Centre for Economic Development, Transport and the Environment

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State of the terrestrial environment in the joint Finnish, Norwegian and Russian border area on the basis of bioindicators

Final technical report of the Pasvik Environment Monitoring Programme

PASI RAUTIO | JARMO POIKOLAINEN

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STATE OF THE TERRESTRIAL ENVIRONMENT IN THE JOINT FINNISH, NORWEGIAN AND RUSSIAN BORDER AREA ON THE BASIS OF BIOINDICATORS
FINAL TECHNICAL REPORT OF THE PASVIK ENVIRONMENT MONITORING PROGRAMME

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The samples were collected by Heikki Posio and Aarno Niva (Finnish Forest Research Institute Northern Unit; *Metla*), Tatiana Sukhareva and Elena Belova (Institute of the Industrial Ecology Problems in the North of the Kola Science Center in Apatity Russian Academy of Sciences; *INEP*) and Paul Aspholm (Norwegian Institute for Agricultural and Environmental Research, Svanhovd; *Bioforsk*).

The samples from Russian plots were prepared and analyzsed at the laboratory of Terrestrial Ecosystems of the INEP.

Metla's laboratory personnel in Paljakka, Parkano, Rovaniemi and Tikkurila prepared and analysed the moss and foliage samples from Finnish and Norwegian plots.

The samples from the Russian plots were analyzed at the laboratory of Institute of the Industrial Ecology Problems of the North Kola Science Centre, Russian Academy of Sciences in Apatity. The samples from the Finnish and Norwegian plots were analyzed at the laboratories of Finnish Forest Research Institute.

The laboratories participate regularly in international inter-laboratory comparison exercises, with satisfactory results.

Preface

Monitoring of terrestrial ecosystems is a part of the Pasvik Environment Monitoring Programme created by the environmental authorities and researches from the three countries for obtaining comprehensive and current information on the changes taking place under the varying anthropogenic load in the joint border area of the three nations. The main threat to aquatic environments in the border area is the neighbouring Pechenganikel industrial complex, located on the Kola Peninsula in northwest Russia. Emissions from the complex comprise high levels of sulphur dioxide, and particulate material containing a wide range of toxic metals, primarily copper and nickel.

One of the primary aims of the monitoring programme for terrestrial ecosystems is to detect changes in the concentrations and spatial distribution of heavy metal and sulphur dioxide emissions from the smelter complexes.

In the present report results of element concentrations in moss and pine foliage sampled during 2011, on the same sample plots as in the 2003–2006 sampling, are presented.

Study area

The terrestrial ecosystem monitoring network of the Pasvik programme consists of selected plots from the earlier established forest monitoring networks (Derome et al. 2008, Fig. 1). The plots were established in pine forests. In the present study the plots used for moss and pine foliage sampling in the 2003–2006 sampling (Fig. 1): the nine Finnish plots (F-1, F-2, F-3, F-4, F-5, F-6, F-7, F-8 and F-9), the five Norwegian plots (PA, PB, PC, PD and N11) and the two Russian plots (RUS0, RUS1) were sampled again. In addition to these 2004 plots listed above some new plots were included in the present sampling (see following chapters).

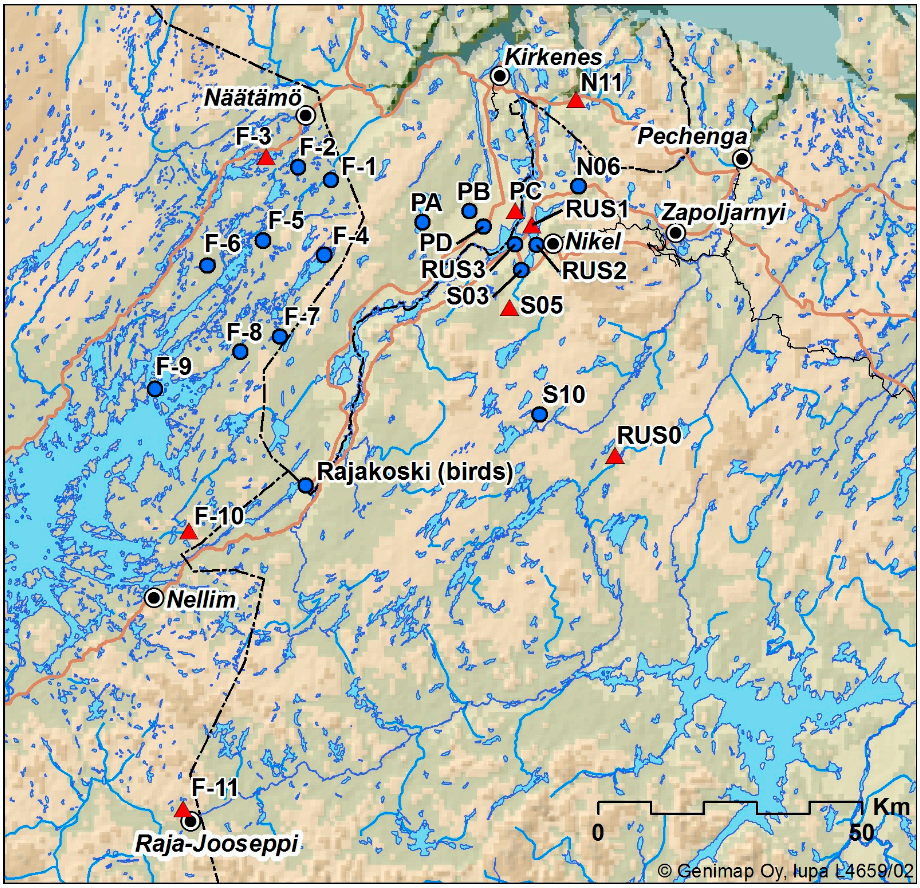


Fig. 1. Location of the Ecosystem Monitoring Network in Russia, Finland and Norway.
▲= Deposition monitoring plots,
● = Ecosystem monitoring plots

Element concentrations in mosses on the plots of the Pasvik programme in 2004 and 2011

Jarmo Poikolainen

Background

The studies of the Pasvik programme in 2003–2006 consisted of many kind measurements related to the terrestrial ecosystem. One research subject was the survey of the element concentrations in plants including also mosses. Carpet forming, ectohydric mosses have been widely used as biomonitors in large-scale, heavy metal and nitrogen deposition surveys (Rühling & Tyler 1970, Zechmeister et al. 2003, Poikolainen 2004, Harmens et al. 2013). Mosses are suitable for this kind of surveys, because they obtain most trace elements and nutrients directly from precipitation and dry deposition. They have also several properties that promote the accumulation of heavy metals. Mosses require small amounts of heavy metals, e.g. zinc (Zn), copper (Cu) and iron (Fe), for their metabolic functioning, but they accumulate also heavy metals, which they do not need for their growth.

