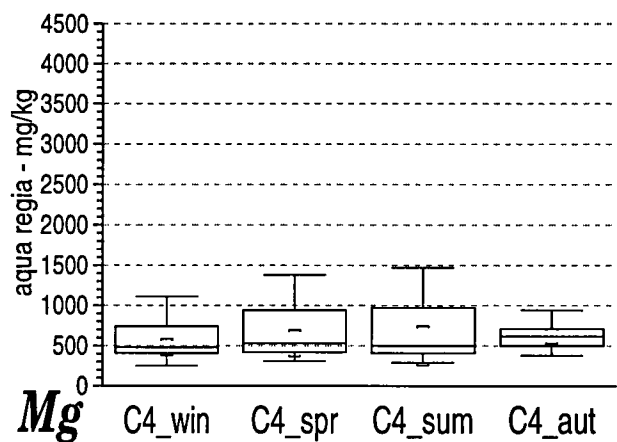
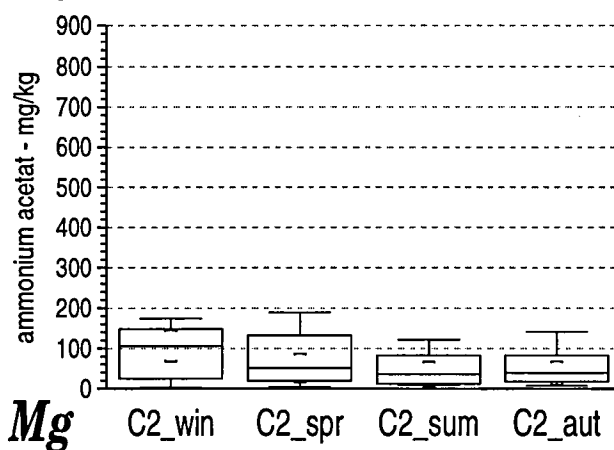
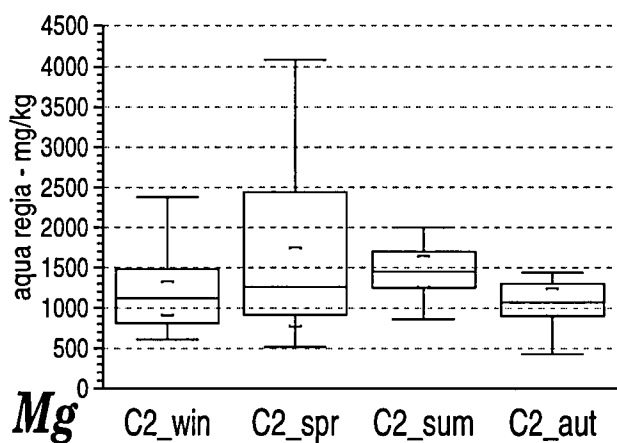
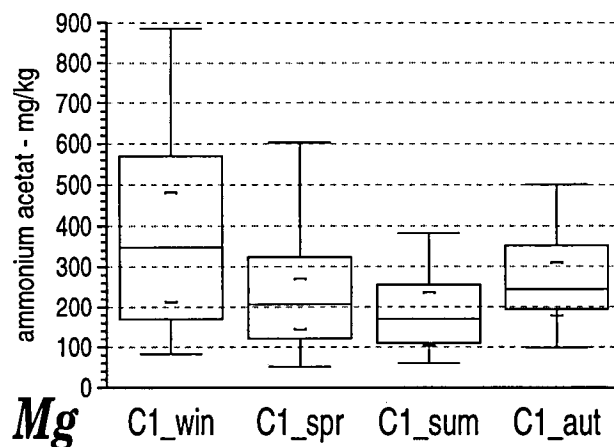
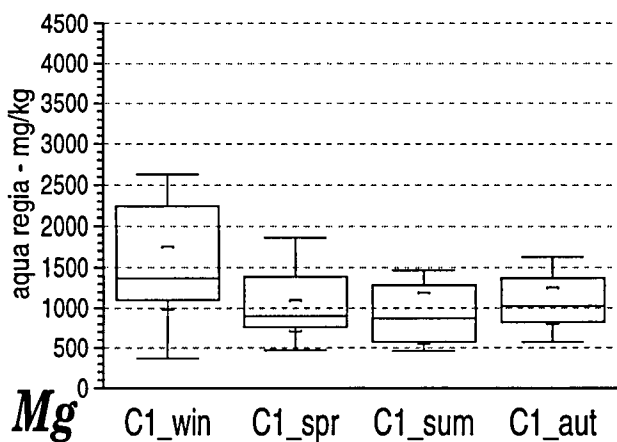
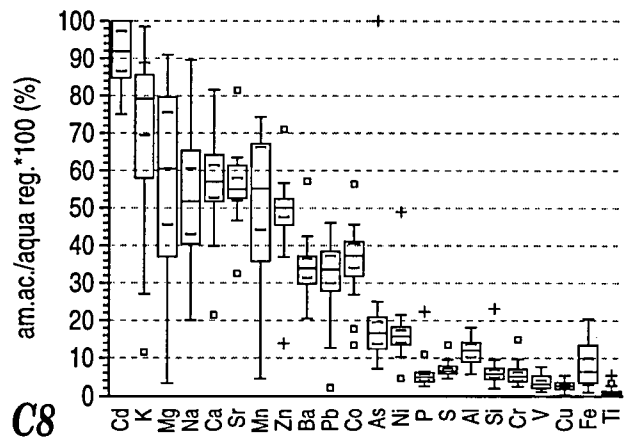
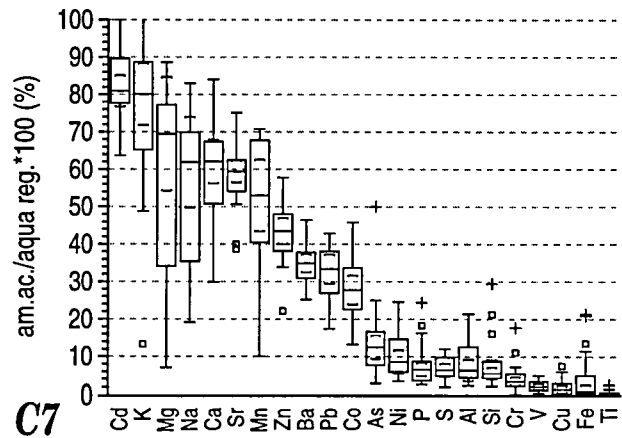
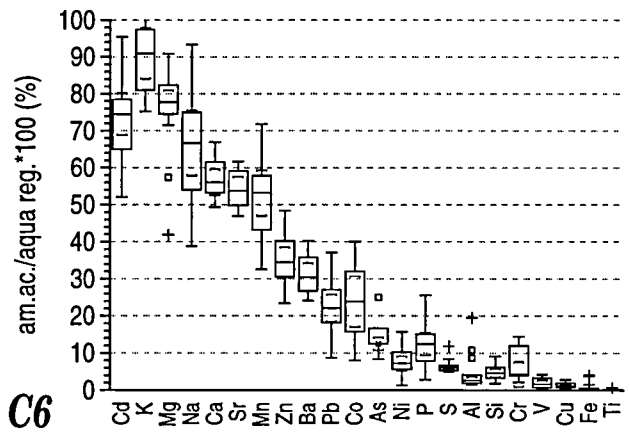
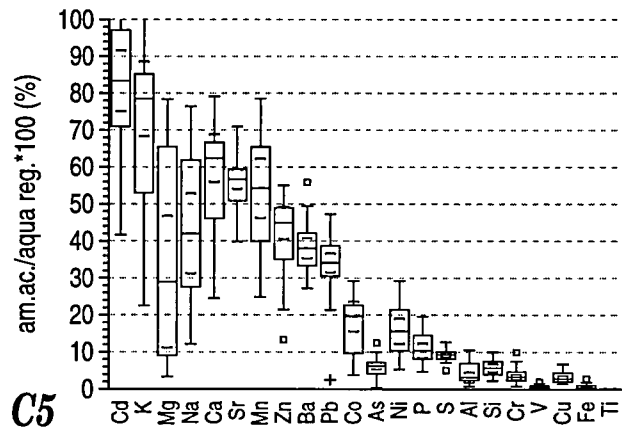
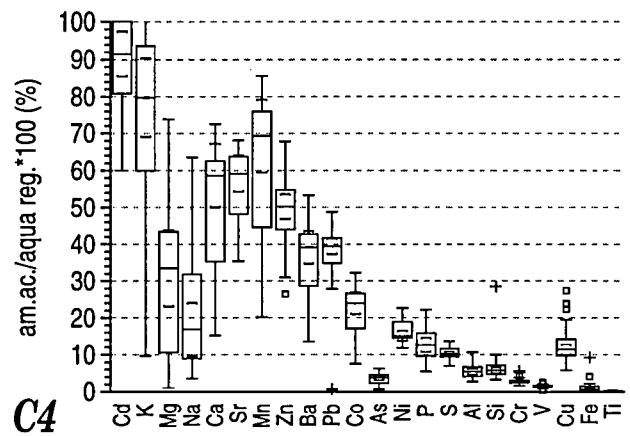
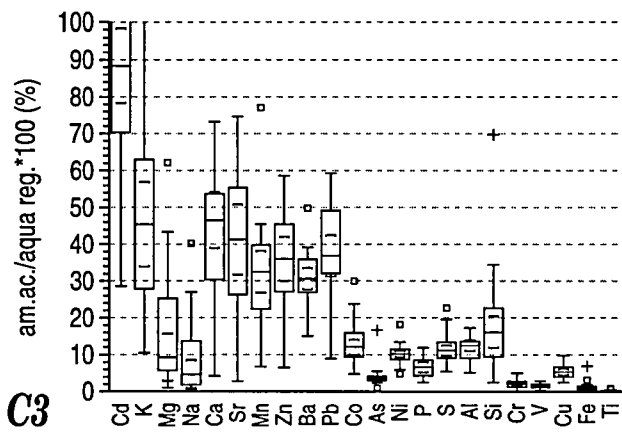
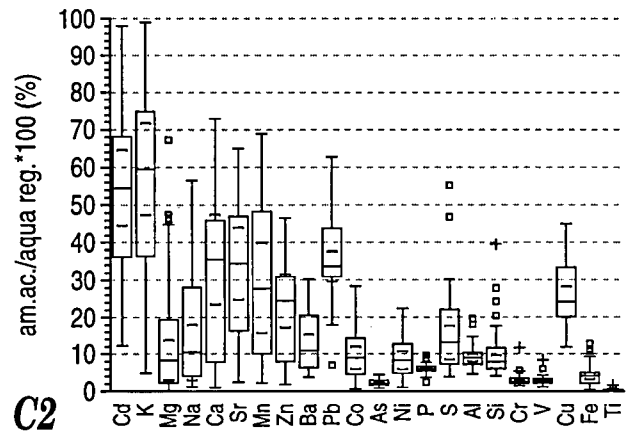
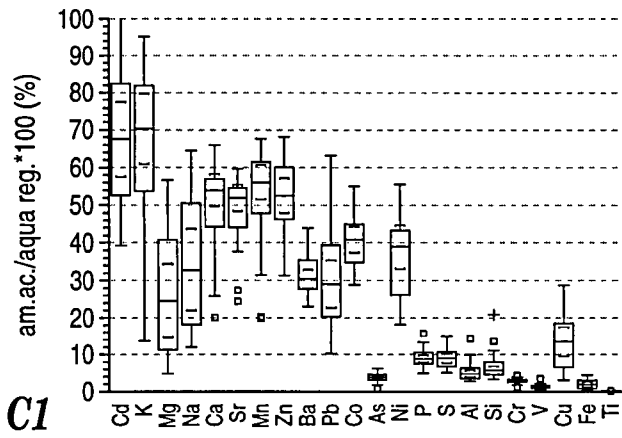




Fig. 7.4.8: Proportion (%) of easily leachable concentration from total concentrations of selected elements in catchments C1 - C8.



## 8 PODZOL PROFILES

### 8.1 Acidity status and mobility of Al in podzols near SO<sub>2</sub> emission sources on the Kola Peninsula, NW Russia

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#### ABSTRACT

The soil acidity status and mobility of Al in podzols were examined on a broad scale near large emission sources for SO<sub>2</sub> and heavy metals on the Kola Peninsula in northwestern Russia (the Severonikel and Pechenganikel smelter complexes) and in neighbouring parts of Norway and Finland. Acidification of the upper podzol horizons and depletion of mobile basic cations were only evident at sites where ecosystems are completely destroyed, in the immediate vicinity of the Severonikel smelter complex. The high content of basic cations in the parent material (till) near the emission sources may mask the acidification effect of the pollution. Both strong anthropogenic (SO<sub>2</sub>) emissions in completely destroyed ecosystems and natural acidification (*in situ* weathering of black schist) intensify weathering and mobilise Al. However, drainage conditions seem to be the most important factor determining the content of mobile Al in the podzols. Under free drainage conditions, mobilised Al leaches out of soil profiles.

#### INTRODUCTION

Three of the largest sources of SO<sub>2</sub> and heavy metal pollution in northern Europe are located on the western part of the Kola Peninsula (northwestern Russia). These are the Severonikel nickel-copper ore smelter at Monchegorsk and the Pechenganikel complex consisting of the ore smelter at Nikel and the ore roasting plant at Zapoljarnij (Fig. 8.1.1). Together they accounted in 1994 for about 300,000 t of SO<sub>2</sub>, 1900 t of Ni, 1100 t of Cu and 94 t of V<sub>2</sub>O<sub>5</sub> emissions yearly (Reimann et al., 1996a). The cumulative SO<sub>2</sub> emission from the Severonikel smelter complex alone was about 6.5 million tonnes from 1960 to 1994 (Caritat et al., 1996a). In addition, there are several other sources of SO<sub>2</sub> emissions in the area (coal-fired power plants, other industry).

The long-term effect of such a high level of pollution is severe and widespread ecosystem degradation in the region, and locally (in the nearest vicinity of Monchegorsk and Nikel) completely disturbed ecosystems are characterised by intensive soil erosion (Kryshkov, 1991, Hogda et al., 1995, Solheim et al., 1995). The accumulation of heavy metals in the various media, including soil, near Monchegorsk and Nikel is quite well documented (Kozlov et al., 1993, Åyrås et al., 1995, Reimann et al., 1995a, b, 1996a, b, Caritat et al., 1996, Niskavaara et al., 1996).

Even though SO<sub>2</sub> is the prevalent component of the emissions, soil acidification has been little investigated in this area. Most studies are based on transect sampling with sparse intervals. Results obtained so far are quite contradictory. On the one hand, experiments to study the direct interaction between precipitation and soil samples showed that pollution from the Severonikel smelter complex may be the cause of podzol acidification and basic cation leaching (Kashulina et al., 1995, Koptsik and Muchina, 1995). In the field, on the other hand, increased

soil acidity has so far only been found in the organic horizon of podzols in severely damaged pine ecosystems near Severonikel (Chertov et al., 1993) and Pechenganikel (Chertov et al., 1990, 1993, Koptsik and Nedbaev, 1992, Koptsik and Muchina, 1995) within 4 km of sources. Investigations into the dynamics of a pine ecosystem near Monchegorsk did not reveal any tendency for increasing acidification over several years (Chertov et al., 1990). The absence of evidence of soil acidification within 7-40 km of Nickel was explained by the high buffering capacity of the soil and a high anthropogenic input of basic cations (Koptsik and Nedbaev, 1992).

Transect investigations in pine ecosystems carried out during the Finnish Forest Damage Project (Tikkanen and Niemelä, 1995) found no connection between the level of pollution and the parameters that characterise soil acidity (pH in water extraction, exchangeable H<sup>+</sup> and Al<sup>3+</sup>) within 10-70 km of Monchegorsk (Motova and Nikonov, 1993). At the same time, these authors describe an increasing content of amorphous Al in the illuvial horizon of podzols near the source of pollution.

During a detailed investigation of spruce ecosystems, lower acidity and higher content of exchangeable basic cations were found in the organic horizon of podzols located 7 and 20 km from the source of pollution, compared with control sites more than 100 km from pollution (Kashulina et al., 1995). In addition, lysimeter observations revealed a sharp increase (about 10 times) in the leaching of Al at sites 7 km from the pollution source (Jevtjugina, 1994). Räisanen et al. (1994) investigated changes in soil mineralogy connected with Al mobilisation at these sites.

The aim of the present paper is to provide data on the status of soil acidity and mobility of Al in podzols in the western part of the Kola Peninsula and neighbouring parts of Norway and Finland where the geological surveys of Finland (GTK) and Norway (NGU) and the Central Kola Expedition (CKE) in Russia are carrying out a major geochemical mapping project (see World Wide Web site <http://www.ngu.no/Kola>) in a 188,000 km<sup>2</sup> area between 24° and 35.5° E and north of the Arctic Circle to the Barents Sea (Fig. 8.1.1). As part of this project, eight catchments (hereafter abbreviated as C1 - C8) widely distributed in this area (Fig. 8.1.1) were investigated in detail in 1994. Media sampled were: snow (meltwater and filter residue), rainwater, stream water, organic stream sediments, terrestrial moss, topsoil (0-5 cm), complete podzol profiles, Quaternary deposits and bedrock. The advantage of this approach compared to transect studies is the availability of data from a large area in connection with up-to-date information about atmospheric element deposition and the natural background (differences in geology) from the same sampling sites.

## FIELD PROCEDURES

Criteria for the selection of sampling sites within the catchments as well as methods used for sampling are detailed in Äyräs and Reimann (1995). Two to four profiles were dug in most catchments (Table 8.1.1) and all major horizons (O, E, B1, ..., BC1, ..., C, ...) were sampled. The depth of the pits varied from 50 to 100 cm, depending on local conditions.

Most of the profiles investigated were haplic or ferric podzols with free drainage conditions. Two profiles (7P05 and 8P18 (the first figure denotes the catchment - see Table 8.1.1)) showed morphological evidence of temporary excess of water, and 1P38, 2P36 and 7P18 were close to the water table.

The parent material at most sites was till with a prevalence of sand fractions and a varying content of stones. Site 5P38 was on glaciofluvial sand. In the E horizon of 5P41, ablation till was mixed with weathering black schist.

Additional important information about the sampling sites is given in Table 8.1.1. All the Finnish catchments have an acidic type of bedrock - granite and quartzite. The other catchments have more basic bedrock, especially C2 with its gabbro and pyroxenite.

Some unusual profiles sampled include 5P41 (Table 8.1.1) where till is mixed with sulphide-rich saprolite in the E-horizon and 2P38 (Table 8.1.1) which is strongly eroded and lacks the humus and E-horizon. The other two profiles in C2 are in the initial stages of erosion. Only one profile was sampled in the Kirakka catchment because this catchment is dominated by outcrops of bedrock and till very rich in stones and large boulders.

The sampling sites also had different types of vegetation and the degree of ecosystem damage varied. Almost total damage of vegetation is characteristic of C2, quite serious vegetation damage was observed in C1 and C4, and slight damage in C3 and C5. C6 is influenced by reindeer grazing and tree felling. C7 and C8 in Finland show no traces of damage to vegetation or soil cover, except at site 8P18 where some spruce trees are dead and ground vegetation is slightly damaged.

Data on the airborne input of S and (Ni+Cu) at the different sampling sites, as calculated from analytical results of snow samples (Chekushin et al., 1996), are also included in Table 8.1.1 and show that all the catchments are to some degree affected by pollution, with the Finnish catchments displaying values that are characteristic for remote background sites worldwide.

#### ANALYTICAL METHODS

After arrival at the GTK laboratory all samples were dried at temperatures <40°C. The O-horizon samples were homogenised by milling in a domestic blender with blades made of non-contaminating material. The samples were then sieved to <2 mm.

For digestion, 0.5 g of humus was mixed with 10 ml of concentrated nitric acid, digested in a microwave oven and diluted to 50 ml with de-ionised water.

The mineral horizon samples were also sieved to <2 mm. 2.0 g of the <2 mm fraction were then mixed with 9 ml concentrated HCl and 3 ml concentrated HNO<sub>3</sub> (aqua regia) in a borosilicate tube. The mixture was left at room temperature overnight and then heated to 90°C for two hours in an aluminium block. The digest was diluted to 60 ml with de-ionised water, mixed thoroughly, decanted to polystyrene tubes and centrifuged.

In addition, ammonium acetate extraction buffered at pH 4.5 (shaking time 2 h), was carried out on all samples.

After centrifugation, the clear solutions were analysed with a Thermo Jarrell Ash Polyscan 61E ICP-AES apparatus for 30 elements. For the mineral soils (aqua regia extraction) and in the ammonium acetate extraction, As and Cd were also determined by graphite furnace AAS. For the O-horizon samples, where conc. HNO<sub>3</sub> was used for extraction, 29 elements were also determined by ICP-MS.

For more details on analytical techniques and quality control procedures, see Niskavaara (1995). The GTK laboratory is accredited according to ISO 9001 and ISO-Guide 25.

In addition, the total content of major elements in the mineral horizons was determined by XRF (fused beads) at NGU.

pH was determined in a water extract (2 g of soil in 20 ml of de-ionised water for the mineral soils, 2 g of sample and 40 ml of de-ionised water for the O-horizon samples) at NGU.

## RESULTS

Total element contents in the C-horizon

Table 8.1.2 summarises the analytical results (XRF) for the major elements in the C-horizon. Differences in lithology (Table 8.1.1) result in quite significant differences in the chemical composition of the parent material (till) between, and sometimes within, catchments (Table 8.1.2).

