The effects of nickel smelters on water quality and littoral fish species composition in small watercourses in the border area of Finland, Norway and Russia

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The Pechenganickel smelters in Nikel and Zapoljarnyi, northwestern Kola Peninsula, have been among the world's largest point sources of SO_2, Ni and Cu emissions. In order to examine the effects of airborne emissions, the water quality of 35 small lakes and brooks in Finland, Norway and Russia, 1–50 km from the smelters, was surveyed, and fish of stony shores of these lakes and brooks were sampled by electrofishing in 2004 or 2005. The results demonstrated that in the study area the airborne emissions of the smelters have not caused any widespread damage to fish populations even in the most sensitive small waters. The small waters close to the smelters (roughly within a 10 km radius) are well buffered against the effects of high sulphate deposition. Extremely high concentrations of the heavy metals Ni and Cu, however, are a local threat to biota in small waters there, and a few lakes that have apparently lost their fish populations were found. In the border area of Russia, Norway and Finland, acidification is currently only a problem in some very small and sensitive waters located in the local highland areas, 15–50 km from the smelters. There, lakes with a low buffering capacity (alkalinity < 0.05 mmol l^{-1}) can be found and some fish populations, mainly minnows, have probably been lost. The SO_2 emissions from the smelters have declined to approximately one third of the maximum level in the late 1970s, and this can be seen in a general recovery of the buffering capacity of small lakes such as those in the Finnish border region, 40–50 km west of the smelters.

Introduction

Major metallurgic industries are located in the northwestern Kola Peninsula, close to the border area of Russia, Norway and Finland. The Pechenganickel smelters in Nikel and Zapoljarnyi have been among the world's largest point sources of SO_2 emissions (Gunn *et al.* 1995). The smelters were constructed to process local ores and have been in operation since 1932 and 1955, respectively. Since 1971, the smelters have processed copper and nickel ores from Norilsk, central Siberia, which have a particularly high sulphur content (Kashulin *et al.* 2001). Annual SO_2 emis-
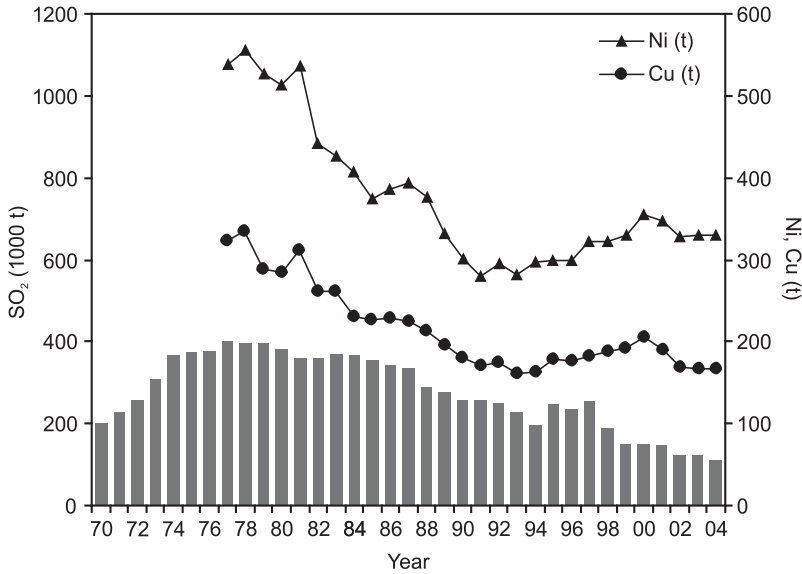
Fig. 1. Annual emissions of SO_2, Ni and Cu from the Pechenganickel smelters (Nikel and Zapoljarnyi) during 1970–2004. Data obtained from the Pechenga nickel company.

sions from the Pechenganickel smelters peaked at around 400 million kg in the late 1970s. Thereafter, emissions began to decline, being at the beginning of the 2000s around 100–150 million kg annually (Fig. 1).

In addition to sulphur deposition, high heavy metal deposition is a serious threat to the environment, with nickel and copper being the main heavy metal pollutants in the area. The highest annual Ni and Cu emissions, 555 tonnes and 335 tonnes, respectively, were reported in 1978. The emissions of not only SO_2 but also heavy metals have decreased since the 1970s. At the beginning of the 2000s, the annual Ni and Cu emissions from the Pechenganickel smelters were around 350 and 180 tonnes, respectively (Fig. 1).

Severe damage to the terrestrial ecosystem of this area can be detected by satellite imagery (Tommervik *et al.* 1995) and the area severely impacted by air pollution was more than 5000 km² in 1988. Terrestrial and aquatic ecosystems have similarly been affected by SO_2 and heavy metal emissions from Sudbury smelters in North America (Gunn *et al.* 1995, Keller *et al.* 1999), where the annual SO_2 emissions at the beginning of the 1960s reached 2500 million kg and the estimated number of acid-damaged lakes was as high as 7000 (Gunn *et al.* 1995). In the Sudbury area, however, there was a 90% reduction in SO_2 emissions between 1960 and 1985, and emission control has been shown to have a positive effect

on water quality and fish populations (Gunn *et al.* 1995, Keller *et al.* 1999).

The sulphur emissions from Pechenganickel smelters are known to cause surface water acidification in the Norwegian–Russian border area (e.g. Nost *et al.* 1991, Moiseenko 1995), but the extent of the acidification problem in the Kola region is still unclear (Kashulina *et al.* 2003). Reimann *et al.* (2000) argued that the emissions "do not seem to present a major threat on a regional scale". Acid-induced damage to fish populations has mainly been studied in the Norwegian Jarfjord area, 30 km north-east of the Nikel and Zapoljarnyi smelters. There, the extent of damage has been small, as only 14 Arctic char (*Salvelinus alpinus*) or brown trout (*Salmo trutta*) populations of small (3–15 ha) lakes were identified as either lost or in various stages of extinction (Hesthagen *et al.* 1998). Surveys conducted in the Finnish border region, 40–50 km west of the smelters, demonstrated that the buffering capacities of a set of small (3–6 ha) lakes was below 0.05 mmol l⁻¹ and an electrofishing survey revealed signs of acid-induced damage to minnow (*Phoxinus phoxinus*) populations (Lappalainen *et al.* 1995, Tammi *et al.* 2003). In the industrial areas close to the smelters, however, the buffering capacity of lakes is generally high (> 0.2 mmol l⁻¹) due to natural factors (e.g. bedrock geology) and local dust emissions containing buffering particles (Moiseenko 1995, Reimann *et al.* 1999).