Moss samples were collected first in the Pasvik programme in 2004, and the collecting was replicated in 2011. In this report results for heavy metal, sulphur and nitrogen concentrations in mosses in 2011 are presented and compare to the results obtained in 2004.

The moss Pleurozium schreberi, which is widely used to monitor airborne heavy metal pollution, was sampled on the plots located at the distances ranging from 5 km to 79 km from the Nikel smelter. Photograph: Paul Aspholm.

Study area

The moss samples were collected in August 2004 on the nine Finnish plots (F-1, F-2, F-3, F-4, F-5, F-6, F-7, F-8 and F-9), on the five Norwegian plots (PA, PB, PC, PD and N11) and on the two Russian plots (RUS0, RUS1). The new moss samples were collected on the same plots in 2011. Moreover samples were collected on the three Russian plots (N06, S03, S05), where no moss samples were collected in 2004. The plots in 2011 located at distances ranging from 5 km to 79 km from the Nikel smelter.

Material and methods

The moss samples were collected in 2011 according to the guidelines of the European moss survey (Harmens et al. 2008). The moss species, Hylocomium splendens (Hedw.) Schimp and Pleurozium schreberi (Willd. ex Brid.) Mitt., were sampled on the Finnish and Russian plots and only H. splendens on the Norwegian plots. The samples from the Finnish and Norwegian plots were analyzed at Finnish Forest Research Institute´s laboratory in Vantaa. After collection, they were air-dried at +35 °C and litter and other debris removed. The last three full years growth (ca. 2 g/plot) was separated for elemental analysis. Next they were homogenized in a ceramic mill and digested with HNO_3/H_2O_2 in the microwave oven. The concentrations of heavy metals (aluminium (Al), cadmium (Cd), chromium (Cr), Cu, Fe, manganese (Mn), nickel (Ni), lead (Pb) and Zn) and sulphur (S) were determined by ICP-ES and nitrogen using a modified Micro-Kjeldahl method (Kubin & Siira 1980). Quality control of the analyses was ensured by means of moss reference material M2 and M3, prepared for the European moss surveys (Steinnes et al. 1997, Harmens et al. 2013). The samples from the Russian plots were analyzed at the laboratory of Institute of the Industrial Ecology Problems of the North Kola Science Centre, Russian Academy of Sciences in Apatity. The Ca, Mg, K, Fe, Mn, Cu, Ni, Zn concentrations were determined, following microwave-assisted digestion, by ICP or 30 AAS, and S and P photocolorimetrically. Total C was determined by the Tiure method and total nitrogen by the automated Kjeldahl method.

Heavy metals in mosses

The emissions of the Nikel smelter contain first of all nickel (Ni) and copper (Cu). Their concentrations in mosses were very high on the Russian plots in the vicinity of the smelter (Table 1, Fig. 2 and 3). The winds blow in this area mainly from the west and south-west, and so the highest Ni and Cu concentrations were found in 2011 on the Russian plot N06 located 12 km north-east from the smelter (Ni 1 074 mg/kg, Cu 636 mg/kg). Also the concentrations of other heavy metals (Al, Cd, Cr, Fe, Pb, Zn) apart from manganese were highest on this plot (Fig. 4–10). The Ni concentration varied on the Russian plots located 5–7 km west from the smelter (RUS1, S03) between 545–572 mg/kg and the Cu content between 404–452 mg/kg. Also the Fe, Pb and Cd concentrations were relatively high on these plots (Fig. 4, 5 and 6).

The Ni and Cu concentrations were still relatively high on the Norwegian plots (PA, PB, PC, PD, N11). However, they were clearly lower than on the Russian plots, although the nearest plots (PC, PD) located only 8–12 km west from the smelter. The Ni concentration varied on the Norwegian plots between 66–196 mg/kg and the Cu concentration between 44–131 mg/kg (Table 1, Fig. 2 and 3). The concentrations of other heavy metals were not especially high in 2011.

The Finnish plots located already so far from the smelter (42–79 km) that the heavy metal concentrations were there relatively low compared to the concentrations on the Russian and Norwegian plots. The Ni concentrations varied between 11–37 mg/kg and the Cu concentrations between 8–20 mg/kg. The concentrations decreased gradually with increasing distance from the smelter. The concentrations of other heavy metals were relatively low (Fig. 4–10).

Table 1. Heavy metal, sulphur and nitrogen contents in mosses 2004 and 2011 on the plots of Pasvik programme in Finland, Norway and Russia (mean, minimum, maximum) and in the national surveys in 2010/2011 in Finland and Norway (Harmens et al. 2013).

		Al	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn	S	N
Finland												
Mean	2004	166	0,09	1,15	13,6	209	451	18,5	2,73	24,4	801	
	2011	108	0,08	0,49	16,0	148	311	20,5	1,28	19,2	712	0,74
	2010[1]	206	0,12	0,95	4,9	240		2,5	2,04	31,0		0,77
Median	2004	161	0,09	1,02	13,0	174	407	17,4	2,9	23,5	825	
	2011	110	0,09	0,46	15,5	150	309	21,4	1,39	19,7	716	0,73
	2010[1]	187	0,11	0,80	3,9	209		1,2	1,87	29,5		0,70
Min	2004	130	0,06	0,83	8,4	136	319	9,7	1,8	19,4	657	
	2011	85	0,06	0,38	9,5	94	<275	10,5	<0,82	17,0	599	0,64
	2010[1]	44	<0,05	0,34	0,74	53		0,42	<0,75	11,5		0,38
Max	2004	249	0,11	1,84	20,2	363	702	31,4	3,55	33,1	896	
	2011	137	0,11	0,59	26,8	210	436	37,1	1,63	2,03	809	0,85
	2010[1]	958	0,44	14,0	55,1	2 230		88,2	6,57	102		2,06
Norway												
Mean	2004	245	<0,22	1,15	88,4	823	640	145	5,01	48,9	1 123	
	2011	285	0,17	1,32	76,3	525	383	112	2,47	33,8	1 094	0,75
	2010[1]	346	0,12	0,98	6,43	449		5,40	2,29	35,9		
Median	2004	206	0,22	1,09	73,0	740	651	119	4,97	42,2	1 120	
	2011	264	0,19	1,27	75,0	515	399	108	2,49	32,0	1 070	0,75
	2010[1]	283	0,08	0,59	4,04	278		1,16	1,54	30,7		
Min	2004	147		0,82	45,4	429	236	78,0	3,32	36,9	1 010	
	2011	235	0,12	1,00	44,4	344	292	66,2	1,87	27,1	1 000	0,67
	2010[1]	46	0,01	0,16	1,38	27		0,15	0,33	7,40		
Max	2004	434		1,82	145	1 371	1 212	234	6,63	77,2	1 233	
	2011	355	0,23	1,91	131	832	517	196	3,25	41,4	1 270	0,87
	2010[1]	4581	1,87	47,9	443	24 684		857	20,8	368		
Russia												
Mean	2011	286	0,43	7,35	351	2 385	474	511	6,19	45,0	1 727	0,77
Median	2011	297	0,50	6,71	404	2 478	491	545	6,36	39,8	1 753	0,84
Min	2011	134	0,08	3,38	26,1	300	129	44,6	1,63	28,3	1 096	0,58
Max	2011	394	0,79	12,6	636	5 019	886	1 074	9,15	78,3	2 326	0,94