The greatest differences between catchments and sites were found for  $\text{Fe}_2\text{O}_3$ , MgO, CaO and  $\text{K}_2\text{O}$ . At about 10%, the sum of CaO and MgO in C2 is at least twice as high as in the Finnish catchments. This sum varies from 7 to 10% in C1, C4 and C5.

Alkalies show only slight variation between catchments and sites. Potassium contents are slightly elevated in the Finnish catchments. The differences in the  $\text{Al}_2\text{O}_3$  content can be quite significant, from 12% (all profiles in C8) to 17% (profile 7P18).

Surprisingly, the soil profiles in C3, where parts of the catchment contain alkaline rocks, show no peculiarities with regard to the chemical composition of the C-horizon. The Quaternary deposits sampled in this catchment show that till occupied most of the area in C3 has no geochemical peculiarities either. Only weathered sediments of Hibiny alkaline rocks which, are located in the upper part C3, are enriched in Na and Al (Chekushin et al., 1996).

The question remains whether or not the C-horizon still represents the natural geochemical background for this area. Even the chemical composition of a deep soil horizon may be altered by natural soil-forming processes and/or anthropogenic factors. For example, comparison of the chemical composition of 2P37, 2P38 and 5P41 with other profiles from the same catchments (Table 8.1.2) and with Quaternary deposit samples (Pavlov et al., 1996) allows us to assume that the low content of alkalies in the C-horizon of 2P37 and 2P38 and the low contents of K, Na, Ca, Mg and Al in 5P41 are related to anthropogenic factors in C2 and to an intensification of weathering and leaching of all these cations from the C-horizon due to natural factors in C5. An extremely high content of basic cations in C2 due to natural conditions (bedrock composition) may mask their leaching from the C-horizon in 2P37 and 2P38.

### *pH in water extract*

Figure 8.1.1 shows the pH values in a water extract for all profiles in the main horizons. For most of the profiles investigated, both the pH and its variation through the profile are quite typical for this type of soil.

### *C-horizon*

No correlation could be found between the pH measured in the C-horizon and major element contents or their molar ratios, nor with pollution as expressed in the aerial input of heavy metals or S (Table 8.1.1).

The pH of the C-horizon is weakly acidic in most profiles and varies from 5.7 to 6.4. The exceptions are 3P52 which gave a neutral reaction (pH 7.3) and 1P38, 2P36 and 5P40 which gave the most acidic reactions (pH 5.4, 5.6 and 5.5, respectively). However, when these extreme values are compared with overlying horizons (1P38 and 3P52) and neighbouring profiles (5P40 and 2P36), they can be explained by a specific aspect of the individual horizon (1P38, 3P52, 2P36), or the geochemistry of the whole profile (5P40).

Variation in pH in the C-horizon within one catchment is especially large between the profiles from C3, whereas pH is most constant and relatively high in all the C4 profiles.

On the whole, the parent material in the Finnish catchments (C6: granites, C7: gneisses, C8: quartzites) may be regarded as the most acidic. In contrast, the Russian catchments are characterised by very basic lithologies (Table 8.1.1). These major lithological differences should normally find their clear expression in the pH values measured in the C-horizon. That pH even in C2, which has the highest natural content of basic cations, does not differ significantly from the Finnish background catchments, may indicate that the C-horizon has also been affected by the industrial pollution.

### *O-horizon*

The organic horizon of podzols is usually characterised by acidic conditions with wide variations in pH, from 3 to 6 (Targuljan, 1971). In contrast, the variation in pH measured in the O-horizon in the eight catchments is surprisingly low. For most profiles investigated (14 of 22), the pH value in the water extract varies by only 0.2 pH units, from 4.3 to 4.5. There is no good correlation between the pH of the O-horizon and any of the other parameters studied here, including the degree of pollution.

The lowest pH value of all O-horizon samples was found in 2P37 (pH 3.9) and is no doubt related to pollution. The next lowest was observed in C6 (pH 4.0) and must have some natural cause. It is surprising to note that the pH of the O-horizon of all the other profiles from C2, every profile from C3 and C4, both of which are clearly influenced by pollution, and even C1, which receives the highest input of S of all the catchments (Table 8.1.1), does not differ from the pH results in the Finnish background catchments.

### *E-horizon*

The correlation between the pH of the E-horizon and other parameters studied here, including the degree of pollution, is poor. For most profiles (18 of 22), the pH of the E-horizon varies by only 0.3 pH units, from 4.8 to 5.1.

The lowest pH value was found in the E-horizon of 5P41. This, however, can be explained by *in situ* weathering of sulphide-rich black schist - a natural acidification factor.

Pollution, however, seems to have some influence on the pH of the E-horizon. All three profiles from catchment 2 have significantly lower pH values than any other profile.

### *B1-horizon, BC1-horizon*

The pollution level can be seen to strongly affect the pH of the B1-horizon. All the lowest pH values occur in C2 near Monchegorsk, despite the basic bedrock here. Lithology is seen to have a strong influence on the BC1-horizon, two of the three profiles in C2 showing rather high pH values. We can thus conclude that soil acidification in C2 has already completely destroyed the natural signature of the B-horizon, but not the BC1-horizon yet.

#### *Mobile cations: content and composition*

An interesting aspect of soil acidification is the interrelated mobilisation of Al and depletion of basic cations. To study these effects, we have used total analysis (XRF results for mineral soils and extraction in conc. HNO<sub>3</sub> for the organic horizon) compared with the results of ammonium acetate extraction (buffered at pH 4.5).

#### *Mobility of Al*

Figure 8.1.3 shows the mobile Al contents measured in the ammonium acetate extraction (bars - left-hand scale). These are also expressed as percentages of the total Al (dots and black line, right-hand scale). Up to 1% of the total Al is available in the C-horizon samples. In the BC1-horizon, up to 2.5% is mobile. In the B-horizon, it reaches 4%, but is only just over 0.4% in the E-horizon, whereas in the O-horizon up to 10% of the total Al is mobile (Fig. 8.1.3, right-hand scale). There is generally a close correlation between the total Al content and the mobile Al content.

In all the soil horizons, considerable variation in these parameters is evident from profile to profile, even within single catchments. This cannot be explained by pollution alone. Differences within the profiles can be large, too, and are due to differences between individual sampling sites.

In the most polluted catchment (C2, Monchegorsk), only profile 2P36 shows a high level of Al mobility in the O-, E- and BC1-horizons. The other two profiles from this catchment, in contrast, show some of the lowest contents of mobile Al, especially in their upper part. Parent material and the extent of pollution are very similar at these three sites. We propose that the differences observed here in the contents of mobile Al are caused by varying drainage conditions. Free surface and vertical drainage, in the case of 2P37 and 2P38, promote the leaching of mobilised Al from the profiles. At 2P36 the drainage is complicated by flat ground and close proximity to the water table.

Complicated drainage conditions can also help to explain the high content of mobile Al in some horizons of the profiles (1P38, 7P05, 8P13 and 8P18) (Fig. 8.1.3). All these profiles developed on flat ground and close to the water table.

In profile 8P18, which has one of the highest contents of Al and proportions of mobile Al in most of its horizons, the poor drainage conditions are not only related to relief and proximity to the water table, but also to the formation of an organo-mineral horizon (Ah) with a high clay content, impeding drainage. In combination with low contents of basic and alkaline cations, the high content of mobile Al may help to explain the damage to spruce trees observed at this site. The toxicity of the soil here for vegetation is also confirmed by the distribution of roots within the profile. The content of roots in the O-horizon is low and they are completely absent in the mineral horizons.

Indirect evidence of the importance of surface drainage for removal of mobile Al from the upper part of the soil may come from the chemistry of surface water during snow melting.

Despite the diluting effect of maximum discharge, a sharp increase in the Al content was observed during snow melting (while lower soil horizons were still frozen, preventing vertical flux) in both polluted and background catchments (Caritat et al., 1996b). Seasonal soil investigations have shown a sharp decrease in all mobile elements in upper soil horizons after snow melting (Levina, 1969).

Profiles from C3, especially 3P51 and 3P52, display the highest content of mobile (and total) Al in the illuvial horizon (B1), the reason for this being the proximity to the alkaline nepheline syenite intrusion. In addition, differences in the pH between eluvial (E) and illuvial (B1) horizons were especially great in this catchment (Fig. 8.1.2), creating a favourable situation for the accumulation of Al in B1.

Catchment 5 generally has the lowest content and proportion of mobile Al of all the catchments in almost all horizons. This is reflected in the low Al content in stream water here (Caritat et al., 1996a). Strong natural acidification owing to *in situ* weathering of sulphide-rich black schist in the eluvial horizon of profile 5P41 intensifies weathering throughout the profile and results in the removal of Al from the profile. This finds its expression in very low contents of mobile Al in this profile.

#### *Mobile cations composition*

The toxic effect of mobile Al on the vegetation depends upon its ratio to the basic cations (Sverdrup et al., 1992). Figure 8.1.4a shows this relationship between the Al and basic cation content in the main soil horizons of every profile. In addition, proportions are given for the O- and E-horizons (Fig. 8.1.4b).

At most sites investigated, the principal mobile cation in the O-horizon is Ca. Mg occupies the second position, K the third and Al the fourth. The proportion of Na is negligible (Fig. 8.1.4b). Drastic changes in the mobile cation composition as a result of pollution were found in the organic horizon of profiles 2P36 and 2P37. In 2P36, a high Al content and a reduction in the content of basic cations causes Al to become the dominant cation and total 50% of the sum. In 2P37, the content of all the mobile cations is very low (Fig. 8.1.4a) and there is a comparatively high content and, hence, proportion of mobile Al.

Occasionally, a high proportion of mobile Al was, however, observed at significantly less polluted sites. For example, in the organic horizon of 5P38, formed on glaciofluvial deposits, mobile Al reaches about 40%. In the organic horizon of profile 8P18 the mobile Al content is 32% in the upper part (O1, Fig. 8.1.4b) and reaches 66% in the lower part (O2, is not shown).

C3, with its special lithology, shows a quite peculiar composition of mobile cations in the organic horizons of the profiles. These profiles show some of the highest mobile Ca contents and, at the same time, rather low relative and absolute contents of Mg.

C1, with the highest input of atmospheric pollutants, is characterised by a low content and proportion of mobile Al and the highest content and proportion of mobile Mg in the organic horizon. Reimann et al. (1995a, b) described a significant input of Mg due to anthropogenic activities here.

Results obtained from 5P41, containing sulphide-rich black schists, did not show drastic changes in the proportions of the mobile cations. Profile 8P15 shows surprisingly high contents of mobile Ca and K and one of the highest contents of Mg in the O-horizon. All the parameters



which determine the nutrient status are very positive here, even though the profile is developed on quartzites (Table 8.1.1) which should result in rather unfavourable conditions. Possible explanations may be topography (foot of a hill with input of elements through runoff), or an intensive biological turnover.

The E-horizon contains considerably smaller amounts of mobile cations than the O-horizon (Fig. 8.1.4a). The principal mobile cation here is Al, followed by Ca and Mg (Fig. 8.1.4b).

In the C- and illuvial horizons the content of mobile basic cations is normally negligible and only Al remains mobile. This is a general feature of podzols and no anthropogenic activity can worsen this situation.

## CONCLUSIONS

It has been possible to show the importance of natural conditions (bedrock lithology and relief) on soil acidity and Al mobility in podzols by studying profiles from eight catchments characterised by different lithologies (natural background) and situated at different distances from some of Europe's largest point sources of SO<sub>2</sub> emissions (anthropogenic input).

The total element content (XRF) in the C-horizon varies considerably between the catchments and also within some of them. The chemical composition of even this deepest horizon can be affected by anthropogenic pollution, as observed in C2, or by natural acidification and subsequent leaching of cations due to sulphide-rich saprolite in one profiles (5P41). Generally, the area investigated is characterised by having the highest contents of basic cations in the parent material of the catchments closest to pollution sources and the lowest contents of basic cations in the parent material of profiles from background catchments. The natural geochemical conditions of the area thus tend to mask any acidification effects near the pollution sources.

Not even a long-lasting, very high input of S and heavy metals as is characteristic for C1 and C2, has led to detectable signs of soil acidification in the podzol profiles developed on till overlying basic lithologies. Moreover, strong degradation of the ecosystem, including soil erosion, seems to be necessary to give measurable effects in the O-, E- and B-horizons, as is the case in C2. However, even here, the O-horizon, which has a very high buffering capacity, shows only slight reaction to acidification, and the E-horizon is naturally leached. Of the upper horizons of the podzol profile, the illuvial horizon (B1) appears to be most sensitive to soil acidification.

Strong natural acidification, as observed at site 5P41 in C5, affected only the pH of the E-horizon containing the sulphide-rich material, and not the pH of any other soil horizon. Weathering of the lower soil horizons, however, is clearly affected. Vegetation is not damaged here. On the contrary, one of the best pine stands of the whole catchment is developed at this site.

In general, the content of mobile Al varies significantly between, and even within, catchments. Anthropogenic contamination plays a surprisingly minor role in the regional variation of mobile Al. Comparison of profiles 2P37 with 2P36 and 1P33 with 1P38 shows that even under very high input of anthropogenic S, high contents of mobile Al can only be found under very poor drainage conditions. Although acidification will mobilise Al and basic cations, these will be leached out of the profile under free drainage conditions.

Geogenic factors (as observed for all profiles in C3, and at sites 7P18, 7P19 and 5P40) and drainage conditions (sites 8P18, 2P36, 1P38), and not soil acidification, are thus the factors governing the availability of mobile Al in the podzol profiles. It is consequently not surprising that the highest content of mobile Al in all the profiles was measured at a background site in C8 (8P18) where drainage conditions were especially poor. The vegetation is clearly affected by Al poisoning because the natural content of basic cations (and all other nutrients) is low here, too.

Mobile Al generally makes up a rather large proportion of the total mobile cation pool in the E-horizon and can become the prevailing mobile cation here, independent of anthropogenic pollution levels. In the deeper soil horizons (illuvial layers, C-horizon), Al is always the predominant mobile cation and no pollution whatsoever has any influence on this situation.

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## TABLES

Table 8.1.1: Location and main characteristics of the podzol profile sampling sites. Information on the aerial input of S and (Cu+Ni) from Chekushin et al. (1996).

Table 8.1.2: Total element content (XRF analysis) of the C-horizon samples from all profiles (LoI corrected values).

## FIGURES

Fig. 8.1.1: Location of the survey area for the regional mapping project and of the catchments where the profiles discussed here were collected.

Fig. 8.1.2: pH of water extraction measured in all the major horizons of all the profiles.

Fig. 8.1.3: Content of mobile Al (ammonium acetate extraction) (left-hand scale, bars) in all the major horizons of the podzol profiles from all eight catchments (mg/kg of dry weight) and proportion of mobile Al to total Al (right-hand scale, black dot connected with lines) measured by XRF (mineral soil horizons) and conc. HNO<sub>3</sub> extraction (O-horizon).

Fig. 8.1.4a: Content of mobile cations measured by ammonium acetate extraction (expressed in meq/100g) in the major soil horizons of all the profiles.

Fig. 8.1.4b: Proportions of mobile cations in the O- and E-horizons of all the profiles.

Profile No	Location, m	Topography	Bedrock type <sup>1</sup>	Vegetation type	S load <sup>2</sup> , kg/km <sup>2</sup>	Ni+Cu load <sup>2</sup> , kg/km <sup>2</sup>
1P28	N: 7 708 000; E: 422 605	Top of moraine hill	Amphibole-biotite and granite gneisses	Dwarf shrub birch forest tundra	440	240
1P38	N: 7 707 250; E: 422 950	Flat, valley	-"	Dwarf shrub sparse forest tundra	460	220
2P36	N: 7 525 550; E: 495 100	Top of moraine hill	Dacite-andesite, tuffs	Empetrum dead standing spruce forest	310	510
2P37	N: 7 527 900; E: 495 600	Top of moraine hill	Norites, gabbro-norites	Technogenic barren with sparse birch shrubs	260	350
2P38	N: 7 529 650; E: 494 400	Gentle mountain slope	Pyroxenites	Technogenic barren	250	460
3P15	N: 7 493 400; E: 533 700	Mountain terrace, flat	Mafic tuffs	Dwarf shrub-green mosses birch forest with spruce	215	10
3P51	N: 7 495 350; E: 531 750	Mountain terrace, flat	Gabbro-norites	Dwarf shrub-green mosses birch mountain tundra forest	200	5
3P52	N: 7 495 350; E: 532 650	Mountain terrace	Gneisses, nephelin sienite	-"	190	3,5
4P06	N: 7 508 750; E: 493 050	Foot of hill, flat	Amphibolites	Dwarf shrub birch forest with spruce	80	32
4P38	N: 7 512 400; E: 493 550	Upper part of hill slope	Gabbro, gabbro-norites	Dwarf shrub sparse spruce forest	125	80
4P39	N: 7 511 900; E: 491 100	Valley, flat	Two-mica gneisses	Dwarf shrub spruce forest	125	50
5P37	N: 7 696 550; E: 597 200	Upper part of moraine hill	Amphibolitic gneiss	Green moss-dwarf shrub birch forest with pine	30	1
5P38	N: 7 697 700; E: 597000	Glaciofluvial delta; flat top	Andesitic sandstone	White lichen pine forest	30	1,2
5P39	N: 7 699 620; E: 597 550	Moraine hill; flat top	Mica schist	Green moss-dwarf shrub sparse pine forest with birch	40	1,2
5P40	N: 7 699 550; E: 596 390	Moraine hill; flat top	Andesitic laves, silt stone	Green moss-dwarf shrub birch forest with pine	45	1,2
5P41	N: 7 700 450; E: 600 100	Moraine hill; gentle slope	Andesitic laves, basalt	Green moss-dwarf shrub pine forest	45	1,3
6P12	N: 7 720 190; E: 454 380	Foot of moraine hill	Granite	Dwarf shrub-green moss pine forest with birch	33	0,5
7P05	N: 7 474 560; E: 602 500	Moraine plain	Granitoids	Bilberry-green moss sparse birch with spruce	70	0,5
7P18	N: 7 476 340; E: 606 040	Foot of moraine hill	-"	Bilberry-green moss spruce forest	80	0,5
7P19	N: 7 474 640; E: 605 100	Moraine hill, gentle slope	Granitoids	Dwarf shrub-green moss forest with pine and birch	100	0,5
8P13	N: 7 565 360; E: 370 180	Moraine plain, flat	Quartzite	Dwarf shrub-green moss spruce forest with birch and pine	70	0,5
8P15	N: 7 565 510; E: 368520	Foot of moraine hill	Quartzite	Grass-dwarf shrub-green moss spruce forest	60	0,5
8P18	N: 7 564 540; E: 371 020	Moraine plain, flat	Quartzite	Dwarf shrub-green moss sparse birch forest with spruce	60	0,5

1 - from geological maps (Chekushin et al., 1996)

2 - Winter 1993-1994 load (Chekushin et al., 1996)

Table 8.1.1: Location and main characteristics of the podzol profile sampling sites. Information on the aerial input of S and (Cu+Ni) from Chekushin et al. (1996).

Profile	SiO2	TiO2	Al2O3	Fe2O3	MnO	MgO	CaO	Na2O	K2O	P2O5	LOI
No											% of d.w.
1P28	67.4	0.73	13.7	5.7	0.08	2.4	4.2	3.5	1.8	0.14	1.5
1P38	63.2	0.96	14.3	8.3	0.11	3.4	4.6	3.1	1.5	0.14	4.3
2P36	62.2	0.68	15.1	7.3	0.11	4.2	5.3	3.1	1.3	0.12	2.7
2P37	60.6	0.75	14.3	8.2	0.12	5.7	5.9	2.7	1.2	0.13	1.9
2P38	60.2	0.64	15.6	7.6	0.11	5.7	5.9	2.7	1.1	0.11	2.0
3P15	69.7	0.47	13.9	4.2	0.07	2.1	4.1	3.7	1.6	0.11	1.3
3P51	65.4	0.89	13.7	6.6	0.13	2.8	4.4	3.4	1.6	0.11	1.8
3P52	64.8	0.89	14.8	6.2	0.14	2.6	4.1	3.8	2.2	0.12	3.0
4P06	60.2	0.78	13.6	9.5	0.14	5.1	5.0	3.1	1.9	0.14	2.3
4P38	67.0	0.67	14.0	5.7	0.09	2.5	4.6	3.6	1.6	0.18	1.3
4P39	63.7	0.83	13.9	7.6	0.11	3.5	5.0	3.2	1.7	0.17	1.8
5P37	62.7	0.68	14.8	8.6	0.10	3.0	4.9	3.1	1.1	0.18	1.9
5P38	66.8	0.54	14.8	5.5	0.08	2.5	3.9	3.3	1.8	0.13	1.2
5P39	65.4	0.75	14.5	6.8	0.10	2.5	4.4	3.3	1.4	0.17	1.2
5P40	62.8	1.03	15.2	9.0	0.14	3.5	4.3	3.1	1.4	0.17	2.0
5P41	68.3	0.68	13.1	8.6	0.09	1.7	3.2	3.1	1.1	0.11	2.6
6P12	73.1	0.32	13.3	2.2	0.04	0.8	1.8	3.6	3.5	0.12	0.8
7P05	73.3	0.42	13.2	2.9	0.05	1.6	2.7	3.6	1.6	0.07	1.6
7P18	66.0	0.50	17.3	5.1	0.04	2.5	2.9	3.6	2.0	0.11	4.2
7P19	68.2	0.56	15.9	4.5	0.05	2.2	2.8	3.6	1.7	0.11	3.7
8P13	72.7	0.43	12.4	3.3	0.05	1.5	2.7	3.4	2.6	0.09	0.9
8P15	72.1	0.48	12.6	4.2	0.09	1.8	2.7	3.0	2.5	0.09	2.4
8P18	73.4	0.46	12.0	3.4	0.05	1.6	2.5	3.0	2.6	0.09	1.1

Table 8.1.2: Total element content (XRF analysis) of the C-horizon samples from all profiles (LoI corrected values).