Heavy metals have received considerable attention due to their toxicity to aquatic biota (Mance 1987), and the toxic effects of heavy metals may influence physiological functions, growth rates, reproduction and mortality. According to Moiseenko *et al*. (1995), the area of high nickel and copper concentrations in water and lake sediments is limited to a 30-km zone around the Pechenganickel smelters, where the range of observed concentrations in lake water was 10–145 μg l^{-1} for Ni and 1–117 μg l^{-1} for Cu. Moiseenko *et al*. (1995), Amundsen *et al*. (1997), Moiseenko and Kudryatseva (2001) and Lukin *et al*. (2003) investigated the contamination and effects of heavy metals, especially Ni and Cu, on local fish populations, mainly whitefish (*Coregonus lavaretus*), brown trout and Arctic char in the large basins of the Pasvik River system or from other lakes in the Kola Peninsula. The general conclusion has been that the prevalence of fish diseases and abnormalities in organs, particularly in the kidney and liver, has been very high (around 90%) in waters with high Ni and Cu concentrations.

The main objective of this study was to provide an overview of the present effects of airborne pollution on water quality and littoral fish populations in small headwater lakes and brooks, which are considered to be the most sensitive waters to anthropogenic effects. The study area covered the industrial area around the Nikel and Zapolarnyi smelters and extended 40–50 km west over the Norwegian and Finnish border areas. In earlier fish surveys in Norwegian and Russian areas (Hesthagen *et al*. 1998), standard gill net series were used. In this study, however, we used electrofishing sampling, enabling us to effectively sample fish even in very small waters where the typical species, such as minnow, small-sized burbot (*Lota lota*) and juvenile brown trout, can hardly be sampled by standard gill net series. Another objective was to demonstrate possible trends in water quality and fish populations in Finnish territory at study sites that were previously surveyed in 1993 and 2000 and where some signs of recovery were already observed (Tammi *et al*. 2003). The third objective of this study was to briefly evaluate the usefulness of the survey methods for possible long term monitoring of the effects of airborne pollution on small waters in the region.

Material and methods

The study area was located in the border area of Russia, Norway and Finland, and was divided into three sub-areas (Fig. 2). Sub-area A was located in Vätsäri, an uninhabited area in Finnish territory, 40–50 km west of the smelters. According to results interpolated from the data of Paatero *et al*. (2006), the annual sulphate (SO$_4$) deposition during 2005 in that area was 350 mg m^{-2}, and depositions of the most abundant heavy metals, Ni and Cu, were 1.2 mg m^{-2} and 1.8 mg m^{-2}. Sub-area B consisted of two local upland areas in Norwegian territory, Sametfjället and Brannfjället, located 20–30 km west of the industrial area, with respective annual sulphate, Ni and Cu depositions in 2005 of 430 mg m^{-2}, 3 mg m^{-2} and 4 mg m^{-2}. Sub-area C was located in the industrial area, between the Nikel and Zapolarnyi smelters and receives the highest sulphate and heavy metal deposition in the Kola region. In 2005, the estimated annual sulphate deposition was 2000–3000 mg m^{-2}, and Ni and Cu depositions were 40–300 mg m^{-2} and 40–200 mg m^{-2}, respectively, depending on the distance from the smelters. The climate in the region is very harsh with long winters and 6–8 months with continuous snow cover. Thus, airborne deposition accumulates as snowpack in winter, and pollutants are released into lakes and brooks during the spring flood.

The objective in the final selection of lakes and brooks inside the sub-areas was to identify waters that are likely to be the most sensitive to airborne pollution. Thus, attention focused on small (2–14 ha) headwater lakes and small (width 1–5 m) brooks running from small headwater lakes. All selected lakes needed to be deep enough (> 3 m) for fish to survive during the winter, while both lakes and brooks needed suitable stony habitats which are important for littoral fish species and where the electrofishing method can be used effectively. Due to the relatively large differences in altitude within a short distance, natural obstacles to migration are common and the fish populations of most of the lakes sampled were isolated from each other.

In sub-area A (Finland), eight small (2–6 ha) lakes, three brooks between small (21–34 ha) lakes and five brooks or outlets of small
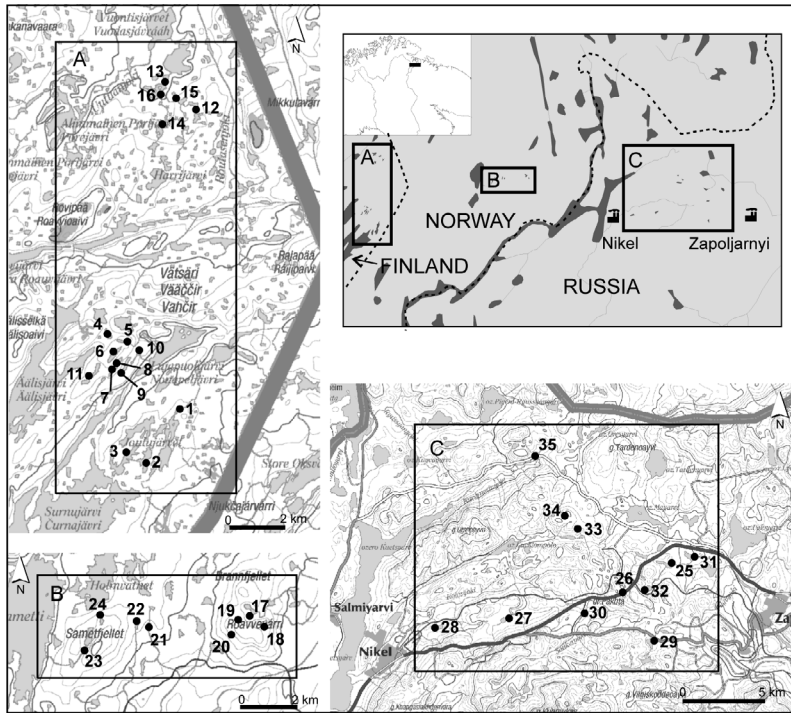
Fig. 2. Location of the study area and the sub-areas A, B and C.