1) Concentrations in mosses in Finland 2010–2011 (Harmens et al. 2013)
2) Concentrations in mosses in Norway 2010–2011 (Harmens et al. 2013).
NB: Maximum Al concentration is near the aluminium factory and maximum Fe concentration near the iron and steel factory.

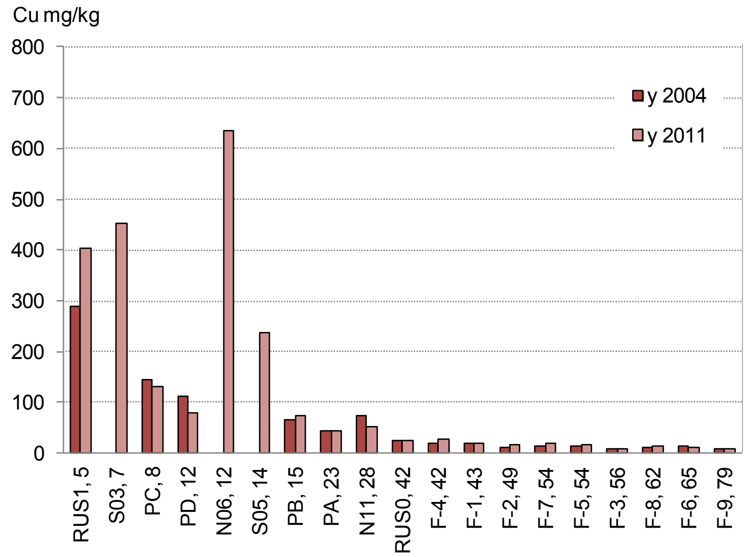


Figure 2. The Cu concentrations in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

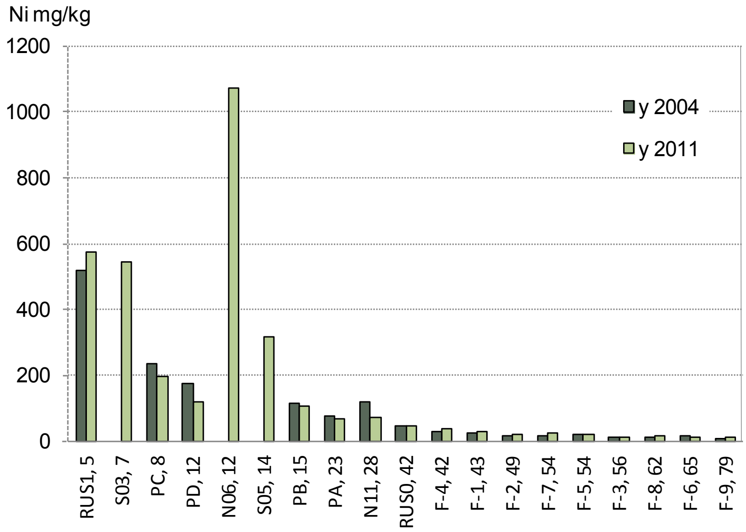


Figure 3. The Ni concentration in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

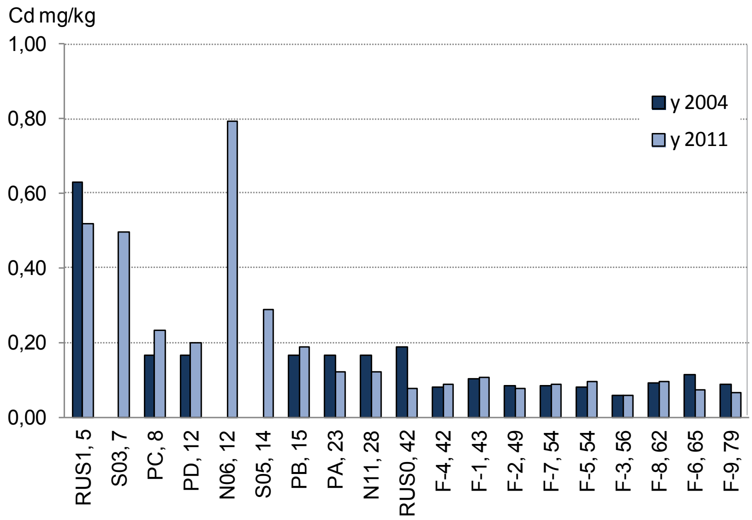


Figure 4. The Cd concentration in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

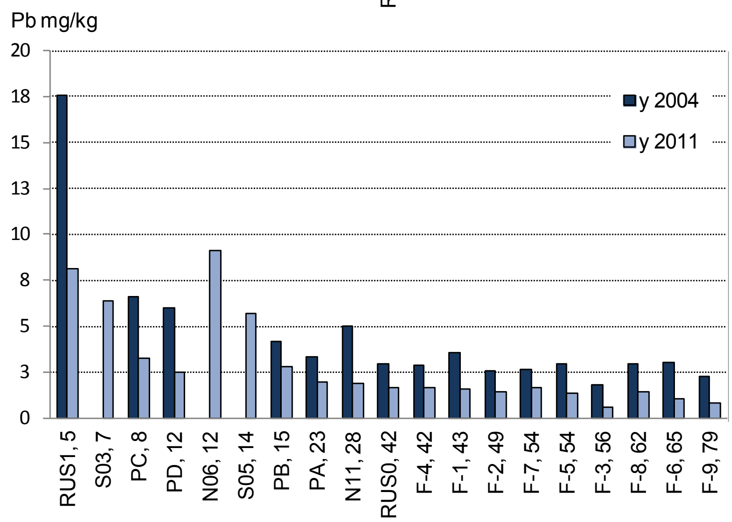


Figure 5. The Pb concentration in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