*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
Catchment locations

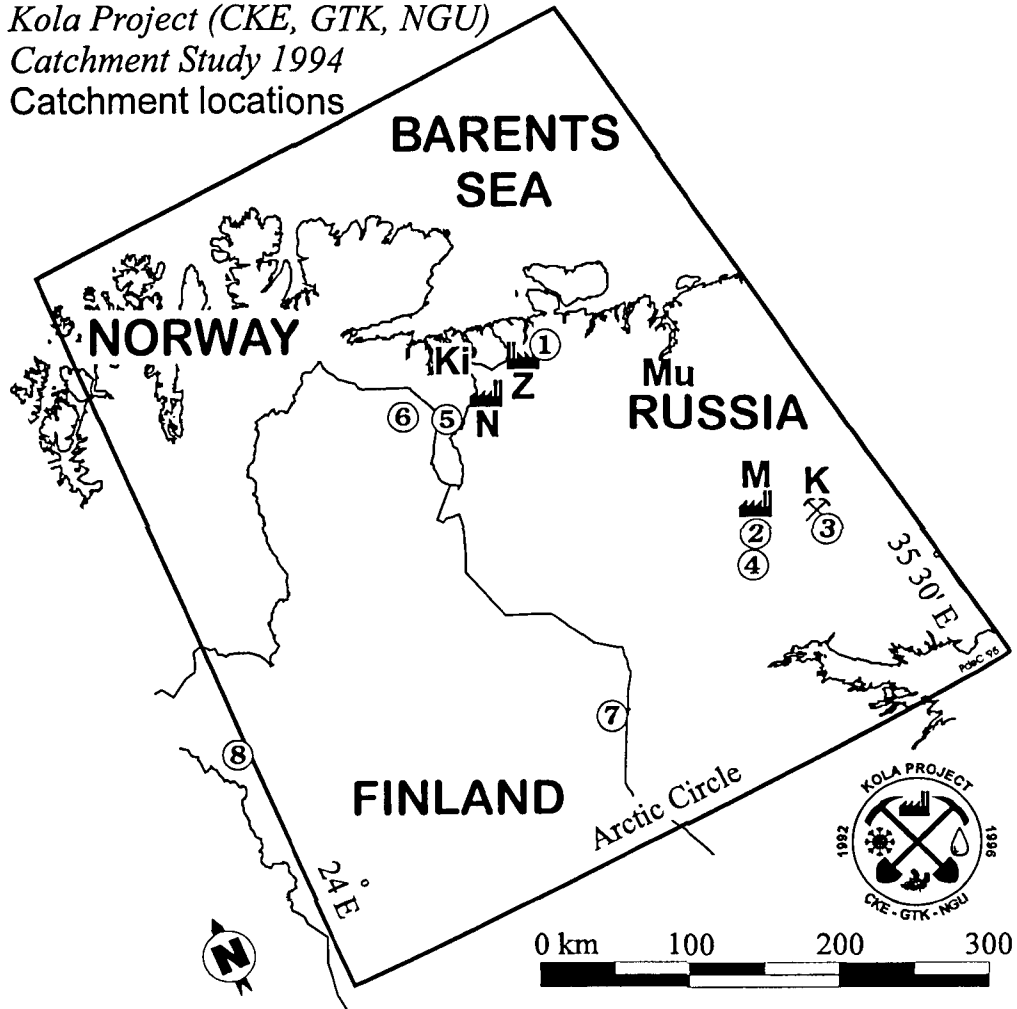


Fig. 8.1.1: Location of the survey area for the regional mapping project and of the catchments where the profiles discussed here were collected.

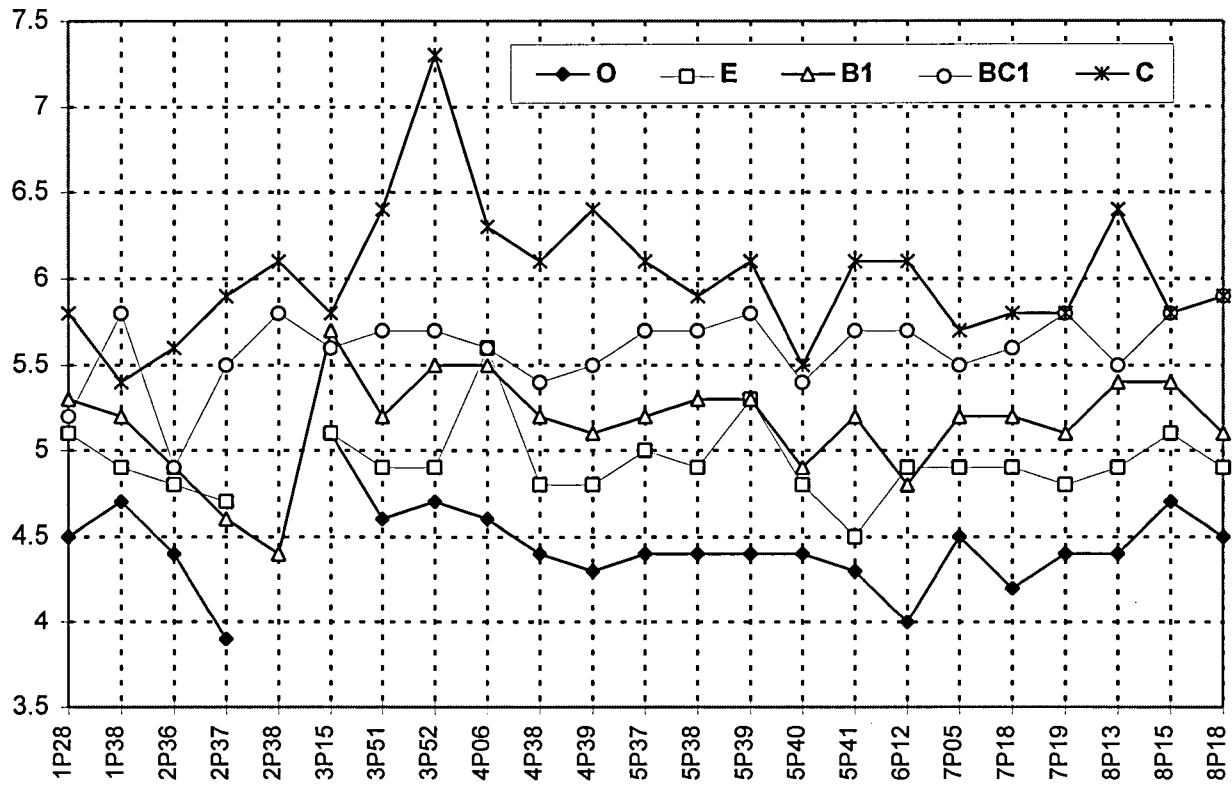


Fig. 8.1.2: pH of water extraction measured in all the major horizons of all the profiles.

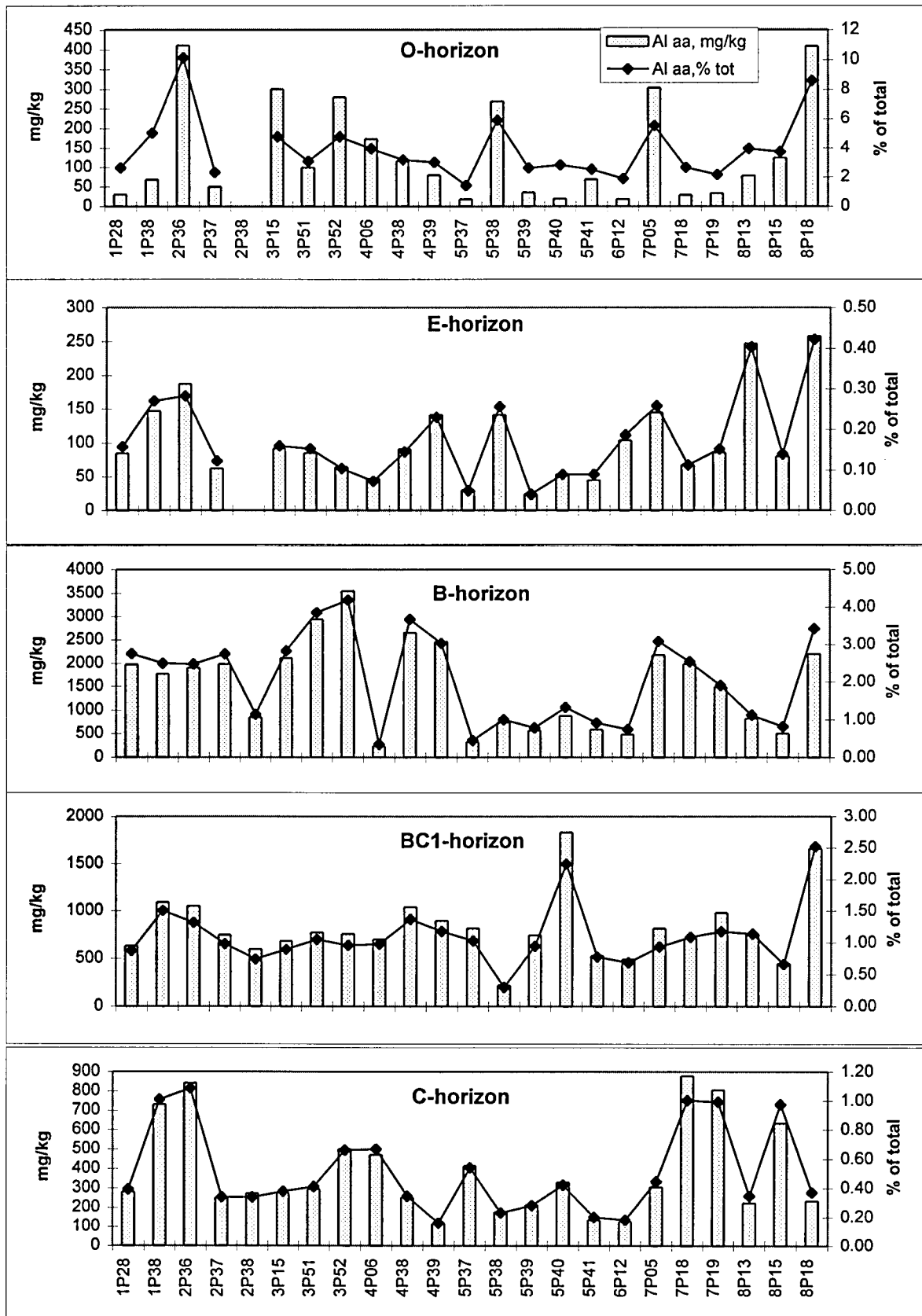


Fig. 8.1.3: Content of mobile Al (ammonium acetate extraction) (left-hand scale, bars) in all the major horizons of the podzol profiles from all eight catchments (mg/kg of dry weight) and proportion of mobile Al to total Al (right-hand scale, black dot connected with lines) measured by XRF (mineral soil horizons) and conc. HNO<sub>3</sub> extraction (O-horizon).

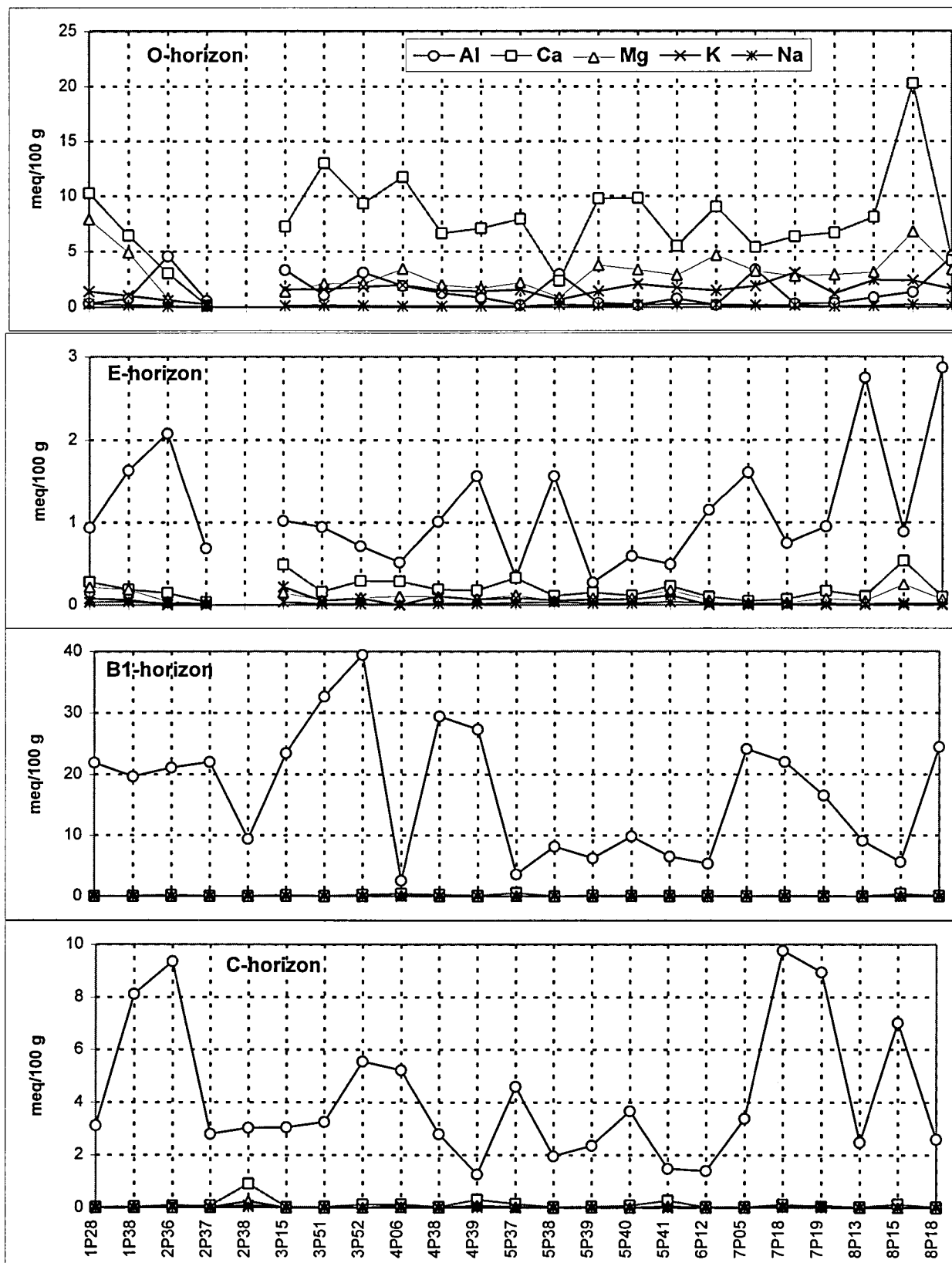


Fig. 8.1.4a: Content of mobile cations measured by ammonium acetate extraction (expressed in meq/100g) in the major soil horizons of all the profiles.

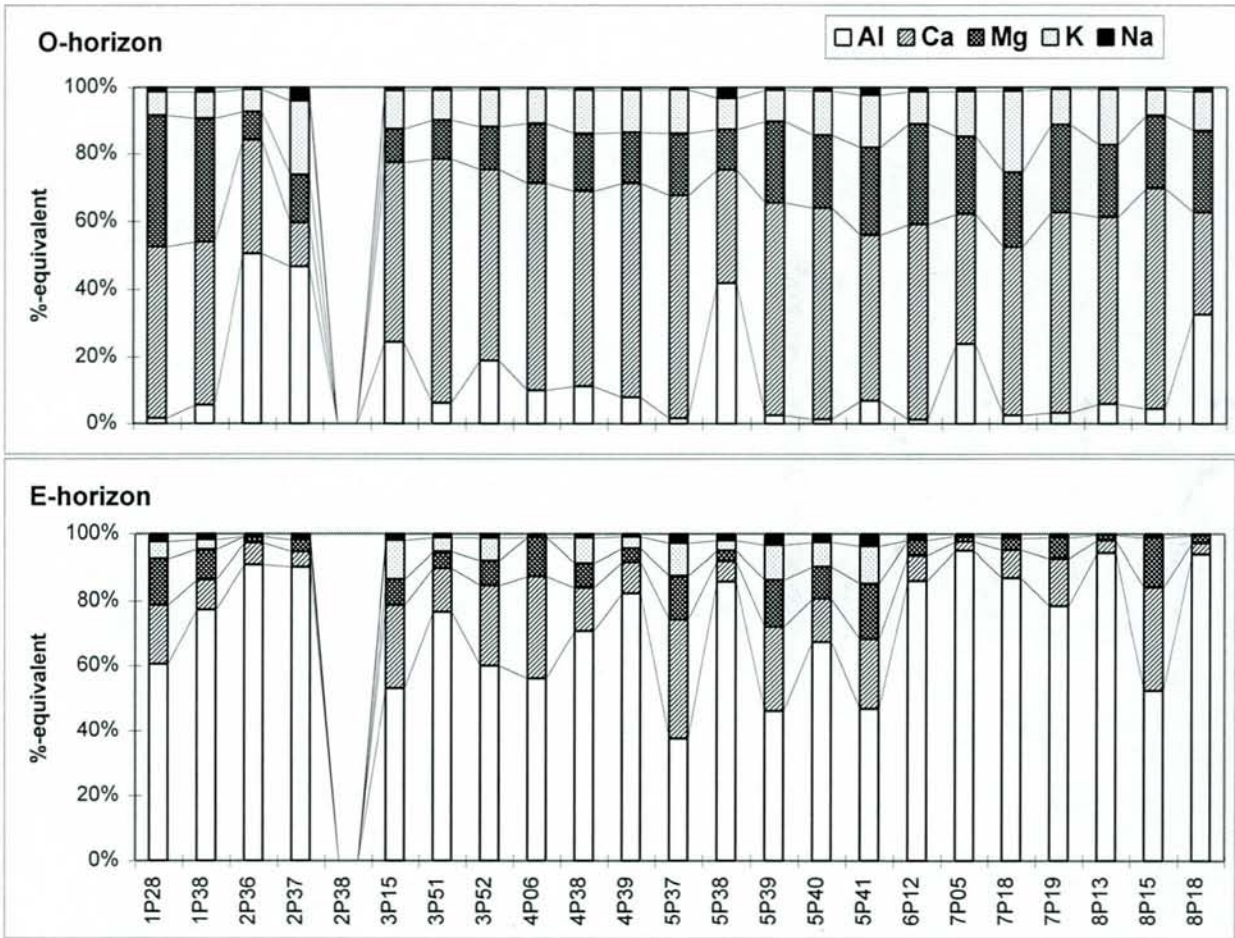


Fig. 8.1.4b: Proportions of mobile cations in the O- and E-horizons of all the profiles.

## 8.2 Mobility and retention of heavy metals, arsenic and sulphur in podzols at eight catchments northern Finland and Norway and the western half of the Russian Kola Peninsula

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### ABSTRACT

The mobility and retention of heavy metals, arsenic and sulphur in podzols from eight catchments located north of the arctic circle in Finland, Norway and Russia was determined by analysing the <2.0 fraction using an ammonium acetate pH 4.5 extraction in addition to a conc. nitric acid digestion for the humus samples, and a hot aqua regia digestion for the mineral soil samples.

Ni, Cu, Co and As were strongly enriched in the humus layer in the Russian catchments when compared to their concentrations in the parent tills and in podzols from Finland and Norway. By contrast, S was accumulated in the illuvial layer and not in the humus layer.

The behaviour of Ni and Cu within the profile showed that at the beginning both elements tend to accumulate in the humus layer. If they are mobilized there they leach down into the groundwater and are not retained by the illuvial layer. An exception was a strongly eroded profile near Monchegorsk, where Ni is tightly fixed in the illuvial layer while Cu and S are mobile.

### INTRODUCTION

Podzols and podzolic soils are the main pedon type in the forest zones in the northern arctic areas. Usually they are developed on tills, which were deposited by the glacier some 10000 years ago (Niemelä et al. 1993). The till consists mostly of fresh (unweathered) rock material mechanically comminuted by glacial erosion. As minerals are enriched to their terminal grades, i.e. minerals are ground to their primary grain size in rocks during the glacial transport (Dreimanis & Vagners 1971), the fine fraction is dominated by Mg and Fe bearing silicates and sulphides and the coarse fraction (>0.06 mm) by quartz and feldspars (Nikkarinen et al. 1984, Mäkinen 1995). Owing to this, the fine fraction (<0.06 mm) is usually enriched by heavy metals and sulphur (Mäkinen 1995, Tarvainen 1995).