(4–13 ha) lakes were sampled in 2005. All sites in sub-area A were located 93–240 m above the sea level (a.s.l.), and the sites had been sampled earlier in 1993 and 2000 using the same methods (*see* Tammi *et al.* 2003). In sub-area B (Norway), eight small (2–14 ha) isolated lakes 120–173 m a.s.l. were sampled in 2004, while in sub-area C (Russia), nine small (2–5 ha) lakes and two small brooks were sampled in 2005. There the distance from the nearest smelters varied from 1 to 16 km and the sites were located at 120–320 m a.s.l..

Electrofishing was carried out in mid-September using Hans Grassl IG200-2 portable equipment. The brooks or stony shorelines of the lakes were fished once and without any closing nets. The actual fishing sites were selected from among the most suitable-looking shores in terms of depth and substrate. At each site, the surface area fished (8–300 m²) was measured to obtain semi-quantitative estimates of littoral fish densities. The length of each fish was measured. Water samples were collected into acid-washed bottles at the same time as electrofishing was carried out, the only exception being the sulphate samples in sub-area B, which were taken in April 2005. The samples from sub-areas A and

B were analysed at the laboratory of the Lapland Regional Environment Centre according to SFS standards (Gran alkalinity, SO_4 by IC and metals by ICP-MS). Water samples from sub-area C were analysed at the laboratory of the Institute of Northern Ecology Problems (INEP), according to the same standards (and metals by GFAAS). Differences in mean water quality between sampling years in sub-area A were tested with a paired *t*-test. Differences in mean water quality between the three sub-areas were tested with ANOVA and Tukey's tests. Heavy metal concentrations were not analysed in the earlier samples taken in 1993 and 2000 from sub-area A.
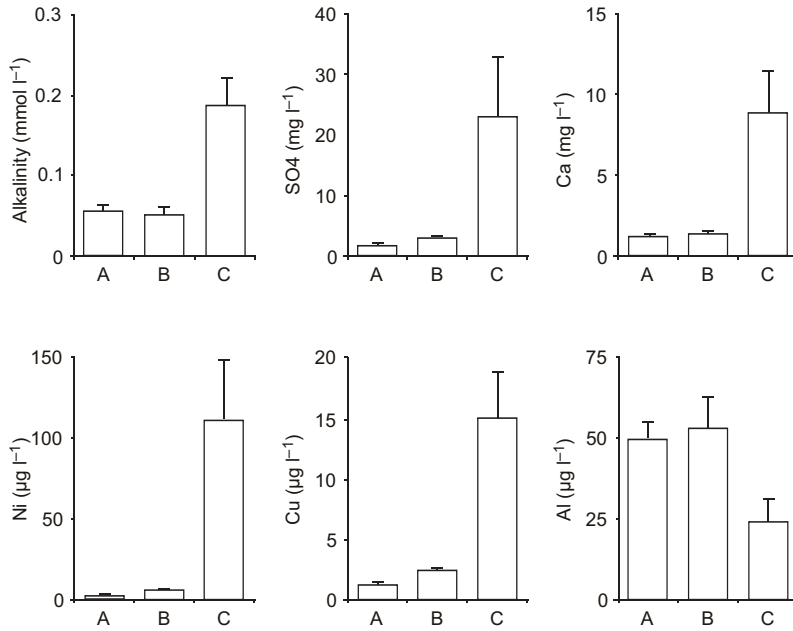
Results

Water quality

Mean alkalinity values as well as mean sulphate concentrations were significantly higher ($p < 0.005$) in sub-area C, close to the industrial area, than in sub-areas A and B (Table 1 and Fig. 3). A similar pattern was recorded for Ca concentrations, although the differences in mean values

were only significant ($p < 0.005$) between sub-areas A and C. Mean concentrations of Ni and Cu were also highest in sub-area C and lowest in sub-area A, the differences being significant ($p < 0.05$) between sub-area C and the other two, but not between sub-areas A and B. For total aluminium the reverse pattern was observed, as mean concentrations were significantly lower in sub-area C, close to the smelters, than in sub-areas A and B ($p < 0.005$). The variation, particularly in Ni, Cu and Ca concentrations, was high in sub-area C, and the highest values were measured close to the smelters (sites 27 and 28, *see* Table 1). In sub-area C, low alkalinities (close to or below 0.05 mmol l^{-1}) were only detected at sites 33 and 34, which were located more than 14 km from the smelters. During the sampling periods, surface water temperatures varied from 4.7 to 7.2 °C, from 6.0 to 7.8 °C and from 5.0 to 6.5 °C in the sub-areas A, B and C, respectively.

Table 1. Altitude, surface area (for lakes only), some chemical parameters and observed fish species at the study sites in September 2004 or 2005. M = minnow, T = Brown trout, B = burbot, Pi = pike, Pe = perch, Te = 10-spined stickleback, Th = 3-spined stickleback. Fish species indicated with boldface had an observed density of more than 10 fish 100 m^{-2}.