The Ni and Cu concentration in mosses were in 2011 on the plots of the Pasvik programme almost at same level than in 2004 (Table 1, Fig. 2 and 3). The concentrations even increased in the vicinity of the smelter. Their mean concentration has decreased a little bit on the Norwegian plots, but not in the Finnish plots. This suggests that the Ni and Cu emissions of the Nikel smelter have not decreased at all during the last 10 years. Contrary to Cu and Ni, Pb concentrations have decreased clearly on all plots (Fig. 5). The decrease is probably partly a result of the decrease in the emissions of the road traffic. Also Fe, Zn and Mn concentrations have decreased on most of the plots (Fig. 6, 8, 10). Al and Cr concentrations have decreased on the Finnish plots, but generally increased on the Norwegian and Russian plots. The differences can be a result of the different analysis methods in 2004 in Norway, Russia and Finland.

According to the concentrations in the mosses, most of the heavy metals emitted from the Nikel smelter are deposited at distances of less than 15 km. The deposition is greatest east and east-north from the smelter because the winds blow in this area mainly from the west and south-west. The Ni and Cu concentrations are very high in the Russian plots and high in the Norwegian plots compared to the mean concentrations in the European moss survey in 2010/2011 (Harmens et al. 2013). The mean Ni content in this survey was in Finland 2,45 mg/kg and in Norway 5,40 mg/kg, and the mean Cu contents respectively 4,90 mg/kg and 6,43 mg/kg.

The heavy metal concentrations in mosses do not directly reflect the total deposition of heavy metals. There are differences in the accumulation of heavy metals in mosses, and the concentrations in mosses are also affected by factors other than atmospheric pollution. Soil dust and the condition of the mosses can be a significant effect on the heavy metal concentrations in the vicinity of the smelter. The mosses are there saturated by heavy metals and metals have stratified mostly on the surface of mosses.

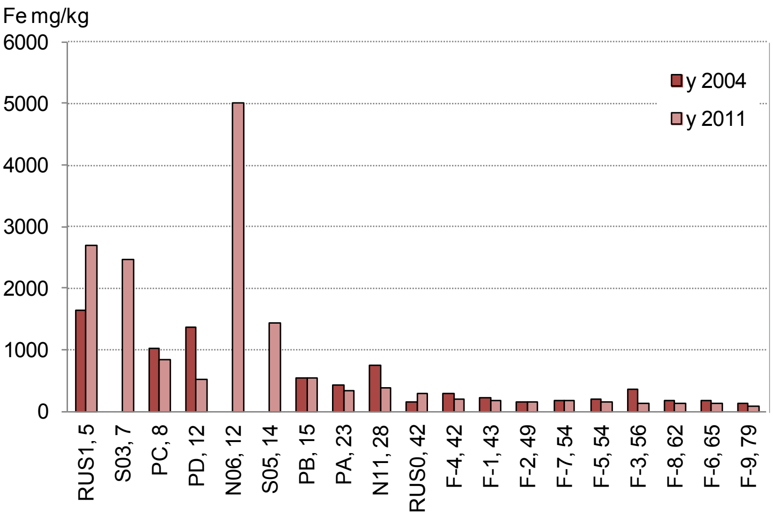
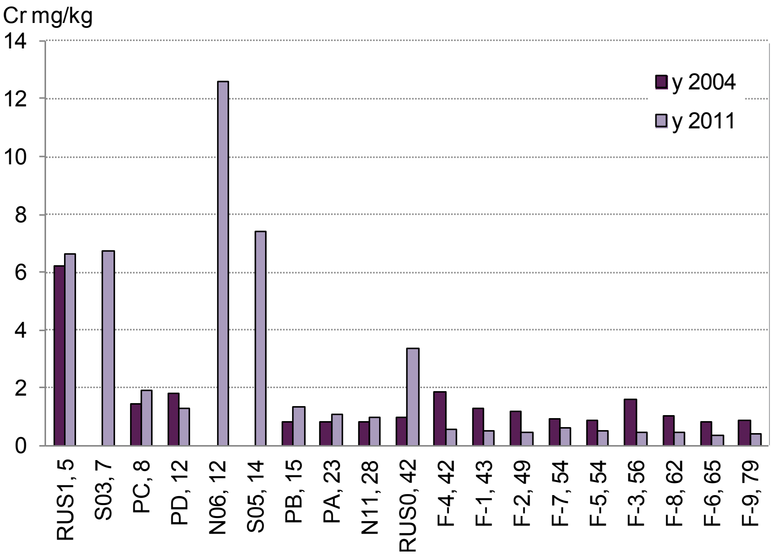


Figure 6. The Fe concentration in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.



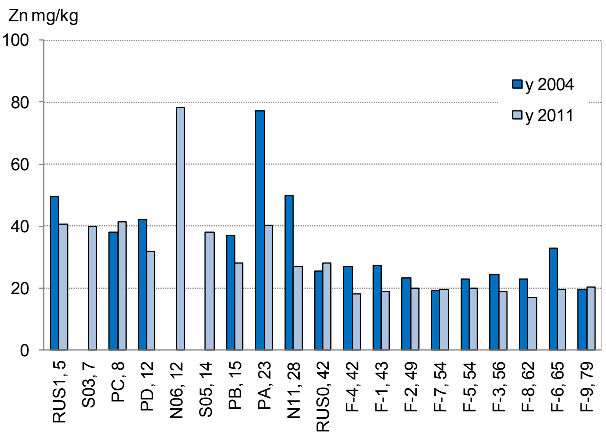
Figures 7. The Cr concentration in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

Sulphur and nitrogen in mosses

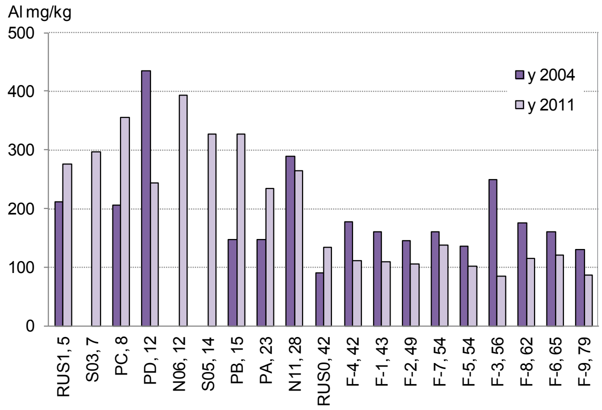
The sulphur concentrations in mosses were in 2011 high (1 753–2 326 mg/kg) near the Nikel smelter (Table 1, Fig. 11). The highest concentration was found on the Russian plot (N06) located 12 km north-east from the smelter. The concentrations decreased clearly with increasing distance from the smelter. They were on the Norwegian plots between 1 000–1 270 mg/kg and on the Finnish plots between 599–809 mg/kg, which are already close to the so-called background levels (Table 1). The sulphur concentration in mosses was on average in 2011 on the plots of Pasvik programme approximately at same level than in 2004. The concentrations decreased slightly on the Finnish plots, but they increased a little bit on the plots near the smelter.