Due to the young age of podzols in the northern Europe (<9000 years) their total content of trace elements and sulphur is still primarily related to the nature of the rocks providing the parent material for soil formation. Here the average age corresponds to the dates (9100 B.P., 9800 B.P.) of the ice retreat in Northern Europe (see Johansson 1995) minus the years of differentiation of podzolic layers (300-1200 years, Petäjä-Ronkainen et al. 1992). Due to pedogenic processes, elements are variably differentiated with depth within the podzol profile. Some of the elements are mobilized and taken up by vegetations leading to an accumulation in the organic layer, others are leached out of the upper soil layers and precipitated in the illuviated layers, or washed out into the groundwater (Berrow & Mitchell 1980).

A typical podzol profile is characterized by a poorly decomposed organic layer (O, humus), a light eluvial (E) layer, a brown illuvial (B1, B2) layers which are considerably enriched in

amorphous Fe, Al, Si and organic matter (leached from the E layer), and the yellowish grey or greenish grey lower layers of less altered parent till (BC1, BC2..., C1...). As Fe and Mn hydroxides have a high sorption capacity for trace metals (Coughlin & Stone 1995) it could be expected that the illuvial layer retains as well airborne as native heavy metals when they are leached from the surface layer (Kabata-Pendias & Pendias 1992). Kabata-Pendias and Pendias (1992) have nevertheless pointed out that the humus layer is the most significant sink for trace elements in polluted soils. Also they conclude that the origin of the trace elements in soils (natural or anthropogenic) has an influence on their behaviour within the profile and on their bioavailability. Räisänen & Hämäläinen (1991) and Räisänen et al. (1993) suggest that abundant mobile heavy metals and arsenic in the surface (O, E) soil layers is indicative of anthropogenic contamination.

The Geological Surveys of Finland (GSF) and Norway (NGU) and the Central Kola Expedition (CKE) in Russia are carrying out a major geochemical mapping project (see World Wide Web site <http://www.ngu.no/Kola>) in a 188,000 km<sup>2</sup> area north of the Arctic Circle, comprising the entire area between 24° and 35.5° E north to the Barents Sea (Fig. 8.2.1). As one part of the project, element input and circling was monitored in more detailed in eight catchments in 1994 (Fig. 8.2.1, Table 8.2.1). Media sampled were: snow (meltwater and filter residue), rainwater, stream water, organic stream sediments, terrestrial moss, topsoil (0-5 cm), complete podzol profiles, Quaternary deposits and bedrock. This paper deals with the podzol profile data. Here the main target is to examine the mobility and retention of heavy metals (Co, Cr, Cu, Mn, Ni, Pb, V and Zn), arsenic (As) and sulphur (S), within podzol profiles located in the catchments with different distances to pollution from the Russian Ni-Cu mining and smelting industry (Fig. 8.2.1). The annual emissions from the Ni-Cu smelter complex at Nikel and at Monchegorsk and the ore roasting plant at Zapoljarnij, are about 296000 tonnes of SO<sub>2</sub>, 1700 tonnes of NO<sub>2</sub>, 1900 tonnes of Ni, 1100 tonnes of Cu, 94 tonnes of V<sub>2</sub>O<sub>5</sub>, and 92 tonnes of Co in 1994 (Report of the Mormansk Region Committee of Ecology and Nature resources 1995).

## MATERIALS AND METHODS

### *Sampling*

The criteria for the selection of the sampling sites within a catchment and the method used for sampling are reported in detail by Räisänen & Kashulina (1995). A short description of landscape, topography, glacial deposits and podzol types at each sampling site is presented in Table 8.2.2 (see also Table 8.2.1).

Horizon samples (O, E, B1, ..., BC1, BC2, ..., C1, ...) of the podzol were taken from the spade-dug pit. Depth of the pits varied from 50 cm to 100 cm depending on the thickness of the podzolic (E, B1, B2) layers and the location of less altered parent till in the bottom of the pit. Some unusual profiles sampled include (Table 8.2.2): the profile 5P41 (Table 8.8.2) in the Skjellbekken catchment was till mixed with sulphide-rich saprolite and the profile 2P38 in the Monchegorsk catchment was strongly eroded and lacked the humus and eluvial layers. The other two profiles in the Monchegorsk catchment were sampled at sites where trees and ground vegetation were partially destroyed and humus was lacking in places. In the Kirakka catchment, only one profile was sampled due to the fact that this catchment is dominated by outcrops of bedrock and till rich in stones and boulders (Table 8.2.1).

### *Chemical analysis*

After arrival at the GSF Lab all samples were dried at a temperature  $<40^{\circ}$ . Mineral soil samples were sieved to  $<2.0$ mm. Before sieving to  $<2.0$  mm, humus samples were homogenized by milling with a domestic blender with contamination-free blades.

The buffered ammonium acetate pH 4.5 method was here used for determining the element concentration in the easily leachable (mobile) fraction of the humus and mineral soil samples (Räisänen & Hämäläinen 1991). A 6 g subsample was extracted in a horizontal shaker for two hours with 30 ml of 1.0 M ammonium acetate solution buffered with acetic acid to pH 4.5. After filtration through a 0.45  $\mu$ m membrane filter, solutions were analysed with ICP-AES for 26 elements and with a graphite furnace -AAS for As and Cd.

In addition, mineral soil samples were analysed with the hot aqua regia method and humus samples with the conc. nitric acid method (Niskavaara 1995). For the digestion of the mineral soil sample, 2.0 g of the  $< 2$  mm fraction was mixed with 9 ml concentrated HCl and 3 ml concentrated HNO<sub>3</sub> (aqua regia) in a borosilicate tube. The mixture was left at room temperature over night and then heated to  $90^{\circ}\text{C}$  for two hours in an aluminium block. The digest was diluted to 60 ml with deionized water, mixed thoroughly, decanted to polystyrene tubes and centrifuged. The clear solution was analysed with ICP-AES for 31 elements. In addition, As, Cd and Pb were determined with a graphite furnace -AAS.

For the digestion of the humus sample, 0.5g of humus was mixed with 10ml of concentrated nitric acid, digested in a microwave oven and diluted to 50 ml with deionized water (Niskavaara 1995). The clear solution was determined with ICP-AES for 30 elements and with ICP-MS for As, Cd, Co, Cr, Ni, Pb and V.

Element concentrations determined with the conc. HNO<sub>3</sub> digestion method can be considered as total element contents as long as the humus does not contain mineral fragments. Element concentrations in the hot aqua regia leaches are not precisely total concentrations, since all minerals are not decomposed during the digestion (Fletcher 1981). For the purpose of this study, however, results obtained for heavy metals, As and S in the aqua regia extraction, are considered as total concentrations. This may be justified due to the fact that hot aqua regia totally decomposes sulphides, trioctahedral micas, clay minerals and amorphous compounds, which are the major sources of these elements in tills (Räisänen et al. 1992).

## RESULTS AND DISCUSSION

For data processing, soil samples of the profile were grouped into a humus layer, an eluvial (E) layer, two illuvial (B1, B2) layers, and an upper transitional zone (BC1) layer and parent till (C). As concentrations of heavy metals, As and S did not markedly vary in the lower layers below the BC1 layer, samples from the lower transitional zone (BC2,...C1,...) layers were here treated as the C layer or parent till. To facilitate easy comparisons between the eight catchments, mean values were calculated for the different layers of all samples from each catchment. These are presented in Tables 8.2.3 (humus layer) and 4 (mineral soil layers). The two abnormal profiles mentioned above (2P38 and 5P41) were not used for the mean calculations.



## *Distribution of heavy metals, As and S within the profile*

### *Total concentrations of the elements in humus*

Table 8.2.3a shows that means of the total concentrations of Ni and Cu were several hundred times greater in the catchment of Monchegorsk than in the Finnish catchments. In the catchments of Kurka and Zapoljarnij, they are ten to a hundred times greater and in the Kirovsk and Skjellbekken catchments, which are situated about 40 km downwind from the pollution sources, they are four to six times greater than in the background area in Finland. The rates of contamination is consistent with the difference in the metal load at these sites. The same pattern was observed for many of the media sampled for the catchment study (snow: Äyräs et al. 1995, rain: Reimann et al. 1996a, streamwater: Caritat et al. 1996, topsoil 0-5cm: Reimann et al. 1996b, moss: Äyräs et al. 1996).

Cobalt, Cr, As and V showed a similar behaviour with considerably lower contrast (Table 8.2.3a). The highest total concentrations were measured in the catchment of Monchegorsk and the lowest in the Norwegian and Finnish catchments. For S, Cd, Mn, Pb and Zn, podzol profile sampling did not reveal any significant correlation between the element concentration in the humus layer and distance to industry, even though these elements are part of the emission spectrum of the Russian mining and smelting industry (Äyräs et al. 1995 and 1996). Noteworthy is the low total concentrations of S in humus near the Ni-Cu smelters, even though total annual emissions of SO<sub>2</sub> were 300 times as high as those of Ni and Cu. In the earlier research from the surroundings of Nikel and Zapoljarnij, Niskavaara et al. (1996) conclude that humus samples can not be used to outline the anthropogenic input of S in the forested area, but they reveal well the contamination of Ni and Cu. A similar finding was also reported by Nikonov (1993).

### *The element concentrations in the mobile fraction of the humus layer*

As seen in Table 8.2.3b, mean concentrations of the easily leachable Ni, Cu, Co, As, V and Cr revealed the similar dispersal pattern as did the means of their total concentrations in Table 8.2.3a, while concentrations of S, Cd, Mn, Pb and Zn was not influenced by the emissions of the smelters at Monchegorsk and the ore roasting plant at Zapoljarnij. It is pointed out here that in contrast to the total concentrations, the mean value of the easily leachable Ni in Table 8.2.3b was 200 times greater, while that of Cu was 4000 times greater in Monchegorsk than in the background areas in Finland. The difference in means of Co, As, Cr and V was 10-20 times. Moreover, the mean concentration of the easily leachable Cu, but not Ni, was abnormally elevated in Kurka compared to those in Zapoljarnij and in the other catchments.

### *Total concentrations of the elements in the mineral soil layers*

It is here assumed that the concentrations of the metals, As and S in the C layer, i.e. the less altered transitional zone layers and parent till, represent the "natural background" levels derived by geology of an area. As Table 8.2.4a shows, means of the total concentrations of Ni, Cu and Cr were greater in tills on mafic bedrock (Monchegorsk and Kurka) than on tills on granite or quartzite bedrock (Kirakka and Pallas). Expectedly, total concentrations of S were greater in tills in Monchegorsk and Skjellbekken, where the mineralizations of Ni-Cu sulphides were exposed as outcrops of bedrock under the till cover. In Zapoljarnij, the moderately high concentrations of S (Table 8.2.4a) may be due to rock material derived from the Pasvic-Pechanga greenstone belt or an unknown mineralisation in bedrock.

Nevertheless, the total concentrations in the C layer between the catchments on mafic and acid bedrock differed less markedly than one could expect. Evidently, this is due to the coarse grain size (>2.0 mm) of the samples, which was selected here to follow the international soil scientific concentrations. Räisänen et al. (1995) report that the composition of till is more granitic in the <2.0 mm than <0.06 mm fraction on the mafic bedrock. They also conclude that geochemical differences between the sandy and silty tills are less detectable in the <2.0 mm than in the <0.06 mm fraction (see also Tarvainen 1995). It is a known fact that regional differences in till geochemistry are best outlined in the fine fraction (Nikkarinen et al. 1984, Räisänen et al. 1992, Mäkinen 1995, Tarvainen 1995).

Table 8.2.4a also shows that differences in metal concentrations between the C layer and the overlying illuviated (B1, B2, BC1) layers were in most cases surprisingly small compared to the difference between the eluvial layer and the illuvial layers. This must again be attributed to the coarse grain size fraction analysed.

As seen in Table 8.2.4a, As, Co, Cr, Cu, Mn, Ni, V and Zn generally show the lowest mean concentrations in the eluvial layer. The exception of this exhibited in the Monchegorsk catchment, where Cu and As were markedly enriched in the eluvial layer. Cobalt (except in the Monchegorsk catchment), Cu, Mn (except in the Kirovsk and Naruska catchments) and Ni show a tendency to increasing element levels with increasing depth. Mean concentrations of Cr and V were at maximum either in the upper or lower illuvial layer (B1, B2) while means of Mn, Zn and As show no general pattern. Mean concentrations of Pb are highest in the eluvial layer, an exception being the samples from the Kirakka and Naruska catchments where the greatest concentrations of Pb are exhibited in the B1 layer. The contrast in the mean Pb concentrations between eluvial layer and the uppermost illuvial layer was unusually big in the Kirovsk catchment, where humus was not characterized by abnormal concentrations of Pb.

Sulphur behaved somewhat differently to the heavy metals and As in the mineral soil layers (Table 8.2.4a). The lowest mean values occur in the eluvial layer and the highest immediately beneath in the B1 layer from which the concentrations decreased downwards in the parent till. This pattern is, however, more distinct here than that for the heavy metals and As. The highest mean concentration of S was obtained in the illuviated layers (B1, B2, BC1) of the Monchegorsk catchment where also the contrast in the concentrations between the parent till and the B1 layer is greatest.

Unusual element concentrations exhibited in the following profiles: the profile 1P38 from Zapoljarnij contained 126 mg/kg Cu and 6 mg/kg As and the two profiles (5P37 and 5P39) from Skjellbekken contained 6-7 mg/kg As in the parent till. However, the concentrations could not be followed with the similar levels upwards in the profile and the elements must therefore be derived from a mineralisation in bedrock.

#### *The element concentrations in the mobile fraction of the mineral soil layers*

The comparison of Table 8.2.4b with Table 8.2.4a shows that the distribution of easily leachable elements follows in many ways the above described patterns for the aqua regia extraction. Mean values for As and Cd were not reported in Table 8.2.4b because the concentrations obtained were very close to or below the detection limits. Abnormally high concentrations of the easily leachable Cu, Ni and S were measured only from the profiles of the Monchegorsk, Zapoljarnij and Kurka catchments which have the highest input of these elements. Mean concentrations of easily leachable Co, Cr, Mn, Pb, V and Zn were low and

varied from catchment to catchment. The eluvial layer of the Kirovsk catchment, however, exhibited an unusually high concentration of easily leachable Pb.

It is emphasized here that in the Monchegorsk and Kurka catchments, mean concentrations of easily leachable Cu and Ni was distinctly higher in the eluvial (E) and illuvial (B1, B2) layers than in the parent till (C). Furthermore, in Monchegorsk Cu was, on an average, more leachable than Ni in the eluvial layer, but not in the underlying layers.

Mean concentrations of easily leachable S were almost ten times greater in the illuvial (B1, B2) layers than in the parent till in the Monchegorsk, Kurka and Zapoljarnij-catchments, while in all other catchments the difference was at most fivefold.

#### *Mobility and retention of heavy metals, As and S*

Räisänen et al. (1993) suggest that polluted soils and sediments can be recognised by an increase in the element concentration of the mobile fraction. This can be demonstrated also here. Fig. 8.2.2 shows an example of the distribution of total and easily leachable S in two selected podzol profiles. Fig. 8.2.2a shows the results from the profile 5P41 from Skjellbekken where till was mixed with saprolite, and Fig. 8.2.2b the results from the strongly eroded profile (2P38) at Monchegorsk. The comparison of the S distributions within the profiles shows that, despite of the high total concentrations of S, its leachability in the NH<sub>4</sub> acetate was very low in the undisturbed (natural) profile at Skjellbekken (Fig. 8.2.2a). In the disturbed profile at Monchegorsk (Fig. 8.2.2b), a large portion of S was in easily leachable form in the upper (B1, B2) soil layers. Copper and Ni behaved quite similarly in the profiles near the industry.

Fig. 8.2.3 compares the mobility of Cu, Ni and S within the different soil layers of podzol profiles from the eight catchments. Here the mobility of the element was calculated as a proportion (%) of the mean concentrations of the easily leachable element from its mean total concentration. The catchments were sorted from close to industry at the left hand side to background at the right hand side of the diagram. While the mobility of Cu and Ni was highest in the eluvial layer and in some cases in the humus, S showed the opposite behaviour. As also seen in Fig. 8.2.3, the mobility of Cu and Ni increased substantially from background profiles to profiles close to industry. The exception was the profile from the Kirovsk catchment with its very special bedrock lithology. However, differences in the mobility of S between the profiles near the industry and in the background were not that pronounced. Mobilities of all other elements investigated did not exhibit any distinct trend.

Fig. 8.2.4a shows the average accumulation of Cu, Ni and S in the humus layer and Fig. 8.2.4 their average accumulation in the upper illuvial (B1) layer in the eight catchments. The accumulation was calculated as a ratio of the mean total concentration in the humus or illuvial layer to that in the C layer. Humus retained distinctly more metals, but not S in the catchments near the emission sources than in background. Similar results are also reported from a Finnish-Russian transect study in the Lapland Forest Damage Project by Nikonov (1993). We argue that the accumulation of S in the humus layer is associated to microbial activity being greater in the background area than in the strongly polluted areas (Derome et al. 1995). By contrast, the considerable accumulation of metals in the humus layer is due to the contamination.

Compared to the humus layer, the accumulation of the metals and S in the upper illuvial (B1) layer was low (Fig. 8.2.4b). For Cu and Ni there was hardly any difference between the background catchments and the most contaminated catchment near Monchegorsk. Sulphur

was, however, slightly more enriched in the illuvial layer of the Kurka and Monchegorsk catchments than in that in the background catchments.

The above-mentioned findings are in contrast to those of Saur and Juste (1994) who report that long-range transported metallic aerosol lead to highly elevated contents of Cr, Cu, Ni, Pb and Zn in the illuvial layers. In the present study, however, retention of metals in the illuvial layer was almost the same in the background catchments as in the immediate vicinity of the emission sources (Fig. 8.2.4b). This suggests that it is rather associated to podzolization than to the anthropogenic contamination.

Also Kabata-Pendias & Pendias (1992) suggest that the precipitation of metals in the illuvial layer is characteristic of podzols. However, they conclude that the accumulation of metals in the organic rich surface soil layers is a typical feature of polluted soils. The latter feature characterizes the catchments near industry in this study.

The behaviour of Cu, Ni and S within the profile as seen in Figs. 8.2.3 and 8.2.4 suggests that in soils near the emission sources, the metals, when mobilized, percolate throughout the podzol towards the groundwater. The retention of the metals in the deeper till layer is delimited by the small amount of exchangeable clay minerals (Räisänen et al. 1994). By contrast, S remains in the illuvial layers; to a large extent, however, in its mobile form. This may disturb the stability of Al-rich compounds here (Räisänen et al. 1994).

An exception is the behaviour of Ni at the strongly eroded site (2P38) near Monchegorsk. Fig. 8.2.5a shows the different behaviour of total and easily leachable Cu and Ni in this profile. Ni was very fast bound in the upper (B1) layer at the top of the profile, whereas almost half of the total Cu content was in the easily leachable form. Total concentrations of both Cu and Ni were remarkably lower in the B2 layer and in the underlying mineral soil layers. In the B1 layer Ni is either tightly fixed by Fe-oxides, a mechanism suggested by Kabata-Pendias and Pendias (1992) and Coughlin and Stone (1995), or there exists a marked difference in the mineralogy of the deposited Ni and Cu emissions. As Cu remains partly in its mobile form, it moves through the soil profile at barren sites. The earlier study of Niskavaara et al. (1996), however, interprets that Cu is much stronger bound than Ni when the organic layer is intact.

In summary, accumulation and mobility of Cu and Ni within the profile differ substantially between the Monchegorsk and Zapoljarnij catchments. This may be due to the different nature of the emissions coming from the smelter (Monchegorsk) and the ore roasting plant (Zapoljarnij), as seen in snow samples by Äyräs et al. (1995).

## CONCLUSION

Total concentrations of Ni, Cu, Co, Cr, As and V were much higher in the Monchegorsk, Kurka and Zapoljarnij catchments than in all other catchments. These elements remained strongly enriched here when compared to the composition of the parent till in the different catchments, thus proving an anthropogenic source.

The < 2 mm fraction as generally used in soil sciences proved to be less suitable for the study of the behaviour of heavy metals in mineral soils. In the coarse fraction even tills derived from ultrabasic bedrock tend to display a generally granitic composition due to the relative enrichment of quartz and feldspar during the glacial comminution.

The mobility of heavy metals in the surface layers and that of S in the illuvial layer was much higher in catchments close to industry than in the background catchments. Slightly elevated concentrations of mobile Cu and Ni in Skjellbekken and of mobile Cu in Kirakka compared to those in Naruska and Pallas, showed that also the soils of these two catchments in Norway and Finland were influenced by contamination.

The behaviour of Cu and Ni within the profiles showed that at first both elements tend to accumulate in the organic layer. If mobilised here they are not retained by the illuvial layers but percolate through the profile to the groundwater. By contrast, S is enriched in the illuvial layers. Here a large portion of S remains in mobile form and may disturb the stability of Al-compounds.

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## TABLES

Table 8.2.1. Overview of the main characteristics of eight catchments.

Table 8.2.2. Podzol profiles and characteristics of the sampling sites. Soil classification was based on the FAO Soil Classification (FAO-Unesco 1980).

Table 8.2.3. Means of a) the total concentrations of heavy metals, As and S and b) the concentrations of the easily leachable heavy metals, As and S in the humus (O) layer in eight catchments. Keys for the catchments: Mo = Monchegorsk, Zap =Zapoljarnij, Kur = Kurka, Kv = Kirovsk, Skj = Skjellbekken, Kir = Kirakka, Nar = Naruska and Pal = Pallas.