Sub-area and site no.	Altitude (m)	Area (ha)	Alkalinity (mmol l^{-1})	Sulphate (mg l^{-1})	Ni (µg l^{-1})	Cu (µg l^{-1})	Ca (mg l^{-1})	Al (µg l^{-1})	Fish species
Finland (A)									
1	160	–	0.063	1.6	1.2	1.1	1.2	32	–
2	161	–	0.069	1.6	1.4	1.3	1.3	40	Te
3	161	–	0.058	1.5	1.3	1.1	1.3	35	M
4	220	2	0.088	1.6	1.1	1.1	1.5	58	–
5	240	2	0.048	2.0	2.1	1.9	0.8	71	–
6	235	5	0.018	1.7	1.9	1.0	0.5	24	–
7	235	2	0.021	2.2	2.0	1.2	0.7	75	–
8	220	2	0.013	2.0	2.2	1.7	0.6	110	**M**
9	219	6	0.044	2.2	–	–	1.1	37	M
10	210	3	0.038	1.8	1.2	1.2	1.0	36	**M**
11	235	5	0.026	1.8	1.3	1.3	0.7	42	**M**
12	129	–	0.094	1.3	1.6	1.1	1.8	47	Pe
13	93	–	0.077	1.7	1.1	1.0	1.7	44	**M**
14	136	–	0.063	1.5	1.2	1.0	1.5	49	**M**
15	110	–	0.089	1.5	1.3	1.5	1.8	47	B
16	95	–	0.077	1.7	1.0	1.1	1.5	42	M,Pi
Norway (B)									
17	145	9	0.072	2.9	4.8	2.5	1.8	38	T
18	166	3	0.024	2.4	8.3	2.7	1.1	39	–
19	170	2	0.013	3.3	7.3	2.4	1.0	69	–
20	173	2	0.006	3.4	10.6	3.6	0.7	57	–
21	114	3	0.071	2.9	2.9	1.7	1.6	21	B,**Pe**
22	120	4	0.085	3.8	3.6	2.4	1.8	40	**M**,B,**Pe**
23	152	14	0.083	2.8	2.0	1.7	1.9	51	**M**,B,**Th**
24	159	2	0.057	2.4	4.9	2.4	1.6	110	–
Russia (C)									
25	210	4	0.337	14.2	31.6	6.6	9.3	7	**B**
26	187	2	0.104	10.9	89.0	10.8	5.0	5	–
27	280	3	0.267	115.5	260.0	21.4	32.1	81	–
28	120	4	0.331	39.6	360.0	43.8	16.1	33	–
29	318	5	0.142	11.7	28.6	4.4	4.6	25	B
30	255	–	0.136	9.3	23.7	4.5	4.2	11	T
31	215	4	0.197	9.6	64.0	13.1	4.6	12	Pi
32	287	3	0.316	18.6	–	–	11.5	–	–
33	230	2	0.057	7.7	74.0	18.6	3.2	11	–
34	290	2	0.021	8.7	154.0	20.1	2.5	32	–
35	210	–	0.159	7.2	20.5	7.4	3.6	21	**T**

Fig. 3. Average (+ SE) of selected water quality parameters at the study sites in sub-areas A, B and C in 2004 or 2005.

Occurrence of littoral fish species

Electrofishing surveys in 2004 and 2005 yielded a total of seven fish species. The minnow was generally the most abundant species, with burbot being the second most frequently observed species. In the Finnish sub-area A, minnows were found at eight out of sixteen sites. Burbot, ten-spined stickleback (*Pungitius pungitius*), perch (*Perca fluviatilis*) and pike (*Esox lucius*) were each found at one site. In the Norwegian sub-area B, burbot were recorded in three lakes out of eight, minnow and perch were detected in two lakes, and both brown trout and three-spined stickleback (*Gasterosteus aculeatus*) in one lake. In the Russian sub area C, burbot were found in two lakes out of eleven (Table 1). Brown trout were recorded in both Russian brooks surveys (sites 30 and 35), and according to the length distributions, one individual of age group 0+ (5–6 cm) and three individuals of age group 1+ (8–10 cm) were caught at site 30, and five individuals of age group 0+ and three individuals of age group 1+ at site 35. A single pike was captured in one lake.

The proportion of sites where no fish were captured was highest (55%) in sub-area C, close to the smelters. At five out of these six sites without fish, measured Ni and Cu concentrations

were exceptionally high (Ni > 70 μg l^{-1}, Cu > 18 μg l^{-1}). Unfortunately, the heavy metal analysis for site 32 failed in the laboratory, but based on the location of this lake, from which no fish were captured, it can be assumed to have had high heavy metal concentrations. In sub-area B, no fish were recorded at half of the eight sites, and lakes without fish were typically among the smallest, located at a high altitude and having the lowest alkalinity (mostly < 0.05 mmol l^{-1}) (Table 1). In sub-area A, no fish were detected at 5 out of 16 sites (31%). Three of these sites were in small lakes with low alkalinity (< 0.05 mmol l^{-1}), and one was in a small lake where alkalinity was a little higher (0.088 mmol l^{-1}). In addition, no fish were observed in one brook (site 1), but several species and individuals had been found there during surveys conducted in 1993 and 2000. In 2005, sites 1–3 were visited during a day when the weather was harsh and there was temporarily some wet snow, which may have affected the results from these brooks. When results for the sub-areas were pooled, no fish were generally observed when Ni concentrations exceeded 64 μg l^{-1} (Fig. 4). The observation of fish at a Ni concentration of 64 μg l^{-1} was based on one pike caught at site 31. Fish were also rarely observed at sites with alkalinity lower than 0.05 mmol l^{-1}.