The sulphur concentrations in mosses indicate moderately elevated sulphur deposition in the study area. The mosses are not considered generally to be especially good biomonitors of sulphur deposition (Mäkinen 1994, Äyräs et al. 1997). The reason may be that sulphur at high concentrations damages the mosses and alters their accumulation capacity. There is also natural variation in the sulphur concentration of plants in the study area due e.g. maritime climate (Kashulina et al. 2003).

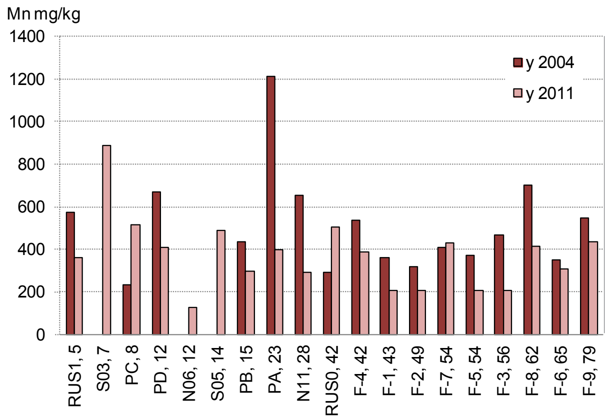
The nitrogen concentration varied in 2011 on the plots between 0,64–0,94 %, and they were highest in the vicinity of the smelter (Fig. 12). These concentrations are not particularly high compared to the concentrations in the European moss survey in 2010/11 (Harmens et al. 2013). N concentrations varied in this survey in Finland between 0,38–2,06 % and they were in northern Finland generally less than 0,60 %.



Figures 8. The Zn concentration in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.



Figures 9. The Al concentration in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.



Figures 10. The Mn concentration in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

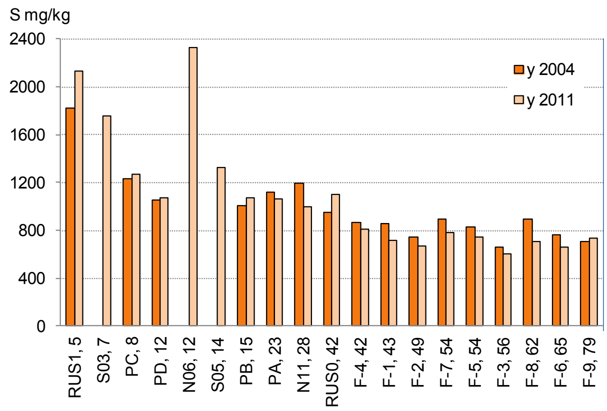


Figure. 11. The sulphur concentration in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

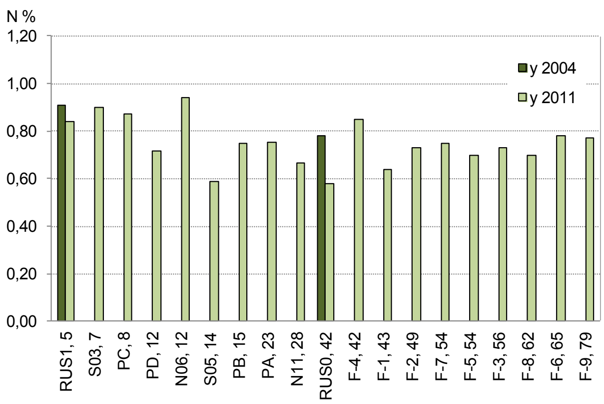


Figure. 12. The nitrogen concentration in mosses in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

Element concentrations in pine oliage on the plots of the Pasvik programme in 2004 and 2011

Pasi Rautio

Background

Sulphur (S) and heavy metals, mainly copper (Cu) and nickel (Ni), are the main pollutants affecting the environment in North-West-Russia. In addition to the direct toxic effect of these pollutants on trees, they can also reduce the availability of macro- and micronutrients as a result of leaching from the soil and from the tree foliage (Lukina and Nikonov 1998, Aber et al., 1989, Darrall 1989, Innes 1995). Disturbances in the nutrient status of the soil take place when acidifying compounds (e.g. SO_2/sulphuric acid, NO_x/nitric acid) displace important macronutrients such as calcium and magnesium (as cations, Ca2+, Mg2+) on the cation exchange sites in the soil (Zoettl and Huettl 1991). However, in the region of the present study area cidifying nitrogen deposition is relatively low. Acidification of the soil may also increase the concentrations of soluble aluminium species (e.g. Al3+) which, in turn, have antagonistic effects with other cations (e.g. Ca), thus reducing the uptake of important nutrient elements (Evers and Huettl 1991). In the present study we studied the chemical composition of Scots pine (Pinus sylvestris L.) foliage in order to assess the spatiotemporal variation in the concentrations of heavy metal and sulphur pollutants, and to assess the possible impact of the pollutants on the nutrient status of the trees. Determination of the element concentrations of tree foliage is one of the most common methods for monitoring forest vitality, assessing the impacts of pollution, and determining the nutrient status of trees (Mitrofanov 1977, Il'in 1991, Huttunen et al. 1985, Helmisaari 1992, Tikkanen and Raitio 1990, Huettl and Fink 1991, Brække 1996, Rautio 2000).

The chemical composition of Scots pine (Pinus sylvestris L.) foliage was studied in order to assess the temporal variation in the concentrations of heavy metal and sulphur pollutants. Photograph: Paul Aspholm.