Table 8.2.4. Means of a) the total concentrations of heavy metals, As and S and b) the concentrations of easily leachable heavy metals and S in the mineral soil layers in eight catchments. See keys for the soil layers in the text and for the catchments in Table 3.

## FIGURES

Fig. 8.2.1. Location of eight catchments in northern Finland and Norway and in the western half of the Kola Peninsula, Russia.

Fig. 8.2.2. Distribution of total S concentrations (black histograms) and concentrations of the easily leachable S (white histograms) in a) the profile 5P41 in Skjellbekken and b) the strongly eroded profile 2P38 in Monchegorsk.

Fig. 8.2.3. Mobilities (%) of Cu (dashed line), Ni (thick line) and S (dotted line) within the profile in eight catchments. The mobility of the element was calculated as a proportion (%) of the concentration of easily leachable element from its total concentration. See keys for the catchments in Table 3a and for soil layers in the text.

Fig. 8.2.4. Accumulation of Cu (dashed line), Ni (thick line) and S (dotted line) into a) the humus layer and b) the upper illuvial (B1) layer in eight catchments. Accumulation was calculated as a ratio of the total concentration of the element in the humus layer to that in the parent till (C layer). See keys for catchments in Table 3.

Fig. 8.2.5. Distribution of a) total Ni concentrations and concentrations of easily leachable Ni, and b) total Cu concentrations and concentrations of easily leachable Cu in the strongly eroded profile 2P38 in Monchegorsk. See keys for the soil layer in the text.



Catchment (No.)	Coordinates of catchment outlet	Size (km <sup>2</sup> )	Elevation (m a.s.l.)	Annual precip. (mm)*	Vegetation	Bedrock	Quaternary Deposits
Russia							
Zapoljamij (C1)	69°27'01"N 31°03'49"E	19.0	25-373	454	Tundra forest (birch)	amphibole-biotite gneiss >> granites	basal till (60%) >> meltout till (26%) >> eluvial dep. (7%) >> rock terrains. water
Monchegorsk (C2)	67°50'30"N 32°54'48"E	22.4	128-507	391	Mostly barren (originally boreal conifer forest: spruce & birch)	andesitic effusives, tuffs > gabbros, gabbro-norite > pyroxenite	basal till (76%) >> glaciofluvial sed. (9%) > eluvial dep. (8%) >> glacio-lacustrine sed. (4%) > water, lacustrine sed. rock terrain
Kirovsk (C3)	67°32'50"N 33°48'55"E	20.0	240-1075	502	Mountain: birch >> spruce	Trachytoid>hibinite >> gabbro-norite > metabasalts & meta-andesites > black schists. mafic tuffs	basal till (57%) > deluvial - eluvial dep. (41.4%) >> rock terrain
Kurka (C4)	67°41'25"N 32°50'14"E	20.5	152-466	502	Boreal conifer forest: spruce > birch	amphibolite > two-mica gneiss > gabbro-norite > gabbro-amphibolite	basal till (54%) > peat (30%) >> glaciofluvial sed. (6%) > lacustrine sed. (5%) > rock terrain. water
Norway							
Skjellbekken (C5)	69°21'25"N 29°27'25"E	34.6	80-297	422	Boreal conifer forest: Pine >> spruce & birch	andesites, andesitic volcanoclastic schists (40%) > tholeiitic basalts & tuffs (30%) > black schists (20%) > others	basal till (59.2%) >> glaciofluvial sed. (17%) > meltout till (12.4%) >> rock terrain, water
Finland							
Kirakka (C6)	69°35'12"N 28°51'46"E	11.9	110-200	386	Tundra forest (pine & birch)	granite (100%)	rock terrain, block fields (60%) > meltout till (34%) > water
Naruska (C7)	67°21'44"N 29°22'05"E	20.2	263-490	513	Boreal conifer forest: spruce & pine > birch	tonalitic gneiss (mica gneiss, amphibolite interlayers)	basal till (63%) >> peat (22%) > rock terrain, block fields (14%) > water
Pallas (C8)	68°09'14"N 23°52'50"E	24.4	303-500	405	Boreal conifer forest: spruce & pine	quartzite >> amphibolites	basal till (69%) >> peat (29%) >> rock terrain (2%) > water

Table 8.2.1. Overview of the main characteristics of eight catchments.

Profile No.	Podzol type	Parent till (C)	Glacial deposit	Landscape	Topography
Zapoljarnij 1P28	haplic	stony sandy till	meltout till	moraine hill	top of the hill
Zapoljarnij 1P38	haplic	stony sandy till	meltout till	moraine plain	flat valley
Monchegorsk 2P36	haplic	silty sandy till	basal till	moraine hill	top of the hill
Monchegorsk 2P37	haplic	stony sandy till	basal till	moraine hill	top of the hill
Monchegorsk 2P38	ferric*	silty sandy till	basal till	mountain	gentle slope
Kirovsk 3P15	haplic	sandy till	basal till	mountain	flat terrace
Kirovsk 3P51	haplic	sandy till	meltout till	mountain	flat terrace
Kurka 4P06	haplic	sandy till	meltout till	moraine hill	foot of the hill
Kurka 4P38	ferric	stony sandy till	meltout till	moraine hill	gentle slope
Kurka 4P39	haplic	sandy till	meltout till	moraine plain	flat plain
Skjellbekken 5P37	ferric	stony sandy till	meltout till	moraine hill	gentle slope
Skjellbekken 5P39	haplic	sandy till	meltout till	moraine hill	top of the hill
Skjellbekken 5P40	ferric	silty sandy till	meltout till	moraine hill	top of the hill
Skjellbekken 5P41	ferric	gravelly sandy till+	meltout till	moraine hill	gentle slope
Kirakka 6P12	ferric	gravelly sandy till	meltout till	ablation moraine	gentle slope
Naruska 7P05	haplic	silty sandy till	basal till	moraine plain	flat plain
Naruska 7P18	haplic	silty sandy till	meltout till	moraine hill	gentle slope
Naruska 7P19	haplic	silty sandy till	basal till	moraine hill	gentle slope
Pallas 8P13	haplic	silty sandy till	meltout till	moraine plain	flat plain
Pallas 8P15	haplic	silty sandy till	basal till	moraine hill	gentle slope
Pallas 8P18	haplic (cryoic)	sandy till	meltout till	moraine plain	flat plain

\*no E layer

+ mixed with sulphide-rich saprolite

**Table 8.2.2. Podzol profiles and characteristics of the sampling sites. Soil classification was based on the FAO Soil Classification (FAO-Unesco 1980).**

a) total concentrations (conc. HNO3)

element	Mo	Zap	Kur	Kv	Skj	Kir	Nar	Pal
	c (mg/kg)							
(n)	(2)	(2)	(3)	(2)	(3)	(1)	(3)	(3)
S	1188	1435	1413	1220	1527	1360	1463	1490
Ni	2205	360	669	46	33	6.4	6.7	4.7
Cu	2490	49	378	27	23	6.8	7.6	6.5
Co	103	15	29	3.4	1.8	.44	1.2	3.0
Cr	30	3.4	12	10	2.5	1.3	2.6	2.7
As	28	3.6	5.3	2.8	1.7	1.6	1.5	1.3
Cd	.55	.34	.65	.30	.29	.26	.56	.50
Mn	45	46	252	472	484	39	144	149
Pb	42	13	32	34	14	12	33	23
V	31	5.5	13	19	5.3	1.7	3.6	5.0
Zn	33	36	44	45	70	43	62	27

b) easily leachable concentrations (NH4 acetate)

element	Mo	Zap	Kur	Kv	Skj	Kir	Nar	Pal
	c (mg/kg)							
S	93	55	86	66	90	75	94	102
Ni	105	88	72	2.9	4.0	.52	.54	.51
Cu	454	1.8	40	1.0	.37	.04	.07	.11
Co	6.7	6.1	5.6	.30	.39	.09	.27	.81
Cr	.47	.05	.15	.12	.11	.03	.06	.05
As	.78	.08	.09	.05	.03	<0.02	.03	<0.02
Cd	.16	.19	.38	.18	.17	.17	.30	.25
Mn	10	31	159	187	322	26	93	82
Pb	10	2.9	10	9.0	3.4	2.7	7.9	5.4
V	.75	.09	.22	.23	.06	.03	.08	.11
Zn	5.4	19	20	18	31	15	25	12

Table 8.2.3. Means of a) the total concentrations of heavy metals, As and S and b) the concentrations of the easily leachable heavy metals, As and S in the humus (O) layer in eight catchments. Keys for the catchments: Mo = Monchegorsk, Zap = Zapoljarnij, Kur = Kurka, Kv = Kirovsk, Skj = Skjellbekken, Kir = Kirakka, Nar = Naruska and Pal = Pallas.

a) hot aqua regia leach

element	Mo	Zap	Kur	Kv	Skj	Kir	Nar	Pal
E layer								
(n)	(2)	(2)	(3)	(2)	(3)	(1)	(3)	(3)
S	60	95	68	76	43	33	27	67
Ni	29	7.7	7.2	3.3	1.2	.50	2.7	2.1
Cu	72	1.3	4.3	2.2	.25	.25	.68	.59
Co	3.5	1.5	1.6	1.5	.81	.25	.76	1.0
Cr	17	5.7	7.0	7.6	5.4	3.0	8.1	8.7
As	1.6	.23	.57	1.0	.49	.12	.12	.10
Cd	.03	.03	.04	<0.02	<0.02	<0.02	<0.02	<0.02
Mn	61	29	50	55	44	25	26	38
Pb	1.9	3.8	4.7	8.3	2.4	4.0	2.2	2.8
V	24	31	17	28	16	11	8.8	19
Zn	5.3	3.9	4.1	5.6	4.3	2.1	4.2	2.7
B1 layer								
(n)	(2)	(2)	(3)	(2)	(3)	(1)	(3)	(3)
S	500	289	338	280	156	146	161	124
Ni	69	16	25	20	9.6	4.5	22	11
Cu	43	21	26	18	7.8	3.2	6.4	6.0
Co	12	8.5	8.1	6.8	6.4	2.7	7.1	4.5
Cr	70	34	55	42	28	16	47	28
As	.92	1.1	.49	.80	4.3	.33	1.1	.24
Cd	.08	<0.02	.04	.04	.03	<0.02	<0.02	<0.02
Mn	193	185	156	171	164	74	214	103
Pb	1.0	1.6	1.2	1.6	1.3	4.2	2.8	1.2
V	58	72	52	37	70	102	57	38
Zn	34	30	36	31	41	14	44	14
B2 layer								
(n)	(2)	(2)	(2)	(2)	(2)	(1)	(3)	(3)
S	357	181	178	160	126	128	154	84
Ni	59	21	28	18	14	6.1	29	12
Cu	34	44	34	14	17	5.4	7.6	6.1
Co	14	11	8.8	6.4	9.6	3.1	8.9	4.8
Cr	57	35	57	34	28	12	46	25
As	.29	1.1	.48	.84	5.8	.16	1	.24
Cd	.07	.03	.03	.04	.04	<0.02	<0.02	<0.02
Mn	183	232	161	146	230	97	187	110
Pb	.59	1.3	1.3	1.3	1.9	2.5	1.6	1.3
V	48	57	37	47	46	21	41	30
Zn	37	38	31	24	53	22	53	13
BC1 layer								
(n)	(2)	(2)	(3)	(2)	(3)	(1)	(3)	(3)
S	315	88	95	79	110	44	96	60
Ni	57	24	34	18	19	4.7	32	12
Cu	37	66	47	24	32	6.6	10	6.8
Co	13	13	9.8	7.4	10	3.1	10	5.4
Cr	55	37	53	28	27	6.5	48	27
As	.26	1.3	.44	.46	6.8	.05	1.2	.22
Cd	.05	.05	.04	.04	.04	<0.02	.04	<0.02
Mn	196	293	170	166	213	114	178	127
Pb	.71	1.2	1.4	1.2	1.7	3.7	1.9	1.1
V	46	64	36	30	46	12	42	35
Zn	32	40	35	24	49	21	41	13
C layer								
(n)	(7)	(4)	(8)	(5)	(10)	(3)	(11)	(10)
S	67	72	43	39	43	29	49	24
Ni	52	29	35	18	30	5.8	29	10
Cu	39	104	61	31	46	13	13	6.3
Co	13	15	13	7.4	14	4.5	9.9	5.1
Cr	47	47	53	27	26	7.7	46	21
As	.24	4.3	.46	.57	3.6	.05	1.2	.32
Cd	.05	.06	.03	.04	.06	<0.02	.04	<0.02
Mn	234	342	255	189	286	171	196	182
Pb	.89	1.2	1.7	1.1	1.3	3.7	2.0	1.3
V	43	68	42	33	54	21	44	28
Zn	26	43	36	23	46	27	41	11

Table 8.2.4. Means of a) the total concentrations of heavy metals, As and S and b) the concentrations of easily leachable heavy metals and S in the mineral soil layers in eight catchments. See keys for the soil layers in the text and for the catchments in Table 3.

b) easily leachable (NH4 acetate extraction)

Element	Mo	Zap	Kur	Kv	Skj	Kir	Nar	Pal
E layer								
	c (mg/kg)							
S	5.5	7.0	6.1	7.7	4.8	2.5	2.5	4.4
Ni	7.6	3.5	2.5	.36	.18	.03	.05	.09
Cu	.34	.13	.82	.18	.06	.05	<0.01	.03
Co	.37	.31	.22	.05	.03	.02	.02	.06
Cr	.28	.06	.14	.09	.04	.04	.08	.19
Mn	.46	.68	2.4	3.5	2.4	.19	.75	1.5
Pb	.32	<0.1	.42	1.2	.24	.23	.50	.20
V	.15	.16	.24	.12	.08	.03	.02	.05
Zn	.28	.93	.67	.79	1.6	.40	.46	.25
B1 layer								
S	270	118	129	72	51	41	43	35
Ni	10.8	.69	1.5	.15	.24	.06	.11	.09
Cu	6.2	.45	.89	.27	.16	.08	.10	.07
Co	.59	.05	.19	.06	.08	.03	.08	.05
Cr	3.0	.83	2.1	1.8	.58	.17	1.5	1.2
Mn	7.0	.44	4.3	3.3	7.3	.13	6.8	.32
Pb	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
V	.15	.16	.24	.16	.13	.15	.31	.29
Zn	.82	.29	.91	.39	2.4	.36	.80	.12
B2 layer								
S	221	87	91	54	32	45	67	21
Ni	4.4	.26	.94	.14	.11	.03	.08	.08
Cu	.53	.67	1.4	.28	.35	.14	.11	.07
Co	.26	.03	.15	.07	.15	.03	.08	.04
Cr	1.6	.65	1.4	1.2	.97	.36	.78	.97
Mn	2.4	.53	2.5	2.0	4.6	.47	3.0	.23
Pb	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
V	.18	.13	.20	.33	.15	.02	.21	.20
Zn	.76	.13	.41	.56	1.1	.24	.57	.05
BC1 layer								
S	207	36	38	25	28	18	45	17
Ni	3.2	.21	.30	.03	.13	.03	.08	.04
Cu	.69	.94	1.1	.62	.53	.18	.15	.07
Co	.18	.05	.06	.04	.17	.05	.08	.04
Cr	1.0	.47	.92	.34	.70	.19	.44	.79
Mn	1.6	1.3	.74	.76	3.6	1.8	1.2	.21
Pb	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
V	.21	.11	.17	.15	.14	.01	.16	.14
Zn	.60	.08	.25	.11	.39	.14	.22	.02
C layer								
S	39	16	12	10	8.9	7.9	21	6.5
Ni	2.4	.26	.26	.03	.10	.03	.03	.03
Cu	.48	1.6	.68	1.2	.60	.24	.27	.07
Co	.13	.04	.05	.03	.09	.03	.05	.04
Cr	.30	.36	.40	.13	.26	.11	.21	.28
Mn	1.2	1.4	.67	.63	1.1	.79	.44	.57
Pb	<0.1	<0.1	<0.1	<0.1	<0.1	.24	<0.1	<0.1
V	.10	.08	.08	.07	.05	.03	.13	.09
Zn	.21	.10	.13	.11	.36	.07	.04	.04

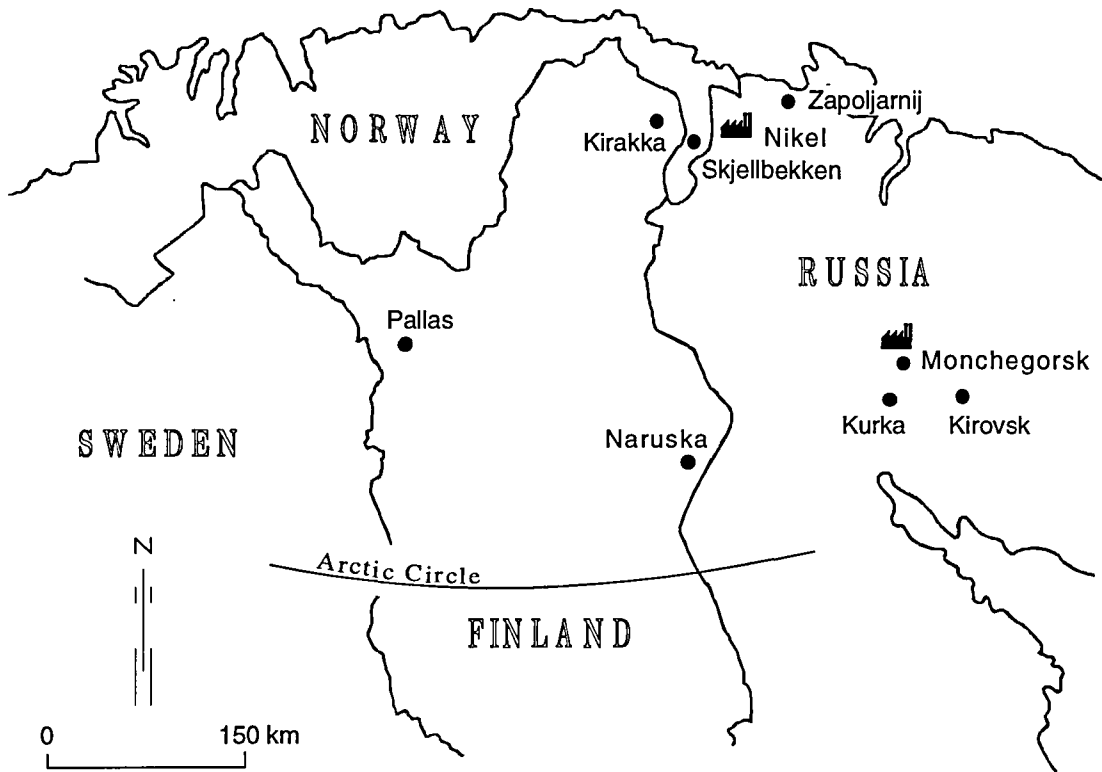


Fig. 8.2.1. Location of eight catchments in northern Finland and Norway and in the western half of the Kola Peninsula, Russia.

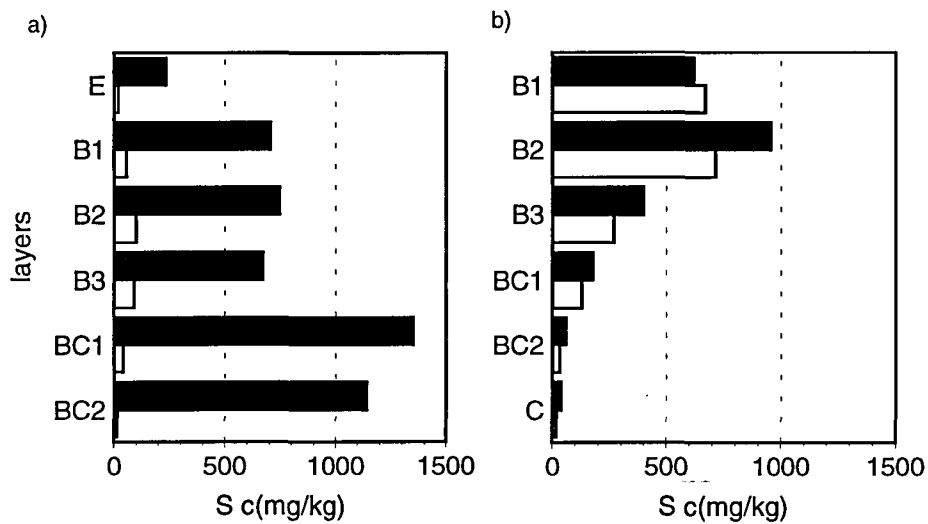


Fig. 8.2.2. Distribution of total S concentrations (black histograms) and concentrations of the easily leachable S (white histograms) in a) the profile 5P41 in Skjellbekken and b) the strongly eroded profile 2P38 in Monchegorsk.