Fig 4. Occurrence of littoral fish in relation to surface water alkalinity and Ni concentrations at the study sites. The vertical dotted line shows the 'critical' alkalinity level (0.05 mmol l^{-1}) and the horizontal dotted line represents a Ni concentration of 70 µg l^{-1}, above which no fish were found.
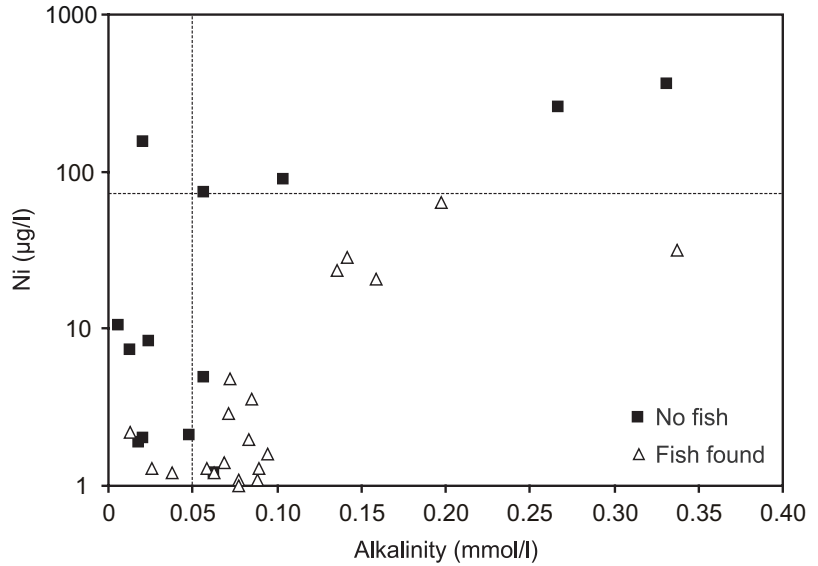
Water quality and fish observations in sub-area A during 1993, 2000 and 2005

At all sampling sites in the Finnish sub-area A, alkalinity was significantly higher and sulphate concentrations lower in 2000 than in 1993 ($p < 0.001$, Table 2). In 2005, alkalinity was even higher and sulphate concentrations lower than in 2000 ($p < 0.001$). The total aluminium concentrations showed no significant differences between 1993 and 2000, but were significantly higher in 2005 that in 2000 ($p < 0.05$). In 2005,

Table 2. Some chemical properties of surface water samples taken in mid-September 1993, 2000 and 2005 from the Finnish study sites, 40–50 km from the smelters. Data from 1993 and 2000 were obtained by Tammi *et al.* (2003).

Site	Alkalinity (mmol l^{-1})			Sulphate (mg l^{-1})			Aluminium (µg l^{-1})		
	1993	2000	2005	1993	2000	2005	1993	2000	2005
Joulujärvi area (brooks)									
1	0.031	0.066	0.063	2.7	2.1	1.6	24	31	32
2	0.040	0.051	0.069	2.7	2.0	1.6	32	29	40
3	0.024	0.041	0.058	2.7	1.9	1.5	33	26	35
Äälisjärvi area (lakes)									
4	0.047	0.076	0.088	2.3	1.8	1.6	49	46	58
5	−0.004	0.015	0.048	2.7	2.0	2.0	20	29	71
6	−0.004	0.013	0.018	2.7	1.9	1.7	15	15	24
7	0.002	0.020	0.021	3.0	2.4	2.2	31	33	75
8	−0.002	0.016	0.013	2.7	2.2	2.0	39	28	110
9	0.020	0.029	0.044	2.7	2.3	2.2	26	26	37
10	0.019	0.026	0.038	2.8	2.2	1.8	22	24	36
11	0.012	0.022	0.026	2.6	2.0	1.8	13	19	42
Vuontisjärvi area (brooks)									
12	0.058	0.075	0.094	2.2	1.7	1.3	35	62	47
13	0.061	0.067	0.077	2.5	1.9	1.7	24	36	44
14	0.047	0.051	0.063	2.6	1.9	1.5	26	51	49
15	0.073	0.083	0.089	2.4	1.8	1.5	33	72	47
16	−	0.074	0.077	−	2.2	1.7	−	30	42

alkalinity was still low (< 0.05 mmol l^{-1}) in small upland lakes (sites 5–11), except at site 4 which is connected to an adjacent larger lake. Surface water temperatures at the sites during the sampling period varied from 6.3 to 9.0 °C, from 7.5 to 9.4 °C and from 4.7 to 7.2 °C in 1993, 2000 and 2005, respectively.

The minnow was generally the most abundant species caught in sub-area A during all three sampling periods (Table 3), the highest observed densities in suitable habitats being roughly 50–100 individuals 100 m^{-2}. In most of the small upland lakes (sites 4–11) and in the outlets and brooks between small lakes (sites 12–16) minnow were consistently observed at almost the same sites during the three sampling periods (Table 3). However, in the brooks connecting larger lakes (sites 1–3), minnow observations were less consistent. Juvenile brown trout and burbot were the second most abundant species observed in sub-area A. These species, particularly brown trout, were consistently observed at same sites in 1993 and 2000, but juvenile brown trout and burbot were absent from most sites in 2005.

Discussion

Surface water acidification

Surface water acidification due to high sulphur deposition and high heavy metal deposition are the most typically reported anthropogenic threats to aquatic biota in the Kola region (e.g. Nost *et al.* 1991, Moiseenko *et al.* 1995, Hesthagen *et al.* 1998, Kashulin *et al.* 2001), and in certain circumstances these two threats may be additive, as a low a pH facilitates the uptake of heavy metals by fish (Moiseenko and Kudryavtseva 2001). The effects of high sulphur emissions from the Pechenganickel smelters were strictly seen in our study area as particularly high sulphate concentrations in the Russian industrial area (sub-area C), and as lower sulphate levels towards the west up to the Norwegian and Finnish border areas. The highest levels measured in the industrial area (up to 115 mg l^{-1}) were similar to the highest levels (116 and 123 mg l^{-1}) measured there earlier in stream waters (Väisänen *et al.* 1998). In the Finnish border area, located 40–50 km west of the smelters and almost against the prevailing

Table 3. Densities (ind. 100 m^{-2}) of minnow, brown trout and burbot at Finnish study sites in 1993, 2000 and 2005 (– = no fish captured.). Data from 1993 and 2000 were obtained by Tammi *et al.* (2003).