Material and methods

A total of 19 sampling plots in pine stands were investigated: 9 in Finland, 5 in Russia, and 5 in Norway (Fig. 1). Needle samples were collected during the dormant period (September-October) 2011. Pine branches (including needles) were taken in the upper third of the tree crown on 3–5 trees and then pooled to form one sample per plot. The needle samples were divided into current year's needles (needles formed during 2011 growing period, below these are referred to as current-year or C-needles) and previous year's needles (2010 formed needles, below these are referred to as previous-year or C+ 1-needles) and analysed as such (for details see Rautio et al. 2010). The Ca, Mg, K, Fe, Mn, Cu, Ni, Zn concentrations were determined, following microwave-assisted digestion, by ICP/OES or AAS, and S and P by ICP/OES or photocolorimetrically. Total C was determined on a CHN analyser or by the Tiure method, and total nitrogen on a CHN analyser or by the automated Kjeldahl method. The methods used by the laboratories in Finland and Russia differed to some extent (samples collected in Norway were analysed by Finnish laboratory), but the laboratories participate regularly in international inter-laboratory comparison exercises, with satisfactory results.

Table 2. Element concentrations in current (C) and previous-year (C+1) Scots pine needles collected in the 19 sample plots in northern Finland, Norway and Russia in 2011. Values marked with < are below quantification limit.

Plot	Distance from Nikel	Age class	Al	B	Ca	Cr	Cu	Fe	K	Mg	Mn	Ni	P	S	Zn	N	C	Pb	Cd
			mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	%	%	mg/kg	mg/kg
Rus1	5,2	C	288		3,30	0,79	41,9	211	6,6	1,33	736	53,5	1,83	1 039	26,1	1,36	56	1,14	0,157
Rus1	5,2	C+1	340		4,68	1,31	52,1	359	5,2	1,19	968	71,4	1,52	1 157	24,0	1,21	58	2,17	0,179
SO3	7,0	C	250		2,99	0,59	36,3	168	6,3	1,28	331	46,1	1,76	1 823	35,2	1,45	54	1,03	0,880
SO3	7,0	C+1	297		4,32	1,31	57,8	346	5,0	1,22	453	82,0	1,41	1 803	39,5	1,18	54	2,45	0,112
PC	8,1	C	193	11,3	1,81	0,32	8,0	38	5,0	0,96	368	12,7	1,36	989	26,3	1,30	53	<1,06	<0,07
PC	8,1	C+1	227	10,5	2,91	0,39	11,6	62	4,1	0,97	552	14,6	1,17	894	25,9	1,19	54	<1,05	<0,07
PD	11,9	C	203	9,36	2,13	0,26	6,6	35	4,4	1,35	444	10,7	1,51	1 080	38,8	1,44	53	<1,06	<0,07
PD	11,9	C+1	234	8,02	3,15	0,45	8,8	56	3,4	1,21	579	11,2	1,26	994	39,7	1,26	54	<1,05	<0,07
N06	12,3	C	247		2,64	0,81	43,3	216	6,2	1,23	474	59,0	1,79	1 502	23,9	1,43	54	1,21	0,074
N06	12,3	C+1	297		3,59	1,45	58,4	402	5,0	1,12	678	90,2	1,49	1548	23,5	1,31	55	3,68	0,090
SO5	14,1	C	404		2,70	0,33	20,0	83	5,7	1,33	623	30,8	2,17	1 443	37,8	1,58	56	0,57	0,070
SO5	14,1	C+1	550		4,12	0,75	33,6	163	4,8	1,28	952	44,7	1,82	1 714	39,6	1,34	57	1,13	0,090
PB	15,3	C	252	12,9	1,86	0,24	6,6	37	4,7	1,16	323	10,7	1,51	1 070	29,7	1,53	54	<1,06	<0,07
PB	15,3	C+1	280	13,6	3,18	0,29	8,7	57	3,7	1,27	539	10,8	1,30	991	32,9	1,34	54	<1,06	<0,07
PA	23,3	C	178	13,3	2,24	0,25	5,6	36	5,3	1,16	291	7,7	1,64	976	39,5	1,49	53	<1,06	<0,07
PA	23,3	C+1	212	13,6	4,05	0,35	6,9	52	4,8	1,24	486	8,5	1,44	1 040	59,5	1,47	53	<1,06	0,085
N11	28,4	C	157	16,1	2,19	1,42	14,6	76	6,0	1,17	381	17,3	1,90	1 200	42,6	1,64	53	<1,06	0,074
N11	28,4	C+1	189	14,8	3,38	0,96	18,4	121	5,4	1,03	620	21,2	1,70	1 180	47,3	1,62	54	<1,06	0,085
Rus0	42,2	C	234		2,46	0,16	5,0	46	4,8	1,35	482	10,6	1,71	1 020	42,3	1,33	56	0,13	0,040
Rus0	42,2	C+1	269		3,85	0,36	7,5	76	4,0	1,30	705	11,9	1,43	962	46,0	1,16	55	0,25	0,045
F-4	42,3	C	206	8,62	1,83	0,23	6,0	28	5,7	1,23	253	5,8	1,60	1 010	41,4	1,46	54	<1,69	<0,113
F-4	42,3	C+1	235	7,52	2,91	0,42	6,6	42	4,3	1,18	382	6,2	1,18	924	48,8	1,21	55	<1,62	<0,108
F-1	42,7	C	279	16,2	1,85	<0,23	5,0	31	4,7	1,24	215	5,4	1,49	873	41,4	1,38	54	<1,71	<0,114
F-1	42,7	C+1	380	16,0	2,94	0,23	4,4	42	4,7	1,43	384	4,9	1,42	859	57,7	1,29	55	<1,61	<0,107
F-2	49,4	C	235	15,0	1,40	<0,22	4,1	24	5,8	1,16	165	2,9	1,56	897	39,8	1,37	54	<1,68	<0,112
F-2	49,4	C+1	328	15,1	2,42	0,37	3,5	35	4,7	1,14	272	2,2	1,26	851	54,2	1,23	55	<1,66	<0,111
F-7	53,7	C	151	11,2	1,98	0,22	5,2	28	5,2	1,12	360	4,6	1,61	990	45,9	1,33	54	<1,64	<0,109
F-7	53,7	C+1	199	10,7	3,37	0,39	6,0	39	4,4	1,16	666	5,0	1,24	945	71,2	1,21	55	<1,66	<0,111
F-5	54,0	C	198	7,40	1,23	0,23	5,0	24	6,0	1,04	229	4,9	1,51	864	42,3	1,30	54	<1,69	<0,112
F-5	54,0	C+1	246	6,63	2,04	0,79	5,0	40	4,3	1,04	343	4,8	1,20	855	49,8	1,15	55	<1,60	<0,106
F-3	55,8	C	217	12,0	2,03	0,23	4,8	33	5,6	1,35	396	5,2	1,82	991	44,4	1,46	55	<1,69	<0,112
F-3	55,8	C+1	273	9,00	3,03	0,57	3,9	45	4,4	1,32	634	4,0	1,47	910	51,4	1,26	56	<1,63	<0,108
F-8	61,7	C	98,5	11,3	1,96	0,38	5,1	29	5,3	0,98	547	3,6	1,37	960	48,2	1,34	55	<1,68	<0,112
F-8	61,7	C+1	132	13,4	3,19	0,31	5,0	39	4,6	1,05	936	3,6	1,18	919	70,6	1,22	55	<1,66	<0,111
F-6	65,0	C	211	20,8	1,69	0,45	4,2	26	6,0	0,88	319	2,8	1,36	887	43,4	1,20	54	<1,68	<0,112
F-6	65,0	C+1	261	22,9	2,95	0,39	3,8	34	5,1	0,84	602	2,3	1,14	839	60,1	1,11	55	<1,67	<0,112
F-9	79,3	C	141	17,6	1,83	0,38	4,6	26	5,7	1,02	275	3,4	1,59	936	35,7	1,28	54	<1,71	<0,114
F-9	79,3	C+1	233	22,5	3,34	0,31	5,1	42	4,8	1,04	540	3,7	1,30	928	51,4	1,13	55	<1,67	<0,111