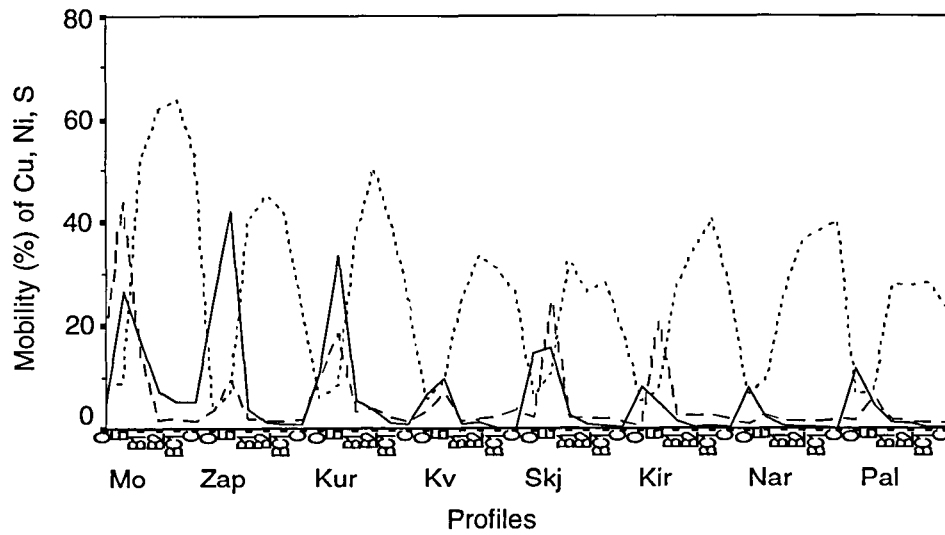


Fig. 8.2.3. Mobilities (%) of Cu (dashed line), Ni (thick line) and S (dotted line) within the profile in eight catchments. The mobility of the element was calculated as a proportion (%) of the concentration of easily leachable element from its total concentration. See keys for the catchments in Table 3a and for soil layers in the text.

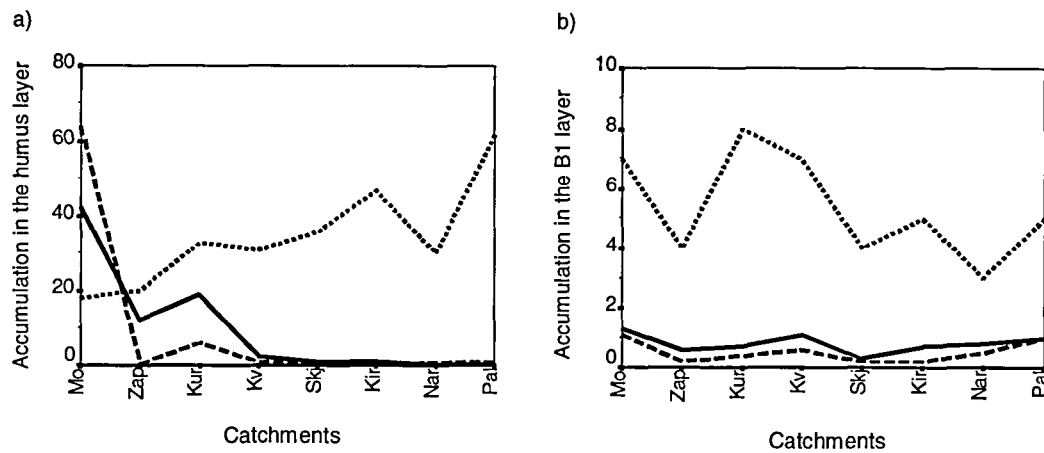


Fig. 8.2.4. Accumulation of Cu (dashed line), Ni (thick line) and S (dotted line) into a) the humus layer and b) the upper illuvial (B1) layer in eight catchments. Accumulation was calculated as a ratio of the total concentration of the element in the humus layer to that in the parent till (C layer). See keys for catchments in Table 3.

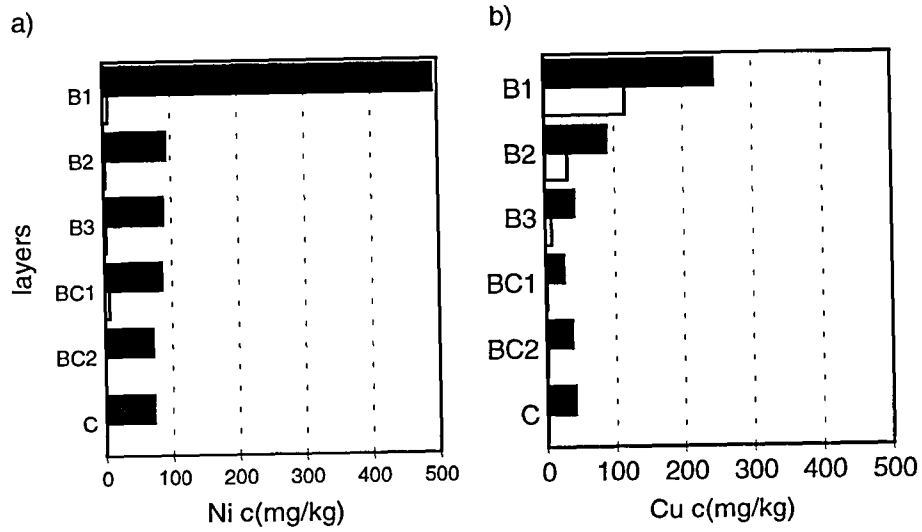


Fig. 8.2.5. Distribution of a) total Ni concentrations and concentrations of easily leachable Ni, and b) total Cu concentrations and concentrations of easily leachable Cu in the strongly eroded profile 2P38 in Monchegorsk. See keys for the soil layer in the text.



## 9 GROUNDWATER

### 9.1 Groundwater composition from two catchments on the Kola Peninsula: evidence for incipient contamination from industrial emissions

*Patrice de Caritat, Svetlana Danilova, Øystein Jæger, Clemens Reimann and Gaute Storø*

#### ABSTRACT

The chemical composition of 185 groundwater samples collected from two catchments in the Barents region over the period April 1994 to November 1995 is reported in terms of Ag, Al, As, B, Ba, Be, Bi, Br, Ca, Cd, Cl, Co, Cr, Cu, F, Fe, K, Li, Mg, Mn, Mo, Na, Ni, NO<sub>3</sub>, P, Pb, PO<sub>4</sub>, Rb, S, Sb, Se, Si, SO<sub>4</sub>, Sr, Th, Ti, Tl, U, V and Zn concentrations as determined by ICP-MS, ICP-AES and IC, pH and electrical conductance. One catchment (C2) is located in Russia 5 km downwind from the smelting industry in Monchegorsk, which is a major SO<sub>2</sub> and trace metal emission source, the other (C5) is located in Norway 30 km off-wind from the smelter in Nikel and 52 km off-wind from the ore roasting plant of Zapoljarniy, which are two other significant emitters of inorganic atmospheric pollutants. C2 is heavily polluted by industrial airborne contaminants, while C5 is only marginally affected. The groundwater in C2 shows clear signs of early contamination from the smelting industry in Monchegorsk (Ni up to 18 mg/l, SO<sub>4</sub> up to 21 mg/l). In C5, the groundwater is unpolluted, but has relatively high natural contents of Al (up to 460 mg/l), U (up to 0.7 mg/l) and Pb (up to 4.5 mg/l). The incipient groundwater contamination in the vicinity of Monchegorsk together with the polluted condition of many streams and lakes in the area can lead to problems in the supply of drinking water to the town's population.

#### INTRODUCTION

The Central Kola Expedition (CKE) and the Geological Surveys of Finland (GTK) and Norway (NGU) are carrying out a major, cooperative regional geochemical mapping project of a 188,000 km<sup>2</sup> area north of the Arctic Circle on the Kola Peninsula (NW Russia) and in adjacent areas in Finland and Norway (see Internet site <http://www.ngu.no/Kola>). One of the aims of the project is to assess the environmental impact of the nickel smelting and other heavy industry in Nikel, Zapoljarniy and Monchegorsk.

As one part of the project, element content in various sampling media (rain, snow, stream water, groundwater, moss, different soil horizons, Quaternary deposits and bedrock) was determined and/or monitored in eight selected catchments (watersheds) located at various distances from the smelters, and in different geological settings (Fig. 9.1.1). Four of these catchments are situated in Russia (C1: Zapoljarniy, C2: Monchegorsk, C3: Kirovsk and C4: Kurka), one in Norway (C5: Skjellbekken) and three in Finland (C6: Kirakka, C7: Naruska and C8: Pallas).

This paper presents the results from groundwater studies in the Skjellbekken (C5) and Monchegorsk (C2) catchments. The composition of the groundwater is reported rather comprehensively (pH, electrical conductance, plus concentration of 40 elements/species) making use of state-of-the-art analytical methods (ICP-MS) with detection limits in the part per trillion range (Stetzenbach et al., 1994). The major, minor and trace element composition

of groundwater is a function of, among others, the geochemical and mineralogical properties of the aquifer hosting it (e.g. Garrels and MacKenzie, 1967, Frapet et al., 1984, Bricker and Jones, 1995, Mather, 1996). Here, the two groundwaters studied are from different types of aquifers, occurring in lithologically distinct provinces, yet the most fundamental control on groundwater geochemistry appears to result from the extreme level of industrial pollution and ensuing environmental contamination at one site (C2) compared to relatively more pristine conditions at the other (C5). At the latter site, natural or background processes (weathering, etc.) dominantly control the composition of the groundwater. In this paper, the composition of the two groundwaters is described and interpreted in terms of element level and source, and it is compared with surface water composition; time-series will be reported separately (Storrø et al., in prep.).

## CATCHMENT DESCRIPTION

### *Skjellbekken (C5)*

C5 covers an area of 34.56 km<sup>2</sup>, ranges in elevation between 80 and 297 m above sea level, received 422 mm of precipitation in 1994 and 581 mm in 1995 (normal range 390-440 mm), and is vegetated by typical north taiga pine forest with birches; outlet coordinates are 69°21'25" north and 29°27'25" east (Fig. 9.1.1). The main bedrock types present are (1) andesite/andesitic volcanoclastic schist (40% areal coverage), (2) tholeiitic basalt/tuff (30%), (3) black schist (20%), and (4) other rock types including ultramafic/mafic intrusions (5%), limestone and quartzite. Surface covers consists of (1) till (71%) and (2) glacio-fluvial (15%) Quaternary deposits, (3) water bodies (6%), and (4) outcrop (5%).

C5 is located 30 km west-southwest (perpendicular to the dominant wind direction) of the Nickel Ni-Cu smelter, 52 km west-southwest of the Zapoljarniy ore roasting plant, and 48 km south-southwest of the Kirkenes Fe ore mine and mill (Fig. 9.1.1). Official figures for emissions from Nickel/Zapoljarniy in 1994 are ca. 198,000 tonnes SO<sub>2</sub>, 297 tonnes Ni, 163 tonnes Cu and 11 tonnes Co (data from Murmansk Region Committee of Ecology and Nature Resources, see Reimann et al., in prep. a). Deposition in 1994 over C5 was estimated to be 164 kg S/km<sup>2</sup>, 6 kg Ni/km<sup>2</sup>, 5 kg Cu/km<sup>2</sup> and 0.17 kg Co/km<sup>2</sup> (Chekushin et al., in prep.).

### 3.2 Monchegorsk (C2)

C2 covers an area of 22.38 km<sup>2</sup>, ranges in elevation between 128 and 507 m above sea level, received 391 mm of precipitation in 1994 (normal range 400-600 mm), and is described as a «technogenic desert» (Alexeyev, 1993, Kryuchkov, 1993) with some birch shrubs subsisting in remaining soil pockets; outlet coordinates are 67°50'30" north and 32°54'48" east (Fig. 9.1.1). The main bedrock types present are (1) gabbro/gabbro-norite (40% areal coverage), (2) andesite/tuff (32%), (3) pyroxenite (16%), and (4) (grano-)diorite. Quaternary overburden is principally represented by (1) till (76%), (2) glacio-fluvial (9%), (3) eluvial (8%), and (4) glacio-lacustrine (4%) deposits.

C2 is located 5 km south and downwind from the major SO<sub>2</sub> and trace element pollution source of the Monchegorsk smelter complex (Fig. 9.1.1). Official figures for emissions from Monchegorsk in 1994 are ca. 98,000 tonnes SO<sub>2</sub>, 1619 tonnes Ni, 934 tonnes Cu, 82 tonnes Co, 341 tonnes Cl and 1267 tonnes NO<sub>2</sub> (data from Murmansk Region Committee of Ecology and Nature Resources, see Reimann et al., in prep. a). Deposition in 1994 over C2 was estimated to be 654 kg S/km<sup>2</sup>, 845 kg Ni/km<sup>2</sup>, 494 kg Cu/km<sup>2</sup> and 54 kg Co/km<sup>2</sup> (Chekushin et al., in prep.).

## SAMPLING AND ANALYSIS

### *Skjellbekken (C5)*

Nine groundwater wells were drilled in mid-August 1994 at three different locations in the Quaternary deposits of the lower part of C5. These deposits consist of 6-11 m of glaciofluvial to glaciolacustrine sediments (fine sand to sand) overlying coarser esker sediments (gravel). At each location, a shallow (2-3 m depth), an intermediate (5-9 m) and a deep (7-12 m) well was cased with white PEH tubing (diameter 50 mm) and sealed with bentonite. Sediments samples were collected at all levels during drilling. A small electric pump (Eijkamp™ submersible pump type Whale) was installed in each of the nine wells for sampling.

Groundwater was sampled for the first time on 20/21 September 1994, and the second time on 9 February 1995. Groundwater was then sampled weekly from 11 April to 6 July 1995, and thereafter monthly until 7 November 1995. The groundwater level was measured in each well before sampling. A volume of water equivalent to at least two well shaft volumes was pumped out and discarded prior to collecting the sample for analysis. Surface water samples were collected weekly from Sjøllbekken, the main stream draining the catchment, from 18 March 1994 to 7 November 1995 (monthly in winter and from July 1995).

Each groundwater and stream water sample comprised two subsamples. The first subsample consisted of 100 ml of water filtered (0.45 mm Millipore™ membrane filters) and acidified (ultrapure nitric acid) in the field and stored in a cool room before dispatching to the laboratory for cation analysis. The second subsample consisted of 500 ml of unfiltered, unacidified water for anion, pH and electrical conductance (EC) analyses.

### *Monchegorsk (C2)*

In this catchment, groundwater was sampled from one hardrock well situated in the upper part of the catchment. The well penetrates ca. 5 m of fine sand and silt, and ca. 45 m of the underlying fractured gabbro. The upper 10 m of the well is cased with a steel pipe (diameter 127 mm), whereas the lower part is uncased (diameter 59 mm). The yield of the well is 1 l.sec<sup>-1</sup>. A small electric pump of the same type as described above was installed at 5 m depth for sampling the groundwater.

The groundwater was sampled weekly from 30 April 1994 to 8 November 1994, then monthly until 28 February 1995. Temperature, EC, pH and water level were measured in the field. Some well water was pumped out and discarded before taking the sample for analysis. The stream draining C2 was sampled weekly over the period 23 March 1994 to 2 June 1995 (monthly in winter). For sampling and preservation of the samples, the same procedures as in C5 apply here.

### *Analysis*

All chemical analyses were carried out at the GTK Laboratory, except for the determination of pH, EC and anion concentration of the first two sampling rounds in C5 (20-21 September 1994 and 9 February 1995), which was done at NGU. The analytical programme at GTK consisted in the quantitative determination of cation concentration by inductively coupled plasma-atomic emission spectrometry (ICP-AES: Ca, Mg, Na, P, S, Si) and inductively coupled plasma-mass spectrometry (ICP-MS: Ag, Al, As, B, Ba, Be, Bi, Cd, Co, Cr, Cu, Fe, K, Li, Mn, Mo, Ni, Pb, Rb, Sb, Se, Sr, Th, Ti, Tl, U, V, Zn), of anion concentration by ion chromatography (IC: Br, Cl, F, NO<sub>3</sub>, SO<sub>4</sub>) and spectrophotometry (PO<sub>4</sub>), and pH and EC (at 25°C) by potentiometry. Due to the wearing off of the teflon coating of an O-ring in a dispenser, some Zn results are spurious (H. Niskavaara, pers. comm. 1996). For more

information about analytical procedures, the reader is referred to Niskavaara (1995). The GTK Laboratory is accredited to meet the requirements of the EN 45001 standard and ISO Guide 25.

## RESULTS

Tables 9.1.1 and 9.1.2 summarise the data obtained for groundwater and stream water in both catchments, respectively. Minimum, 25th percentile, median (50th percentile), mean, standard deviation, 75th percentile and maximum reported values are given. Where results below the detection limit were encountered, half the detection limit value was used for the statistical treatment. The variance of the data results from (1) the contrasting hydrogeological setting of the two wells (overburden vs. hardrock aquifer), (2) the different levels sampled in C5 (shallow, intermediate and deep), and (3) the natural time-dependent variation in groundwater composition throughout the hydrological year (see Storrø et al., in prep.).

## DISCUSSION

### *Comparison of C2 and C5*

Median values in groundwater (Tab. 9.1.1, Fig. 9.1.2) are higher in C5 than C2 for (parameters are ranked in order of decreasing ratio, and if ratio <2 the parameter is given in parenthesis) Al (by a factor of 6.1), U (>6), Pb (3.2), (V), (K), (Mo), (B), (pH), (Si) and (Cd), whilst higher in C2 than C5 for Ni (10.3), Fe, SO<sub>4</sub>, S (5.3), NO<sub>3</sub>, Co (4), Cl, Ca, Mg (2.8), Mn, Zn (2.2), EC, (Ba), (Rb), (Cu), (Li), (Sr) and (Na).

The single greatest difference between the two groundwaters is for Ni, which is more than 10 times more abundant in C2 than in C5. This element can have both a geogenic source (though no Ni-mineralisation is known in C2) and an anthropogenic source (Ni emissions from the smelter). Given that the median Ni/Cu ratio in groundwater (5.6) is closer to the value of this ratio in the emission data (1.7; Reimann et al., in prep. a) or the deposition data (1.7; Chekushin et al., in prep.) than in normal 'background' groundwaters (0.17-0.5; Allard, 1995), and given the extreme Ni emission/deposition values, it appears reasonable to suggest that the high Ni content in groundwater from C2 results mostly from contamination.

Other elements also part of the ore being smelted at Monchegorsk (Cu, Co, S) are similarly higher in groundwater from C2 than from C5. Absence of known mineralisation within C2 and high industrial emission and deposition again likely indicate groundwater contamination from anthropogenic pollutants.

The relatively high median SO<sub>4</sub> (20 mg/l) and Cl (9.5 mg/l) values in C2 groundwater are also taken as evidence supporting the infiltration of contaminants down from the surface, since these elements are not abundant in rock-forming minerals in C2 and both have high emission values from neighbouring Monchegorsk.

High cation (Ca, Fe, Mg) concentration and EC in C2 can be mainly attributed to accelerated weathering reactions under extreme SO<sub>2</sub> loading ('acid deposition'), together with some direct deposition effects as seen in rain water and snow (Reimann et al., in prep. a, Chekushin et al., in prep., Åyräs et al., 1995, in prep.) (limestone and dolomite used as flux in the smelter?). Nitrate levels (0.9 mg/l) in groundwater at C2 also reflect elevated N emissions from Monchegorsk.

Groundwater from C2, being collected from a steel cased well (see above), can be contaminated, e.g. with Fe and heavy metals from the pipe. The degree to which this source of contamination influences groundwater composition is uncertain: on the one hand one might expect its relative importance to be limited in an area with such dramatic deposition of airborne heavy metals, on the other hand the high S deposition could enhance metal corrosion (but the pH of the groundwater sampled is circum-neutral). In addition, the heavy metal composition of the steel is unknown.

Al and U, which are more enriched in C5 than C2 groundwater, are likely to have a geogenic source here. Relatively elevated Pb concentrations in C5 groundwater has a problematic origin as no Pb(-Zn) mineralisation is known within C5. C2 receives 10 times more Pb deposition than C5, which appears to be at, or only slightly above, background Pb deposition levels (Chekushin et al., in prep.) and which has a well-developed humus layer that would effectively bind most deposited Pb (Lewis, 1977, Wang et al., 1995). Therefore, some minor subsurface source(s) (war relics or waste from a small industrial development) could play a role here. The simultaneous, relatively high concentrations of Al, U and Pb point towards some particular physico-chemical (Eh-pH) conditions within the aquifer favouring a high solubility.

#### *Comparison with other groundwaters*

Despite being located within, and on the margins of, an area with some of the world's most intense emission of SO<sub>2</sub> (Gunn et al., 1995) and accompanying trace element spectrum, the median concentrations of the 'standard' heavy metals in C2 and C5 groundwaters are within or below the published 'background' concentration ranges for groundwater (Allard, 1995) for Cr, Ni (C5 only), Cu, Zn (C5 only), As, Cd and Pb. C2 groundwater is above the quoted 'background' only for Ni (by a factor of 10) and Zn (by a factor of 2-3). The overall result that most heavy metal concentrations are below 'background' levels, even in C2 where groundwater is clearly influenced by some of the highest heavy metal emissions in the whole world (Gunn et al., 1995) casts doubt on the general universal applicability and usefulness of such 'background' ranges.