Site	Minnow			Brown trout			Burbot		
	1993	2000	2005	1993	2000	2005	1993	2000	2005
Joulujärvi area (brooks)									
1	1	–	–	19	7	–	1	7	–
2	5	4	–	3	1	–	1	4	–
3	–	13	2	–	–	–	2	2	–
Äälisjärvi area (lakes)									
4	1	–	–	–	–	–	1	1	–
5	–	–	–	–	–	–	2	2	–
6	–	1	–	–	–	–	–	–	–
7	–	–	–	–	–	–	16	–	–
8	12	28	131	–	–	–	–	–	–
9	17	–	8	–	–	–	–	10	–
10	30	73	17	–	–	–	–	–	–
11	6	5	13	–	–	–	–	–	–
Vuontisjärvi area (brooks)									
12	20	–	–	–	–	–	4	–	–
13	16	13	10	9	2	–	–	–	–
14	46	79	101	–	–	–	13	4	–
15	422	755	900	–	–	–	–	–	3
16	87	73	2	–	–	–	13	–	–

southwesterly winds, the mean sulphate concentrations in 2005 were close to the median value, 1.9 mg l⁻¹ (40 μeq l⁻¹) of 184 randomly-sampled lakes in northern Finland (Mannio *et al.* 2000). However, the sulphate levels have decreased from that measured in 1993 (2.2–3.0 mg l⁻¹).

The present high sulphur levels, however, had not led to low buffering capacities (< 0.05 mmol l⁻¹) in the vicinity of the smelters, even though the waters studied were small and thus presumably among those most sensitive to acidification. Particle emissions from the Pechenganickel smelters are ineffectively controlled and basic ash phases falling nearer the smelters than the potentially acidic gaseous phases might result in a rise in pH and buffering capacity in the immediate vicinity of the smelters (Moiseenko 1995, Reinmann *et al.* 1999). Dust from local strip-mining activities might also be a source of base cations (Kashulina *et al.* 2003). In our study area, the few poorly buffered (alkalinity < 0.05 mmol l⁻¹) lakes found were located at least 10 km from the smelters, and they were all extremely small upland lakes above the local coniferous tree line.

In poorly buffered lakes and brooks within the study area, episodic pH drop may occur during the spring snowmelt, and this period is likely to be critical to fish populations. Hesthagen *et al.* (1998) reported acid-induced damage to fish populations in the Norwegian Jarfjord area, 30 km north-east of the Nikel and Zapoljarnyi smelters. There, 14 Arctic char or brown trout populations of small (3–15 ha) lakes were identified as either lost or in various stages of extinction (Hesthagen *et al.* 1998). The minnow is among the most sensitive species to acidification (Muniz 1984, Berqvist 1991). Lappalainen *et al.* (1995) and Tammi *et al.* (2003) suggested that the presence of only few large and old minnows or the total absence of this species in the most poorly buffered lakes in sub-area A are signs of acid-induced damage. Similar potential signs of acid-induced fish population damage were also found in the Norwegian border area, where no fish were detected in the most poorly buffered small lakes. Due to their invasion history, minnows as well as perch, burbot and pike are not distributed in the northernmost parts in the Norwegian border region and Jarfjord area (Hesthagen *et al.* 1998), but minnows are found, for instance, in a large (1700 ha) lake two kilometres from the Nikel smelter (Lukin *et al.* 2003). Thus, the original distribution area of minnows should also cover the small lakes in the industrial area, especially as there were ample habitats that appeared suitable for this species. It is likely that minnow populations in the small waters of the industrial area have disappeared. Minnows may be more sensitive to high heavy metal concentrations than species such as burbot and pike.

Heavy metal pollution

The high Ni and Cu concentrations (maximum 360 and 44 μg l⁻¹, respectively) measured in the vicinity of the smelters were on a same level as reported by Moiseenko and Kudryavtseva (2001), Kashulin *et al.* (2001) and Lukin *et al.* (2003), and were one or two orders of magnitude higher than the averaged background levels (0.5 μg l⁻¹ for Ni and 0.6 μg l⁻¹ for Cu) of the Kola Peninsula (Moiseenko and Kudryavtseva 2001). Even higher heavy metal concentrations have been reported in Canada, near the Coniston nickel-copper smelter, where the highest water Ni concentrations in two extremely polluted lakes before the closure of the smelter were 2000–5000 μg l⁻¹ and Cu concentrations 500–600 μg l⁻¹ (Woodfine and Havas 1995). In our study areas, the effects of Ni and Cu emissions from the Pechenganickel smelters were clear in the small waters of the Norwegian border region as well as in the Finnish border region, as the average values measured in the latter region (Ni 1.5 μg l⁻¹ and Cu 1.2 μg l⁻¹) were still higher than the medium values of small lakes in Finland (Ni and Cu 0.4 μg l⁻¹) (Skjelkvåle *et al.* 2001). Woodfine and Havas (1995) demonstrated a good capacity of heavily polluted lakes to recover from chemical stress over a short period, as the Ni and Cu concentrations in two monitored lakes had declined to less than a tenth of former levels 20 years after the closure of the Conistron smelter in Canada. The heavy metal emissions from the Pechenganickel smelters have also decreased, being at the beginning of the 2000s approximately half of the level in the late 1970s. Thus,

the heavy metal concentrations in lake water in the sphere of influence of the smelters might have been even higher during previous decades.