Chemical composition of pine needles

Maximum accumulation of heavy metals in current-year pine needles occurred at a distance of <15 km from the smelter and the concentrations generally decreased exponentially with increasing distance from Nikel. On the other hand, also the prevailing wind direction had a clear impact on the foliar concentrations. The predominant wind direction in the Paz river valley is from the south-southwest (Bekkestad et al. 1995, Hagen et al. 2006), which means that plots located to the north of Nikel (e.g. N06) receive very high amounts of deposition. As an example, plots PD (in Norway) and N06 (in Russia) are equidistant from the Nikel smelter, but still the foliar Ni concentrations in trees on plot N06 are manifold compared to PD (Fig. 13).

The impact of smelter is the most distinct in case of foliar Ni concentration as in the most severely polluted area C+1-needles had up to 30-fold higher concentrations than those collected on plots in "background" part of the study area (> 45 km from the smelter). The spatial patterns for the foliar Cu and Fe concentrations resembled those of Ni (Fig. 14 and 15). For all these elements the concentrations were exceptionally high at plot N06 (12 km NE of the smelter). In general, the concentrations of these heavy metals were lower in current-year needles than in previous-year needles, suggesting that these elements accumulate in the needles over time. A noteworthy fact is also much higher foliar Cu concentration in Russian side in foliage (C-needles) collected in 2011 compared to foliage collected in 2004. In case of Ni this pattern is seen only on the plot closest to the Nikel smelter (plot Rus1, Fig. 13). As moss samples were not collected in most of these plots in 2004 it is difficult to say if increased foliar Cu concentrations are due to increased deposition, but higher Cu concentration in mosses collected in plot RUS1 in 2011 compared to 2004 samples (Fig. 2) would suggest to this direction. In addition to Cu and Ni some indications for higher concentrations of other heavy metals (Cd, Cr and Pb) in plots close to Nikel smelter were seen (Table 2) but these elements were found in quite low concentrations and in many cases below the quantification limit.

The S concentrations in needles were more constant over the study area than Cu and Ni concentrations. In three Russian plots (S03, N06 and S05) foliar S concentrations raised well above the other plots (Fig. 16). In addition to these plots only in few plots the concentration raised above 1 000 mg/kg that can be considered slightly elevated level, and even there the levels were only slightly higher than on plots that are 60–80 km away from the smelter. In the cold climate that prevails in the study area sulphur that is emitted in combustion processes as sulphur dioxide, remains long in gaseous form before deposition, hence sulphur emissions spread over a large area and clear relation to distance as in case of Cu and Ni cannot be observed in case of S (Fig. 16). Furthermore, sulphate originating from marine sources confounds the effects of SO_2 emissions from the smelters. There is nevertheless a constant increase in S concentrations in C-needles from 2004 to 2011 throughout the study area (Fig. 16), that would suggest that at least part of the increase is due to natural sources.

There was no clear spatial or temporal trend in the foliar Al concentrations (Fig. 17) as on some plots foliar concentrations were higher in 2004 than in 2011 and on some plots the situation was the opposite.

Collecting of needle samples at plot N06 (12 km NE of the smelter). Photograph: Tatina Sukhareva.

Also no clear relation to distance from the smelter was could be seen. Al is common element in soil, hence large part of the Al found in the foliage are due to soil dust.

Other measured elements do not show apparent relation to the vicinity of the Nikel (Table 2), though some indications for higher foliar Ca concentrations in the vicinity of smelters were seen but the differences between plots are not large. Ca is also common element in soil; hence soil dust can contribute to the elevated concentrations as particles deposited on needle surfaces (Rautio et al 1998). There is also some indication for elevated exchangeable Ca concentrations in the organic layer on the plots in the vicinity of Nikel smelters (Derome et al 2008). Consequently higher foliar Ca concentrations observed here can also originate from enhanced availability of Ca in the root layer.