Since the published 'background' levels only apply to very few elements, it is interesting to also compare (hardrock) groundwater from C2 with the hardrock groundwaters from Norway recently analysed by Reimann et al. (in prep. b). They reported the concentration of 64 elements in 145 raw (unfiltered) groundwater samples from the Oslo and Bergen regions. Because the groundwater samples from C2 were filtered, many elements are much more abundant in the Norwegian study, for instance, comparing median values, Cu (by a factor of 11.3), B (11), Li (7.9), Mo (7.4), Na (5.1) and Al (3.8). More interesting here, is the suite of elements more abundant in filtered groundwater from C2 than in the raw Oslo/Bergen groundwaters: Ni (by a factor of 7.8), Co (3.9), Fe (2.8) and Mn (1.8). Thus, two metals (Ni, Co) from the sulphide ore being smelted in Monchegorsk, and two elements (Fe, Mn) of likely geogenic origin (mineral weathering accelerated under intense acid deposition, plus role of Eh-pH conditions) are clearly present in anomalous proportions in the groundwater near Monchegorsk.

Median groundwater composition from C2 and C5 is below the Dutch A-value ('reference level') as given in Ewers (1991, Tab. I.20b-14) for As, Ba, Cd, Co, Cr, Cu, Mo, Ni, Pb and Zn. Even when comparing our maximum values, very few instances of exceeding the Dutch A-value are found in C2: Ni and Zn (>B-value); and in C5: Mo and Zn (>C-value; sample contaminated?). For Cu and Cr, the maximum values in C5 comes close to (>half) the Dutch A-value.

### *Comparison with drinking water standards*

Drinking water standards (DWS) and guidelines (GL) collated by Ewers (1991, Tab. I.20b-7) and including data from the World Health Organization (WHO), the Council of the European Communities (CEC) and the United States Environmental Protection Agency (U.S. EPA) are all (well) above any detect concentrations in C2 or C5 groundwaters for Ag, As, Ba, Ca, Cd, Cr, Cu, K, Mg, Mo, Na, Ni, Pb, Sb, Se and Sr. The only exceedences (all given here in mg/l) noted are for Al (maximum C5 = 463 vs. DWS = 200), Fe (median C2 = 70, maximum C2 = 1230 and maximum C5 = 310 vs. CEC GL = 50 and DWS = 200 or 300), Mn (maximum C2 = 142 and maximum C5 = 53.7 vs. CEC GL = 20 and DWS = 50 or 100) and Zn (maximum C2 = 100 and maximum C5 [contaminated?] = 1970 vs. CEC GL = 100).

The issue of incipient groundwater contamination by industrial emissions is a serious one in an area like Monchegorsk because of the already advanced stage of surface water pollution there (Moiseenko, 1994, Moiseenko et al., 1995, Väisänen et al., in prep., Caritat et al., in prep. a,b). If groundwater can not be considered a safe alternative source of potable water, serious drinking water supply problems may follow, as transport of water in pipe-lines over long distances is precluded by climatic and ground conditions in these arctic regions.

### 6.4 Comparison between groundwater and stream water

The geochemistry of the stream water in all eight catchments of the Kola Ecogeochemistry project is described in detail elsewhere (Caritat et al., in prep. a,b). Accordingly, stream water and groundwater quality is compared here with focus especially on groundwater processes. The composition of stream water and groundwater in C2 is compared in Fig. 9.1.3, and in Tables 9.1.1 and 9.1.2. Parameters higher in stream water compared to groundwater are (parameters are ranked in order of decreasing ratio, and if ratio <2 the parameter is given in parenthesis) Cu (by a factor of 40), Ni (40), Co (29), Al, Cd, V (3.4), As, NO<sub>3</sub>, Cr (>2.5), Sb, Mo, (Ag), (U), (Mg), (Mn), (Ba), (Sr), (S), (B), (Pb), (SO<sub>4</sub>), (Zn), (Li), (EC) and (Si), whilst those higher in groundwater are Cl (2.4), (Fe), (Na), (Rb), (K) and (Ca).

The fact that the three most important sulphide metals, Ni, Cu, Co, together with S and a suite of other trace metals typical of the ore, are (much) more abundant in stream water than in groundwater in C2 is taken as an indication of a much less severe contamination level of the aquifer than of surface water bodies. Many other elements are also more abundant in stream water because of enhanced weathering reaction in the upper soil layers due to acid deposition (SO<sub>2</sub>, NO<sub>x</sub>). Not surprisingly, of the few elements more abundant in groundwater, all but Cl (and partly Na, e.g. feldspar dissolution) likely find their origin in weathering of rock-forming minerals. The relatively high Cl concentration in groundwater (9.5 mg/l) remains unexplained at present.

The composition of stream water and groundwater in C5 is compared in Fig. 9.1.4, and in Tables 9.1.1 and 9.1.2. Parameters higher in stream water compared to groundwater are Ca (by a factor of 4.4), As (4.3), Fe, Br, EC, (SO<sub>4</sub>), (Ba), (Sr), (S), (Ni), (Cu), (Mg), whilst those higher in groundwater are Zn (6.5), Co, Pb, Al (2.5), Si, Mn, (K), (Mo), (V), (B), (Na), (Rb), (Li), (Cl) and (pH).

The higher abundance of typical geogenic elements (Ca, Fe, Mg, Sr) in stream water than in groundwater, together with the opposite relationship for typical seaspray elements (Na, Cl), suggest that these two waters are in very good communication with one another in C5. The bedrock, which includes limestone, and the Quaternary deposits here are known to be generally

Ca-rich (Melezhic, 1995, Olsen, 1995, Caritat, 1995 a,b). Perhaps the occasional acid precipitation events (up to 3 mg SO<sub>4</sub>/l) as recorded in the rain samples (Reimann et al., in prep. a) cause leaching of carbonate and other Ca-bearing minerals in the upper soil layers, the acidity of the infiltrating water being progressively buffered as it percolates down to the aquifer (pH groundwater > stream water).

The median Cl content of groundwater (2.45 mg/l) is higher than in stream water (2.32), but the difference is too small to be significant. Rather, the concentration levels in both waters should be seen as similar. The maximum Cl concentrations show clearly the influence of seasalt transported during storms onto the catchment and its surface water reservoir. Sodium can have a geogenic component (Na-feldspar, etc.), in addition to the seasalt source, hence explaining its higher (median and maximum) concentration in groundwater than in stream water.

Arsenic in C5, being enriched in rain (Reimann et al., in prep. a) and in stream water (Caritat et al., in prep. a) compared to the background catchments of the Kola project, likely finds its source mainly as a technogenic airborne contaminant. This coincides well with its greater abundance in stream water than groundwater in C5 (as it does in C2). A smaller geogenic contribution, however, is also likely given the bedrock and Quaternary deposit compositions in C5 (Pavlov et al., in prep.).

Higher abundances of Zn and Co in groundwater in C5 than in stream water have a natural cause here: both bedrock and Quaternary deposits have unusually elevated Zn and Co content (Pavlov et al., in prep.). For Pb, the relative enrichment in groundwater is more problematic: the content in rocks and overburden is not unusual. If Pb was deposited as an anthropogenic pollutant, one would expect it to be strongly bound to the humus layer (Lewis, 1977, Wang et al., 1995), and not be mobilised either to surface or groundwater.

Other elements result clearly from weathering of aluminosilicates in the soil and aquifer: Al, Si, Mn, K and Li.

To further give an impression of which elements are part of the emission spectrum of the Monchegorsk smelter, it is interesting to compare the median geochemistry of the two streams (Tab. 9.1.2, Fig. 9.1.5). Parameters greater in C2 than in C5 are Co (by a factor of 341!), Ni (241), Cu (42), Zn (19), NO<sub>3</sub>, Mn, Cd, S, SO<sub>4</sub>, Al, Mg, V, Mo, Cr, Sb, Si, Li, (Ag), (Cl), (B), (Ba), (Rb), (Sr), (K), (Na), (Pb) and (EC), whilst those greater in C5 are V (3), (Ca), (As) and (pH).

## CONCLUSIONS

The composition of groundwater from two catchments in the Barents region (C2 in NW Russia and C5 in NE Norway) is reported in terms of pH, electrical conductance, plus concentration of Ag, Al, As, B, Ba, Be, Bi, Br, Ca, Cd, Cl, Co, Cr, Cu, F, Fe, K, Li, Mg, Mn, Mo, Na, Ni, NO<sub>3</sub>, P, Pb, PO<sub>4</sub>, Rb, S, Sb, Se, Si, SO<sub>4</sub>, Sr, Th, Ti, Tl, U, V and Zn as determined by state-of-the-art (e.g. ICP-MS) analysis of 185 samples collected over the period April 1994 to November 1995. C2 is adjacent to the Monchegorsk industrial centre, which is a major SO<sub>2</sub> and trace metal emission source, whilst C5 is located further away and off-wind from the smelter in Nikel and the ore roasting plant of Zapoljarniy.

Median concentrations in groundwater (Tab. 9.1.1, Fig. 9.1.2) are >2 times higher in C2 than C5 for Ni (by a factor of 10.3), Fe, SO<sub>4</sub>, S (5.3), NO<sub>3</sub>, Co (4), Cl, Ca, Mg (2.8), Mn and Zn (2.2). These elements are either components of the ore being smelted in nearby Monchegorsk (Ni, S, Co, Zn), otherwise part of the emission cocktail of this industry (NO<sub>3</sub>, Cl), or base

cations mobilised by intensified weathering under acid deposition (Fe, Ca, Mg, Mn). As a result, EC is also much higher in groundwater from C2 than C5. Together, these findings imply that the groundwater at C2 is strongly influenced, directly and indirectly, by industrial activity in adjacent Monchegorsk, as no mineralisation that could naturally be responsible for elevated heavy metal content in the groundwater is known in C2.

Median concentrations in groundwater are >2 times higher in C5 than C2 for Al (by a factor of 6.1), U (>6) and Pb (3.2). These elements all dominantly have a subsurface source here and are thought to owe their high solubility to peculiar Eh-pH conditions in the aquifer. Al and U probably originate exclusively from natural, geogenic sources, whereas Pb may partly have its origin in some minor subsurface waste deposit.

Compared with published 'background' composition ranges for groundwaters (Allard, 1995) only median Ni and Zn in C2 exceed 'background' (by a factor of 10 and 2-3, respectively). Cr, Cu, As, Cd and Pb concentrations are all below 'background'. This casts doubt on the universal validity of such 'background' ranges, since groundwater in C2 clearly is being contaminated by fallout from one of the largest point-source emitters of sulphur and heavy metals in the world.

The polluted nature of the ecosystem in C2 is beyond doubt («technogenic desert»). Compared with 145 raw (unfiltered) hardrock groundwaters from Oslo and Bergen recently analysed (Reimann et al., in prep. b), the (filtered) hardrock groundwater of C2 contains more Ni (by a factor of 7.9), Co (3.9), Fe (2.8) and Mn (1.8). The two first elements are metals of the sulphide ore smelted at Monchegorsk, attesting to their infiltration down to the aquifer, the latter two are likely geogenic in origin.

The median content of As, Ba, Cd, Co, Cr, Cu, Mo, Ni, Pb and Zn in groundwater from C2 and C5 are all below the Dutch A-value for groundwater. The maximum value reported for Ni and Zn in C2 and for Mo and Zn in C5 exceed the Dutch A-value.

The only elements found to exceed drinking water standards or guidelines in any one instance or more are Al, Fe, Mn and Zn. Given that surface waters around Monchegorsk are under severe environmental stress due to the level of airborne contamination here, groundwater could prove a viable alternative for the source of drinking water. However, the first results for the groundwater in C2 presented here clearly indicate the groundwater resources are experiencing incipient stages of (inorganic) contamination.

Median stream water in C2 contains 40 times more Cu and Ni, and 29 times more Co than the groundwater here, and 341 times more Co, 241 times more Ni and 42 times more Cu than stream water in C5. Thus, at least in terms of Ni and Co, the groundwater in C2 is significantly more enriched than even stream water in C5.



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## TABLES

Tab. 9.1.1 Statistical summary of the parameters determined in groundwater

Tab. 9.1.2 Statistical summary of the parameters determined in stream water

## FIGURES

Figure 9.1.1. Location map of catchments in Russia (C1: Zapoljarniy, C2: Monchegorsk, C3: Kirovsk and C4: Kurka), Norway (C5: Skjellbekken) and Finland (C6: Kirakka, C7: Naruska and C8: Pallas). K: Kirovsk, Ki: Kirkenes (open-pit iron mine and mill), M: Monchegorsk (nickel-copper smelter, cobalt smelter, etc.), Mu: Murmansk, N: Nikel (nickel-copper smelter), Z: Zapoljarniy (nickel-copper ore roasting plant).

Figure 9.1.2. Median composition of groundwater in C5 vs. groundwater in C2 (in mg/l, except for EC, in mS/m, and pH, in pH-units).

Figure 9.1.3. Median composition of stream water in C2 vs. groundwater in C2 (in mg/l, except for EC, in mS/m, and pH, in pH-units).

Figure 9.1.4. Median composition of stream water in C5 vs. groundwater in C5 (in mg/l, except for EC, in mS/m, and pH, in pH-units).

Figure 9.1.5. Median composition of stream water in C5 vs. stream water in C2 (in mg/l, except for EC, in mS/m, and pH, in pH-units).

Detection Limit	GROUNDWATER C2 (n=28; 30 Apr. 1994-28 Feb. 1995)								GROUNDWATER C5 (n=157; 20 Sep. 1994-7 Nov. 1995)							
	Min	25%ile	Median	Mean	Std.Dev.	75%ile	Max	Min	25%ile	Median	Mean	Std.Dev.	75%ile	Max		
Ag	0.01	<0.01	<0.01	<0.01	0.01	0.02	<0.01	0.1	<0.01	<0.01	<0.01	0.01	0	<0.01	0.03	
Al	0.1	0.52	1.63	3.18	4.07	3.41	5.25	12.6	0.59	6.72	19.4	49.24	81.17	47.4	463	
As	0.05	0.08	0.11	0.13	0.13	0.03	0.15	0.24	<0.05	0.05	0.13	0.85	1.16	1.47	3.51	
B	0.5	<0.5	1.39	1.86	3.83	11.36	2.2	62.8	<0.5	1.245	2.12	2.30	1.16	3.275	6.7	
Ba	0.04	4.25	8.08	12.1	12.58	5.40	16	22.9	1.42	4.74	6.22	8.81	5.70	12.65	20.8	
Be	0.1	<0.1	<0.1	<0.1	<0.1		<0.1	0.11	<0.1	<0.1	<0.1	<0.1		<0.1	<0.1	
Bi	0.03	<0.03	<0.03	<0.03	<0.03		<0.03	<0.03	<0.03	<0.03	<0.03	<0.03		<0.03	<0.03	
Br	30/100	<100	<100	<100	<100		<100	<100	<30	<30	<30	40	36	88	120	
Ca	10	8500	10100	12600	12113	2350	14200	15700	1620	2250	3630	8900	8377	11800	26200	
Cd	0.02	<0.02	<0.02	<0.02	0.02	0.01	<0.02	0.06	<0.02	<0.02	0.02	0.03	0.03	0.04	0.16	
Cl	100	5970	9000	9500	9290	1216	10100	11400	1730	2250	2450	2491	397	2590	4080	
Co	0.02	<0.02	0.09	0.24	0.27	0.20	0.42	0.67	<0.02	0.03	0.06	0.11	0.22	0.11	2.31	
Cr	0.2	<0.2	<0.2	<0.2	<0.2		0.24	0.42	<0.2	<0.2	<0.2	<0.2		0.25	0.87	
Cu	0.04	0.21	0.8	1.04	1.32	0.99	1.71	4.24	0.22	0.47	0.69	0.87	1.11	0.91	11.5	
F	50	<50	<50	<50	<50		<50	310	<50	<50	<50	<50		50	83	
Fe	10	<10	30	70	134	224	150	1230	<10	<10	<10	31	49	40	310	
K	10	1050	1310	1360	1363	141	1420	1820	310	1255	1760	1795	1074	2025	4540	
Li	0.1	0.36	0.42	0.46	0.54	0.24	0.54	1.28	<0.1	0.145	0.31	0.42	0.35	0.575	1.67	
Mg	10	1510	2100	2330	2230	333	2470	2700	460	640	840	1098	557	1475	2240	
Mn	0.02	1.38	5.61	13.8	26.94	31.79	34.7	142	0.16	1.835	5.09	9.60	11.67	12.15	53.7	
Mo	0.03	0.06	0.12	0.22	0.34	0.36	0.37	1.78	<0.03	0.12	0.26	0.95	1.69	0.7	10.4	
Na	100	2700	3000	3400	3317	371	3600	3900	1830	2390	2660	2767	466	3130	4200	
Ni	0.06	0.33	2.12	5.79	6.73	4.87	9.6	17.8	0.11	0.425	0.56	0.69	0.47	0.86	4.3	
NO <sub>3</sub>	200	<200	600	900	880	485	1270	2100	<200	<200	<200	<200		<200	1210	
P	50	<50	<50	<50	<50		<50	<50	<50	<50	<50	<50		<50	<50	
Pb	0.03	<0.03	<0.03	0.05	0.05	0.04	0.07	0.19	<0.03	0.09	0.16	0.32	0.52	0.285	4.48	
PO <sub>4</sub>	20/100	<20	<20	<20	32	60	<20	260	<100	<100	<100	<100		<100	<100	
Rb	0.01	0.71	1.04	1.23	1.24	0.31	1.42	1.84	0.19	0.56	0.77	0.92	0.57	1.18	2.7	
S	50	4050	6260	6550	6285	751	6690	7080	340	660	1230	2132	1775	4275	5850	
Sb	0.025	<0.025	<0.025	<0.025	<0.025		<0.025	0.09	<0.025	<0.025	<0.025	0.03	0.03	0.045	0.23	
Se	0.5	<0.5	<0.5	<0.5	<0.5		<0.5	<0.5	<0.5	<0.5	<0.5	<0.5		<0.5	0.68	
Si	100	300	2120	4100	3397	1695	4700	5190	2300	3510	4240	4087	1023	4605	7060	
SO <sub>4</sub>	200	12000	17700	20100	18907	2505	20500	21000	1070	2020	3620	6125	5022	11900	21000	
Sr	0.1	15.5	22.9	25	24.05	3.92	25.7	36.7	7.35	12.35	17.2	21.39	12.15	27.05	44.9	
Th	0.02	<0.02	<0.02	<0.02	<0.02		<0.02	<0.02	<0.02	<0.02	<0.02	0.03	0.05	<0.02	0.3	
Ti	5	<5	<5	<5	<5		<5	<5	<5	<5	<5	<5		<5	10	
Tl	0.02	<0.02	<0.02	<0.02	<0.02		<0.02	<0.02	<0.02	<0.02	<0.02	<0.02		<0.02	<0.02	
U	0.01	<0.01	<0.01	<0.01	0.01	0.01	0.02	0.04	<0.01	0.02	0.06	0.14	0.15	0.3	0.71	
V	0.02	0.1	0.13	0.19	0.34	0.26	0.52	0.94	0.06	0.205	0.29	0.41	0.31	0.53	1.29	
Zn	0.1	2.24	15.6	28.9	75.24	132.54	48.1	616	0.71	3.09	12.9	72.56	275.33	17.2	1970	
EC		8.8	10.3	11.1	11.30	1.34	12.6	13	2.5	3.4	5.1	7.49	4.89	10	16	
pH		6.41	6.61	7	7.01	0.46	7.2	8.56	6.2	7.2	7.6	7.53	0.44	7.9	8.2	