High heavy metal concentrations are a likely reason for the lack of fish observations in the immediate vicinity of the Pechenganickel smelters. Fish reproduction and early life stages are generally considered to be the most sensitive to surface water acidification, as reviewed by Haines (1981) and Magnuson *et al.* (1984), but reports of the effects of Ni and Cu pollution on fish have focused on the accumulation of metals in fish tissues and the prevalence of fish diseases and abnormalities in adult individuals (Moiseenko *et al.* 1995, Amundsen *et al.* 1997, Moiseenko and Kudryatseva 2001, Lukin *et al.* 2003). The serious pathological symptoms observed in the kidney and liver of around 90% of the fish in lakes within a radius of 30 km of the smelters (Moiseenko and Kudryavtseva 2001) can presumably also have serious effects on the population level. Moiseenko *et al.* (1995) presented critical levels for Ni and Cu concentrations in lake waters based on the occurrence of fish diseases. The critical levels in well-buffered waters, such as our lakes in the Russian sub-area A, were 20 μg l^{-1} for Ni and 8 μg l^{-1} for Cu. These figures seem to fit our data fairly well, as only a few burbot and pike were found in lakes with slightly higher concentrations, the highest values with fish being 64 μg l^{-1} Ni and 13 μg l^{-1} Cu. Brown trout in the Russian area were observed in brooks where Ni and Cu concentrations were very close to the presented critical levels. It is evident that all the lakes showing Ni and Cu concentrations over the critical levels are located less than a 10 km radius from the smelters, where the highest Ni concentrations in rainwater are also observed (Karaban and Gytarsky 1995), and the total number of lakes with especially high heavy metal concentrations is likely to be a few dozen.

In addition to heavy metals, aluminium occurring as inorganic ions (labile aluminium) has been shown to be harmful for fish. Concentrations of inorganic aluminium exceeding 60 μg l^{-1} can produce toxic conditions in sensitive fish species at a low pH (Skogheim and Rosseland 1986). In our samples, the concentrations of labile aluminium were not analysed. However, even total aluminium concentrations, including all fractions of aluminium, were in most cases less that 60 μg l^{-1}. Total aluminium concentrations of at least 150 μg l^{-1} at pH between 5 and 6 have been reported to cause high mortality in minnows (Norrgren *et al.* 1991). However, our highest measured values were 110 μg l^{-1}, indicating that aluminium is not a major risk for fish in our study area.

Remarks on the survey methods

Results of earlier studies in this region (e.g. Hesthagen *et al.* 1998, Tammi *et al.* 2003) suggest that the extent of damage to fish populations caused by acidification is relatively restricted. Hence, this study focused on extremely small waters, which are usually the most sensitive to the effects of airborne emissions, particularly to acidification. These waters and fish populations were used as initial or early warning indicators concerning effects of acidification. Electrofishing is a feasible survey method in these small waters as they are typically inhabited by small fish species such as minnow, burbot or juvenile brown trout, which prefer shallow stony habitats and are difficult or impossible to catch with the standard gill-nets used in larger lakes (e.g. Moiseenko *et al.* 1995, Amundsen *et al.* 1997, Hesthagen *et al.* 1998, Lukin *et al.* 2003).

Our results from littoral fish surveys in the Finnish border region during mid-September in three separate years showed that the minnow, inside its distribution area, is an excellent indicator species in small lakes and brooks between small lakes for two reasons: (i) it is regularly and abundantly found at the same sites, and (ii) it is known to be sensitive at least to acidification (Muniz 1984, Bergquist 1991). However, burbot and juvenile brown trout were caught at the same sites in 1993 and 2000, but not in 2005. The almost total lack of these species in 2005 might be a coincidence, as their densities have been low. Our sampling was carried out during or shortly after the autumn overturn, which is generally considered a suitable time for water sampling (Henriksen *et al.* 1998). Electrofishing surveys are usually also carried out during late summer and early autumn.

The sulphur and heavy metal deposition from

the Pechenganickel smelters has been decreasing and the recovery of surface waters is expected and already taking place in this Arctic region (AMAP Assessment, 2006). Basic information on the recovery of the small waters can be acquired by solely monitoring the water quality. A disadvantage in using fish populations as indicators of the recovery of very small and isolated waters is that once totally lost, fish cannot recolonize without the assistance of reintroductions. Colonization is, however, possible in small river systems where, for example, brown trout can return to ancient spawning and nursery areas from lower parts of a river in the absence of migration obstacles. Recovery from less severe fish population damage caused, for instance, by partial or occasional reproduction failures can also be detected in small waters by electrofishing (Tammi *et al.* 2003).

Conclusions

The two Pechenganickel smelters have been among the world's largest point sources of SO_2 emissions, and the emissions of heavy metals have also been high. However, airborne emissions of the smelters have not caused any widespread damage to fish populations, even in the most sensitive small waters in the study area, a border region of Finland, Norway and Russia. The small waters close to the smelters (at a radius of approximately 10 km) are well buffered against the effects of high sulphate deposition. Extremely high concentrations of the heavy metals Ni and Cu, however, are a local threat to biota in small waters close to the smelters, where a few lakes that have presumably lost all fish populations can be found. In the border area of Russia, Norway and Finland, acidification is currently only a problem in certain small and sensitive waters in local highland areas, 15–50 km from the smelters, where some fish populations, mainly minnows, have probably been lost. SO_2 emissions from the smelters have decreased to approximately one third of the maximum level in the late 1970s. This can be seen in the recovery of the buffering capacity of small lakes such as those in the Finnish border region, 40–50 km west of the smelters.

References

AMAP Assessment 2006. *Acidifying pollutants, Arctic haze, and acidification in the Arctic*. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

Amundsen P-A., Staldvik F.J., Lukin A.A., Kashulin N.A., Popova O.A. & Reshetnikov Y.S. 1997. Heavy metal contamination in freshwater fish from the border region between Norway and Russia. *Sci. Total Environ.* 201: 211–224.

Bergquist B.C. 1991. Extinction and natural recolonization of fish in acidified and limed lakes. *Nord. Freshwater Res.* 66: 50–62.

Gunn J.M., Keller W., Negusanti J., Potvin R., Beckett P. & Winterhalder K. 1995. Ecosystem recovery after emission reductions: Sudbury, Canada. *Water Air Soil Pollut.* 85: 1783–1788.

Haines T.A. 1981. Acidic precipitation and its consequences for aquatic ecosystems; a review. *Trans. Am. Fish. Soc.* 110: 669–707.

Hesthagen T., Langeland A. & Berger H.M. 1998. Effects of acidification due to emissions from the Kola Peninsula on fish populations in lakes near the Russian border in northern Norway. *Water Air Soil Pollut.* 102: 17–36.