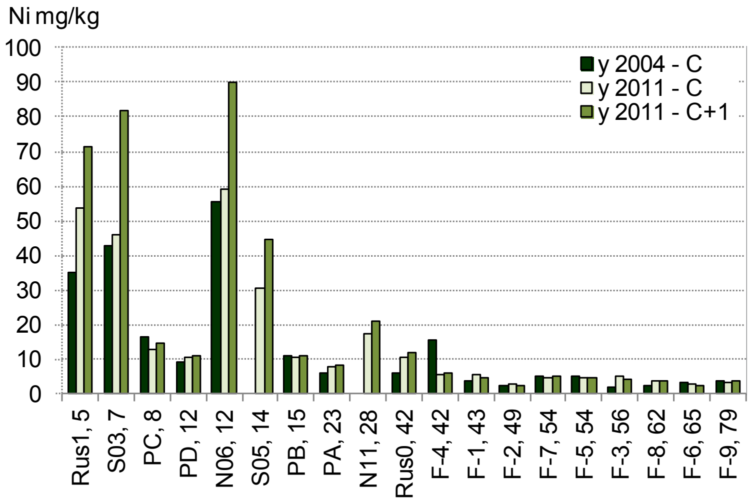
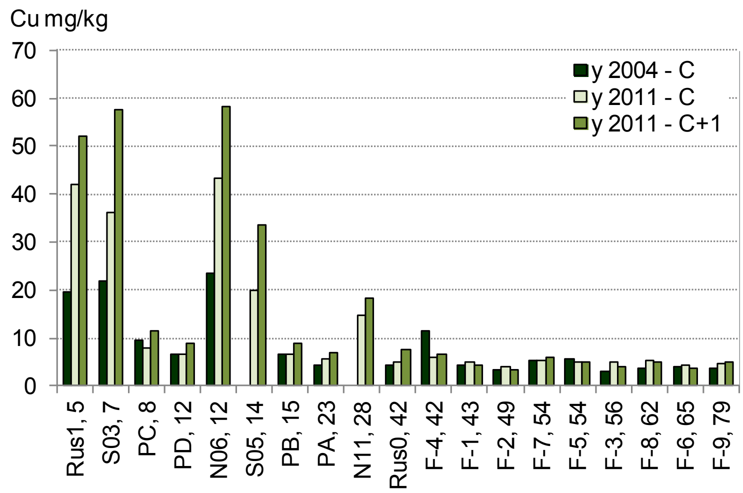
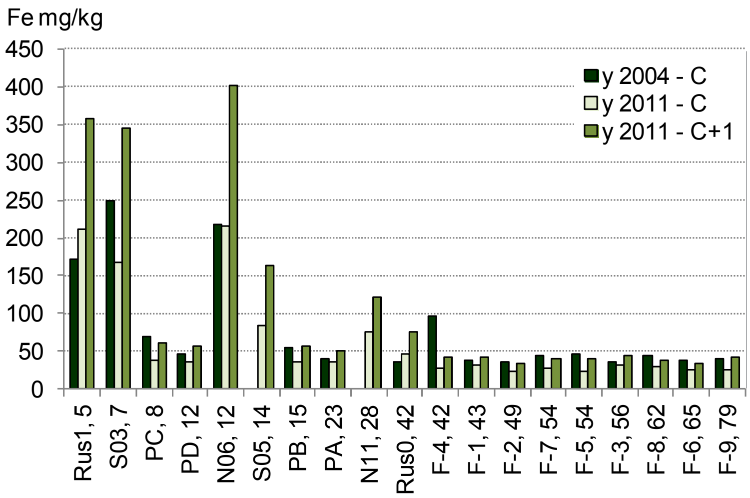


Figure 13. Nickel concentrations in current-year needles (C) and previous-year needles (C+1) collected in 19 sample plots in 2004 and 2011. The number after plot number is the distance (km) from Nikel.



Figures 14. Copper (Cu) concentrations in current-year needles (C) and previous-year needles (C+1) collected in 19 sample plots in 2004 and 2011. The number after plot number is the distance (km) from Nikel.



Figures 15. Iron (Fe) concentrations in current-year needles (C) and previous-year needles (C+1) collected in 19 sample plots in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

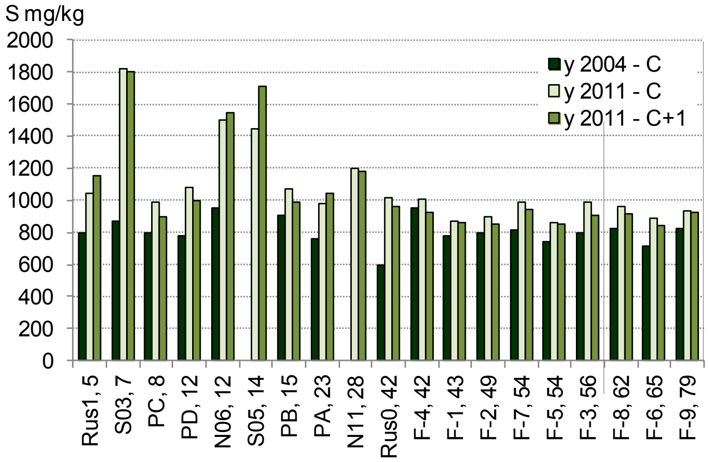


Figure 16. Sulphur concentrations in current-year needles (C) and previous-year needles (C+1) collected in 19 sample plots in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

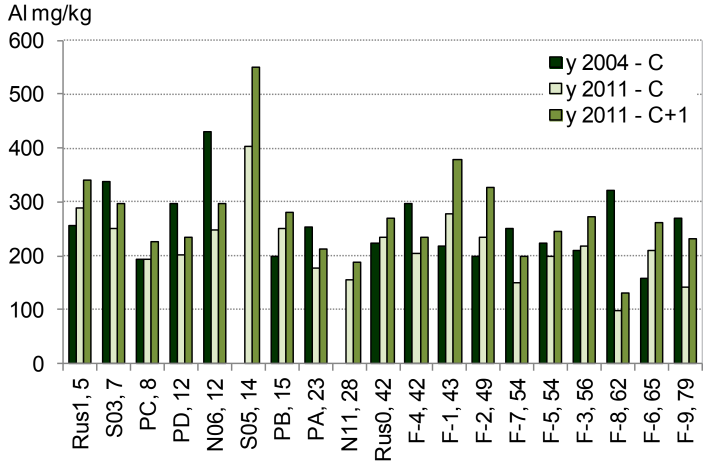


Figure 17. Aluminium concentrations in current-year needles (C) and previous-year needles (C+1) collected in 19 sample plots in 2004 and 2011. The number after plot number is the distance (km) from Nikel.

Conclusions

- In mosses sulphur, copper and nickel concentrations increased clearly towards Nikel smelters
- In foliage samples Cu and Ni followed the same pattern as in mosses but S was not as clearly related to the distance from Nikel smelters
- In addition to the distance to the source the prevailing wind direction played a clear role in sulphur and metal concentrations: in mosses Cd, Cr, Cu, Fe, Ni, S and Zn concentrations and in pine foliage Cr, Cu, Fe, Pb and Ni concentrations were at the highest on the plot N06 that is about 12 km downwind of Nikel
- In foliage samples increasing temporal trend (2004 vs. 2011 collection) was seen for Cu, Ni and S, but in mosses this trend is difficult to evaluate, because moss samples were not collected in some of the plots were highest concentrations were measured in 2011

Recommendations for the future monitoring

- Monitoring efforts in the future should take full advantage of the existing monitoring network to secure the comparability to the historical data
- Different parameters (e.g. moss, foliage etc. samples) should be collected on the same plots to facilitate interpretations
- In case monitoring network is expanded in the future, it is recommended to take the prevailing wind direction into account (e.g. denser sampling downwind of the smelter)
- A special care must be taken to harmonize sampling and analyze methods as far as possible, and in case different methods are used to secure the comparability

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