Tab. 9.1.1 Statistical summary of the parameters determined in groundwater

Detection Limit	STREAM WATER C2 (n=45; 23 Mar. 1994-2 Jun. 1995)								STREAM WATER C5 (n=55; 18 Mar. 1994-7 Nov. 1995)							
	Min	25%ile	Median	Mean	Std.Dev.	75%ile	Max	Min	25%ile	Median	Mean	Std.Dev.	75%ile	Max		
Ag	0.01	<0.01	0.01	0.02	0.03	0.02	0.04	0.08	<0.01	<0.01	<0.01	0.01	0	<0.01	0.03	
Al	0.1	6.02	16.1	31.1	55.26	131.08	47.9	895	1.9	4.4125	7.83	9.18	5.71	12.35	24.8	
As	0.05	0.23	0.36	0.43	0.59	0.49	0.53	2.39	0.35	0.53	0.56	0.57	0.09	0.61	0.83	
B	0.5	0.6	2.07	2.62	2.63	0.88	3.14	5.27	0.52	0.8325	1.52	1.48	0.62	1.97	2.82	
Ba	0.04	13.9	16.4	18.9	18.98	2.97	20.5	26.9	7.41	10.4	11.4	11.78	2.46	13.28	19.7	
Be	0.1	<0.1	<0.1	<0.1	<0.1		<0.1	0.22	<0.1	<0.1	<0.1	<0.1		<0.1	<0.1	
Bi	0.03	<0.03	<0.03	<0.03	<0.03		<0.03	<0.03	<0.03	<0.03	<0.03	<0.03		<0.03	<0.03	
Br	30/100	<100	<100	<100	<100		<100	<100	<100	<100	<100	<100		<100	<100	
Ca	10	4900	9030	12000	11036	2605	12700	14600	7650	12900	16100	15209	3156	17850	19400	
Cd	0.02	0.05	0.1	0.15	0.35	0.54	0.29	2.51	<0.02	<0.02	<0.02	<0.02		<0.02	0.03	
Cl	100	1880	3270	4000	3828	768	4400	5500	1440	2180	2320	2613	1945	2650	16500	
Co	0.02	1.57	3.39	6.81	12.82	16.38	14	83.8	<0.02	<0.02	<0.02	<0.02		<0.02	0.03	
Cr	0.2	<0.2	0.41	0.5	0.55	0.32	0.72	1.98	<0.2	<0.2	<0.2	<0.2		<0.2	0.21	
Cu	0.04	16.5	25.8	41	93.10	169.34	81.4	1100	0.25	0.755	0.97	0.98	0.34	1.16	2.24	
F	50	<50	<50	<50	<50		50	80	<50	<50	<50	<50		<50	50	
Fe	10	10	20	40	48	45	50	240	10	23	40	42	24	50	130	
K	10	730	1010	1080	1157	336	1210	2600	530	730	905	885	190	1030	1230	
Li	0.1	0.15	0.4	0.56	0.68	0.41	0.81	1.99	<0.1	0.2025	0.275	0.30	0.18	0.34	1.04	
Mg	10	1640	3090	4150	3801	904	4500	4940	590	938	1175	1113	224	1300	1500	
Mn	0.02	9.91	12.7	21.8	44.58	44.45	63.9	166	1.03	1.89	2.49	2.70	1.18	2.87	6.16	
Mo	0.03	0.15	0.39	0.46	0.45	0.13	0.54	0.79	0.09	0.1425	0.16	0.16	0.03	0.18	0.21	
Na	100	1000	2280	2500	2365	517	2600	3700	1170	1760	2100	2038	355	2300	2900	
Ni	0.06	96	150	223	261.84	171.74	320	1100	0.41	0.795	0.925	1.00	0.26	1.19	1.64	
NO <sub>3</sub>	200	1400	2000	2430	2829	1109	3500	5500	<200	<200	<200	219	228	280	1470	
P	50	<50	<50	<50	<50		<50	<50	<50	<50	<50	<50		<50	<50	
Pb	0.03	<0.03	0.04	0.07	0.12	0.14	0.14	0.57	<0.03	0.04	0.06	0.07	0.05	0.09	0.25	
PO <sub>4</sub>	20/100	<20	<20	<20	10		<20	20	<20	<20	<20	<20		<20	<20	
Rb	0.01	0.47	0.76	0.96	1.05	0.40	1.11	2.15	0.45	0.63	0.665	0.68	0.07	0.72	0.85	
S	50	5600	8370	9230	9169	1400	9940	12500	1450	2040	2190	2244	337	2550	2980	
Sb	0.025	<0.025	0.04	0.06	0.06	0.03	0.08	0.15	<0.025	<0.025	<0.025	<0.025		<0.025	0.03	
Se	0.5	<0.5	<0.5	<0.5	0.50	0.32	0.68	1.4	<0.5	<0.5	<0.5	<0.5		<0.5	0.6	
Si	100	1600	3600	4200	4071	891	4640	5500	1170	1443	1900	1970	595	2490	3400	
SO <sub>4</sub>	200	14600	25000	27500	27496	4978	30200	39000	4040	6020	6600	6647	1028	7600	9100	
Sr	0.1	20.2	31.9	38	36.79	6.48	41.2	48.5	13.6	23.5	31.1	28.70	6.81	33.33	39	
Th	0.02	<0.02	<0.02	<0.02	<0.02		<0.02	<0.02	<0.02	<0.02	<0.02	<0.02		<0.02	<0.02	
Ti	5	<5	<5	<5	<5		<5	<5	<5	<5	<5	<5		<5	<5	
Tl	0.02	<0.02	<0.02	<0.02	0.02	0.01	0.02	0.07	<0.02	<0.02	<0.02	<0.02		<0.02	<0.02	
U	0.01	<0.01	0.01	0.02	0.02	0.01	0.02	0.05	0.02	0.04	0.06	0.06	0.02	0.07	0.09	
V	0.02	0.47	0.58	0.64	0.72	0.29	0.74	1.86	0.11	0.17	0.19	0.20	0.04	0.22	0.32	
Zn	0.1	6.45	30.8	38.1	181.63	638.48	44.9	3820	0.42	1.1625	1.97	4.69	5.54	5.34	25	
EC		6	9.8	12.1	11.52	2.19	13	14.8	5.3	8.55	10.4	9.95	1.99	11.55	13.3	
pH		5.78	6.7	7	6.96	0.44	7.28	7.6	6.65	7.2	7.3	7.40	0.39	7.65	8.1	

Tab. 9.1 2 Statistical summary of the parameters determined in stream water

*Kola Project (CKE, GTK, NGU)*  
*Catchment Study 1994*  
 Catchment locations

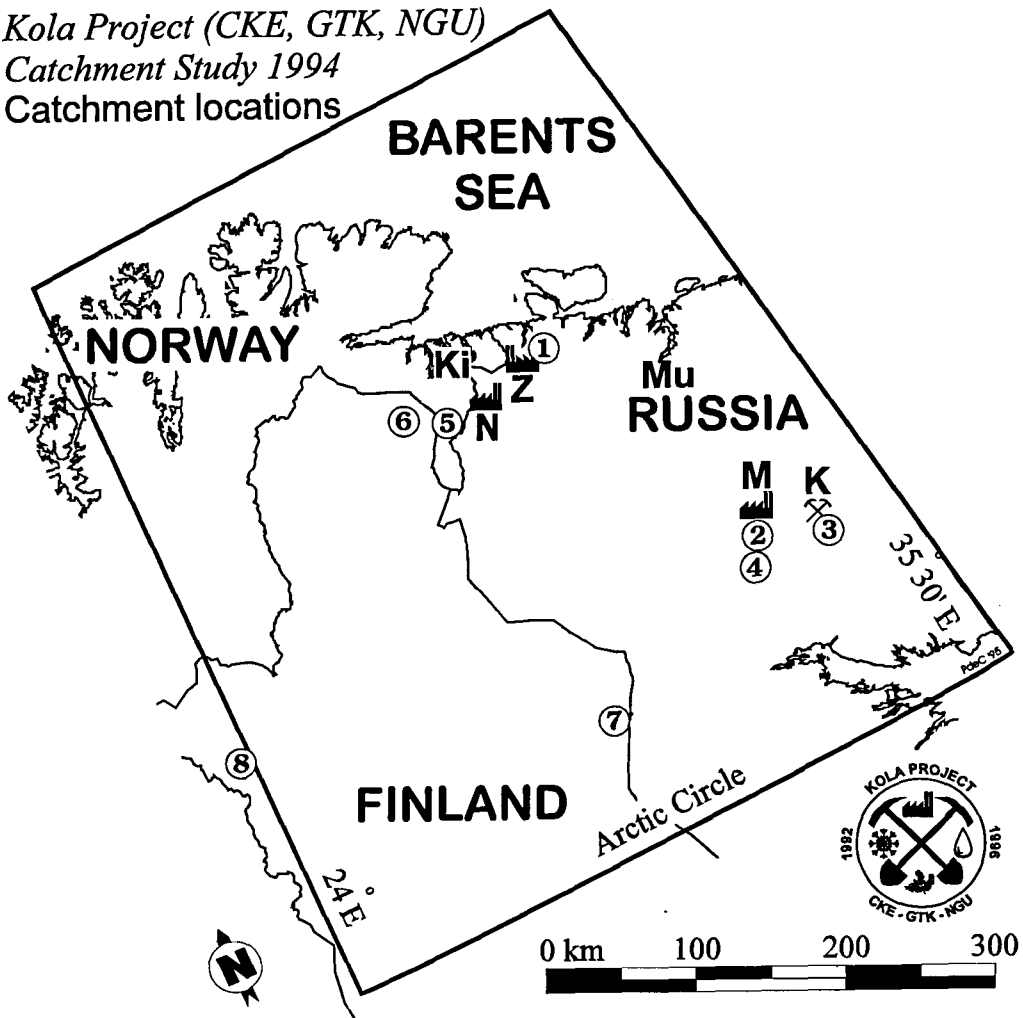


Figure 9.1.1. Location map of catchments in Russia (C1: Zapoljarniy, C2: Monchegorsk, C3: Kirovsk and C4: Kurka), Norway (C5: Skjellbekken) and Finland (C6: Kirakka, C7: Naruska and C8: Pallas). K: Kirovsk, Ki: Kirkenes (open-pit iron mine and mill), M: Monchegorsk (nickel-copper smelter, cobalt smelter, etc.), Mu: Murmansk, N: Nikel (nickel-copper smelter), Z: Zapoljarniy (nickel-copper ore roasting plant).

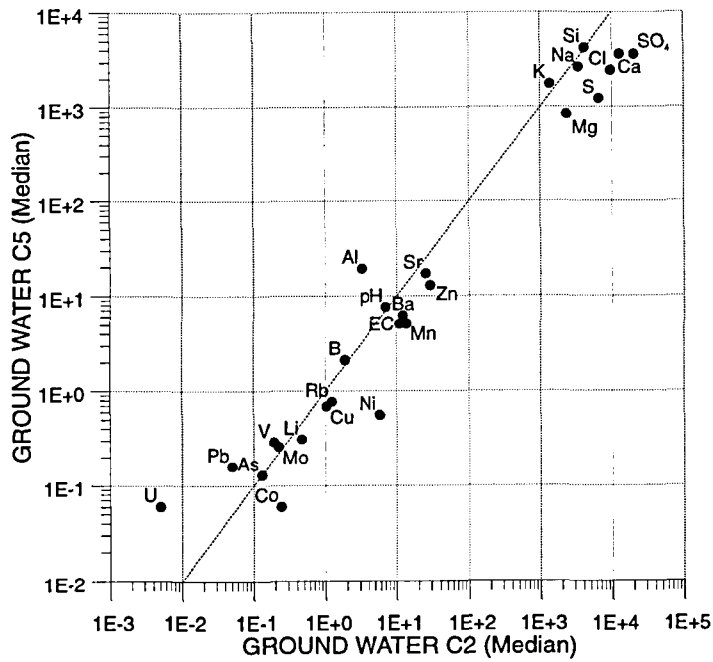


Figure 9.1.2. Median composition of groundwater in C5 vs. groundwater in C2 (in mg/l, except for EC, in mS/m, and pH, in pH-units).

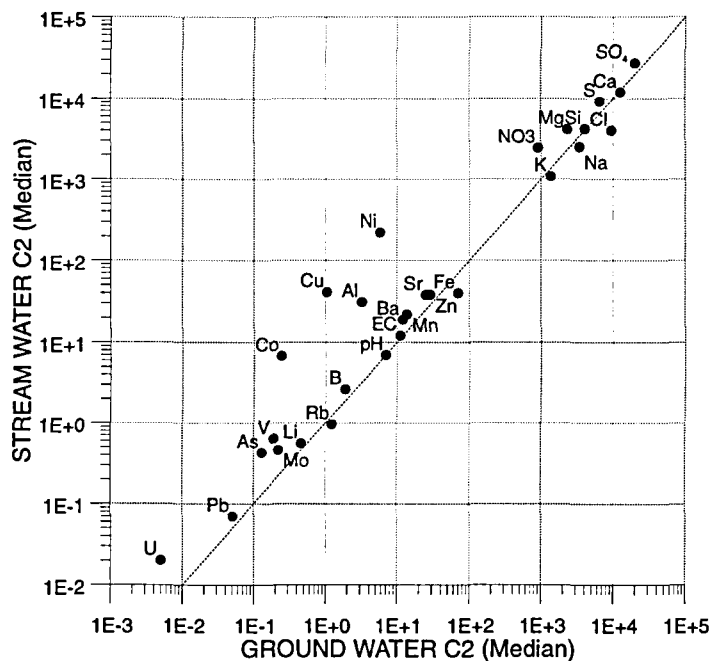


Figure 9.1.3. Median composition of stream water in C2 vs. groundwater in C2 (in mg/l, except for EC, in mS/m, and pH, in pH-units).



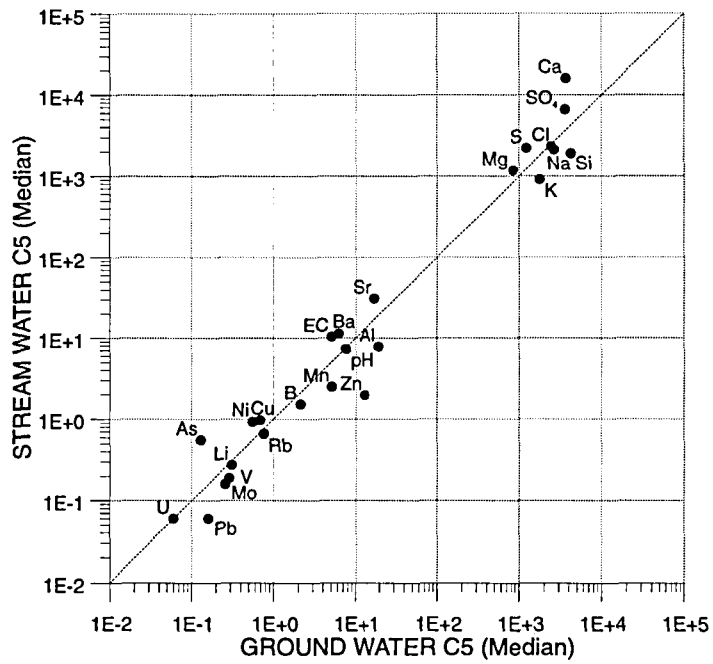


Figure 9.1.4. Median composition of stream water in C5 vs. groundwater in C5 (in mg/l, except for EC, in mS/m, and pH, in pH-units).

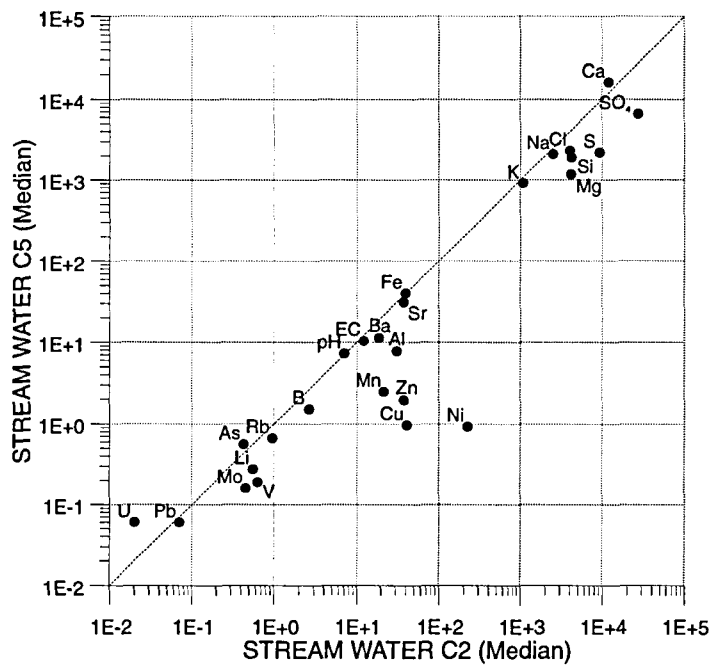


Figure 9.1.5. Median composition of stream water in C5 vs. stream water in C2 (in mg/l, except for EC, in mS/m, and pH, in pH-units).

## 10 CONCLUSIONS

During the year of 1994 the Central Kola Expedition and the Geological Surveys of Finland and Norway carried out a multimedia ecogeochemical study in 8 catchment basins of 12 - 35 km<sup>2</sup> each, spread over a 188000km<sup>2</sup> area in the European Arctic. This naturally sensitive part of Northern Europe encompasses some of the world's most strongly polluted places in the surroundings of the Russian nickel mining and smelting industry on the Kola peninsula. On the other hand it contains still areas virtually untouched by human activities. This contrast contributes to its fascination in a research context.

Four catchments in Russia (C1, C2, C3 and C4), one catchment in Norway (C5) and three catchments in Finland (C6, C7, C8) were chosen to represent the different natural conditions in the survey area as well as different distances to industry. Media sampled in these catchments were snow, rain, stream water, organic stream sediment, overbank sediment, terrestrial moss, other vegetation where moss was missing, topsoil (0-5cm), complete podzol profiles, Quaternary deposits, groundwater and bedrock.

The levels of contamination identified in the eight catchments studied show a clear relationship to the distance to pollution sources, major wind directions and topographical features. C1, C2 and C4 are strongest polluted whilst C3, C5 and C6 show some signs of pollution for some elements. C7 and C8 show background concentration levels. Element concentrations and variations in almost all media are such that regional mapping at the proposed density of 1 sample site per 300 km<sup>2</sup> should result in reliable results.

Snowpack samples, collected at the end of the winter season, are best suited to identifying the suite of elements emitted from any one source. The ratio of element concentrations as measured in snow meltwater to filter residue enables the identification of single pollution sources. Disadvantages of snowpack samples are that their information is valid only for the last winter and that the technique can only be applied to areas with stable snow cover where element redistribution due to thaw events during the winter is minimal.

Rainwater samples collected monthly, give rather similar results to those observed for the meltwater of the snow samples in background areas, not, however, close to industry, where particulate deposition predominates. Logistics of rainwater sampling are rather difficult, limiting its usefulness for regional studies. Results from single points will thus have to be used to calibrate other media in order to obtain a regional overview. Monthly sampling of rainwater allowed to even detect changes in the ore feed from local (Pechenga) to imported (Norilsk) ore in the Monchegorsk smelter.

The results of rainwater and snowpack sampling are combined to calculate yearly average deposition values of heavy metals at different distances from pollution. A shortcoming of our investigation was that we were not able to collect dry deposition during summertime. A method, based on the characteristic changes of the ratios of snow meltwater to filter residue and snow meltwater to rainwater with distance to industry, allowing to calculate annual total deposition data for any wanted point within the survey area, was thus developed. Using this method it is even possible to estimate the relative input from different industrial sources.

The composition of stream water is known to vary strongly with time. Water chemistry is strongly influenced by lithology, thus a rather good knowledge of regional geology is necessary for the interpretation of element levels as observed in surface waters. Nevertheless, in the

catchments close to industry, stream water is so strongly polluted that it would be no problem to use surface waters for regional pollution mapping. For example, all values measured in C2 are at least by a factor of two above the current drinking water action level for Ni. Snow melt results in a strong peak of heavy metals in the stream water of the polluted catchments. For some elements this peak is visible in the stream water of C5, but not in the Finnish background catchments.

Organic stream sediments reflect both, pollution and geology. Their usefulness for environmental studies depends strongly on a good understanding of the local geology. In general, this medium appears to be best suited to application in mineral exploration.

Overbank sediments represent rather well the geogenic background levels throughout the area. Some samples show strongly enriched heavy metal values due to industrial activities. The main pollutants Co, Cu, Ni and S are mobile in a overbank sediment profile near Monchegorsk.

Moss sampling proves to be very effective for the regional mapping of airborne contamination by sulphur and many (but not all) heavy metals, when there is a major pollution source in the area investigated. An obvious disadvantage of mosses for airborne pollution monitoring is their absence at the most heavily polluted sites. For a number of elements, geogenic dust strongly influences the observed element concentrations in the mosses. For others, growth rate and competition in uptake between different elements seem to play a role for the element levels observed.

Compared to lichen and crowberry, moss is the most sensitive bioindicator for heavy metal pollution. For the major pollutants (Co, Cu, Ni) moss, however, seems to reach a saturation level and crowberry may be a better indicator of very high pollution levels. Crowberry could be found even in the most polluted areas.

The detailed study of heavy metal profiles as visible in tree rings by the use of laser ablation ICP-MS shows great variations between trees and within profiles, but no direct correlation with the pollution history of the area.

Topsoil (0-5cm) samples show some of the strongest enrichments of heavy metals in the catchments close to industry. Cu and Ni contents in C2 are up to 600 times higher than in the Finnish background catchments. Many other elements show, however, as large variations within one catchment as between catchments, and topsoil is not an ideal sample medium for low density geochemical mapping. For several elements the observed levels are strongly influenced by the content of organic material in the sample. The content of organic matter must thus be determined when using topsoil samples for regional mapping. Sulphur is not enriched in the topsoil.

Investigations of elements availability and seasonal effects on element contents as observed in topsoil show some striking differences between the Russian catchments and the background areas. Due to the severe degradation on the ecosystems in the catchments closest to industry, high levels of heavy metals are washed out of the catchment with the snow melt at spring time. The available portions of the base cations, important for plant growth and health, are washed out together with the heavy metals and this process leads to a further degradation of vegetation.

Precious metals (gold and platinum group elements) reach very high contents in topsoils in C2, indicating that substantial financial gains could be expected by reducing the smelter's emissions.

Concentrations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were only determined for some few selected samples. Results obtained so far are low and highly variable on the local scale. Rather large composite samples will have to be collected for regional mapping.

Podzol profiles have the greatest accumulation capacity for heavy metals of all media sampled. Podzol profiles offer the opportunity to study element distribution and the effect of anthropogenic contamination not only in two but in three dimensions. Their study allows predictions to be made on the future development of an area. Results obtained so far indicate that the mineral soil layers are generally well protected against heavy metal input from anthropogenic sources if the organic layer is intact. The behaviour of sulphur as the main pollutant, however, gives reason for concern as it does not accumulate in the organic layer but moves directly downwards in the soil profile. These weathering processes and the availability of geogenic heavy metal pools are affected.

In C2 even the groundwater is affected by anthropogenic contamination as can be seen by generally elevated S-contents and peaking Ni-levels some weeks after snow melt.

In summary, the different media used in this study yielded quite different pieces of information that complement each other in understanding the results. Knowledge about the geological composition of the study area proves very important for the correct interpretation of the obtained results.

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