Henriksen A., Skjelkvåle B.L., Mannio J., Wilander A., Harriman R., Jensen J.P., Fjeld E. & Moiseenko T. 1998. Northern European lake survey 1995. *Ambio* 27: 80–91.

Karaban R.T. & Gytarsky M.L. 1995. Studies of precipitation contamination levels over the north-western forests of Russia subject to emissions from the two nickel smelters. *Water Air Soil Pollut.* 85: 2071–2076.

Kashulin N.A., Ratkin N.E., Dauvalter V.A. & Lukin A.A. 2001. Impact of airborne pollution on the drainage area of subarctic lakes and fish. *Chemosphere* 42: 51–59.

Kashulina G., Remann C. & Banks D. 2003. Sulphur in the Arctic environment (3): environmental impact. *Environ. Pollut.* 124: 151–171.

Keller W., Heneberry J.H. & Gunn J.M. 1999. Effects of emission reductions from the Sudbury smelters on the acid- and metal-damaged lakes. *J. Aquat. Ecosyst. Stress Recovery* 6: 189–198.

Lappalainen A., Mähönen O., Erkinaro J., Rask M. & Niemelä E. 1995. Acid deposition from the Russian Kola Peninsula: are sensitive fish populations in north-eastern Finnish Lapland affected? *Water Air Soil Pollut.* 85:

439–444.

Lukin A., Dauvalter V., Kashulin N., Yakovlev V., Sharov A. & Vandysh O. 2003. Assessment of copper-nickel industry impact on a subarctic lake ecosystem. *Sci. Total Environ.* 306: 73–83.

Magnuson J.K.J., Baker J.P. & Rahel F.J. 1984. A critical assessment of effects of acidification on fisheries in North America. *Phil. Trans. R. Soc. Lond. B* 305: 501–516.

Mance G. 1987. *Pollution threat of heavy metals in aquatic environments.* Elsevier, London.

Mannio J., Räike A. & Vuorenmaa J. 2000. Finnish Lake Survey 1995: Regional characteristics of lake chemistry. *Verh. Internat. Verein. Limnol.* 27: 362–367.

Moiseenko T. 1995. Critical loads of SO_4 for surface waters in the Kola region of Russia. *Water Air Soil Pollut.* 85: 469–473.

Moiseenko T.I., Kudryavtseva L.P., Rodyushkin I.V. Dauvalter V.A., Lukin A.A. & Kashulin N.A. 1995. Airborne contamination by heavy metals and aluminium in the freshwater ecosystems of the Kola Subarctic region (Russia). *Sci. Total Environ.* 160/161: 715–727.

Moiseenko T.I. & Kudryavtseva L.P. 2001. Trace metal accumulation and fish pathologies in areas affected by mining and metallurgical enterprises in the Kola Region, Russia. *Environ. Pollut.* 114: 285–297.

Muniz I.P. 1984. The effects of acidification on Scandinavian freshwater fish fauna. *Phil. Trans. R. Soc. Lond. B* 305: 517–528.

Norrgren L., Wicklund-Glynn A. & Malmborg O. 1991. Accumulation and effects of aluminium in the minnow (*Phoxinus phoxinus* L.) at different pH levels. *J. Fish Biol.* 39: 833–847.

Nost T., Yakovlev V., Berger H.M., Kashulin N., Langeland A., Lukin A. & Muladal H. 1991. *Impacts of pollution on freshwater communities in the border region between Russia and Norway. I. Preliminary study in 1990.* Norwegian Institute for Nature Research, Report 26.

Paatero J., Makkonen U., Kauhaniemi M., Salmi T., Stebel K. & Grekelä I. 2006. Effects of the Pechenganikel smelter to air quality in the Finnish–Norwegian–Russian border area. In: *Modern ecological problems of the North.* Proceedings of the International Conference 10–12 October 2006, Apatity. Part 2. Russian Academy of Sciences, Kola Science Centre, Apatity, Russia, pp. 223–227.

Reimann C., Banks D., Bogatyrev I., de Caritat P., Kashulina G. & Niskavaara H. 1999. Lake water geochemistry on the western Kola Peninsula, north-west Russia. *Appl. Geochem.* 14: 787–805.

Reimann C., Banks D. & de Caritat P. 2000. Impacts of airborne contamination on a Regional soil and water Quality: The Kola Peninsula, Russia. *Environ. Sci. Technol.* 34: 2727–2732.

Skjelkvåle B.L., Andersen T., Fjeld E., Mannio J., Wilander A., Johansson K. Jensen J.P. & Moiseenko T. 2001. Heavy metal surveys in Nordic lakes; concentrations, geographic patterns and relation to critical limits. *Ambio* 30: 2–10.

Skogheim O.K. & Rosseland B.O. 1986. Mortality of smolt of Atlantic salmon, *Salmo salar* L., at low levels of aluminium in acidic softwater. *Bull. Environ. Contam. Toxicol.* 37: 258–265.

Tammi J., Lappalainen A. & Bergman T. 2003. Water quality and fish populations of acid sensitive waters in the Vätsäri area, north-eastern Finland: responses to reduced sulphur emissions from the Kola Peninsula, Russia, in the 1990s. *Boreal Env. Res.* 8: 1–7.

Tommervik H., Johansen B.E. & Pedersen J.P. 1995. Monitoring the effects of air pollution on terrestrial ecosystems in Varanger (Norway) and Nikel-Pechenga (Russia) using remote sensing. *Sci. Total Environ.* 160/161: 753–767.

Väisänen U., Misund A. & Chekushin V. 1998. Ecogeochemical investigations: stream water quality as an indicator of pollution in the border areas of Finland, Norway and Russia. *Water Air Soil Pollut.* 104: 205–219.

Woodfine D.G. & Havas M. 1995. Pathways of chemical recovery in acidified, metal-contaminated lakes near Sudbury, Ontario, Canada. *Water Air Soil Pollut.* 85: 797–803.