BOREAL ENVIRONMENT RESEARCH 8: 61–70 ISSN 1239-6095
Helsinki 25 March 2003

Metal contamination of streams in relation to catchment silvicultural practices: a comparative study in Finnish and Russian headwaters

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Vuori, K.-M., Siren, O. & Luotonen, H. 2003: Metal contamination of streams in relation to catchment silvicultural practices: a comparative study in Finnish and Russian headwaters. *Boreal Env. Res.* 8: 61–70. ISSN 1239-6095

We studied metal contamination of streams in the forestry dominated and close-on pristine subcatchments in Finland and Russian Karelia. In this area, atmospheric deposition and point loading are insignificant as sources of metals, while geochemical sources and silvicultural activities are important. Aquatic moss *Fontinalis antipyretica* Hedw. was used as a sentinel organism measuring the metal contamination. Tufts of *Fontinalis* were collected at 10 Russian and 8 Finnish stream sites for the measurement of whole-plant Al, Fe, Cd, Zn, Cu, Pb and Ni concentrations. The average Al, Fe, Cd, Cu, Zn and Ni concentrations were statistically significantly higher in the Finnish moss samples than in the Russian ones. In particular, concentrations of Al, Fe, Cu, Ni and Zn in many Finnish streams clearly exceeded natural background concentrations and were comparable to the earlier results from streams affected by metal loading from point and non-point sources. The results suggest that silvicultural practices in combination with specific geochemical features may cause metal contamination of streams in areas considered otherwise unpolluted. We stress the importance of taking precautions against forestry-induced metal contamination

Introduction

Intensive silvicultural practices have caused impairment of water quality and ecological integrity of many headwater streams and lakes (Heikurainen *et al*. 1978, Seuna 1982, Ormerod *et al*. 1987, Ahtiainen 1988, Binkley and Brown 1994). In Finland, forestry practices during the past four decades have included extensive clear fellings, large-scale drainage of wetlands, fertilization and the construction of a comprehensive network of forest roads. These practices have caused long-term deterioration of headwater streams and lakes (Turkia *et al*. 1998, Ahtiainen and Huttunen 1999). Despite the recent development of environmental guidelines of forest management, including many water protection measures, forestry-induced adverse effects to streams are still obvious (Vuori and Joensuu 1996, Joensuu *et al*. 1999).

Impact assessment of silvicultural practices has largely focused on the load of nutrients and suspended solids to watercourses. Forestry-induced load of metals has received much less attention, although the number of studies report that forestry may significantly increase leaching and concentration of such metals as aluminium and iron in streams (Ahtiainen 1988, Reynolds *et al*. 1992, Miller *et al*. 1996, Manninen 1998, Vuori *et al*. 1998). Recent studies have demonstrated that the increased load of metals from silvicultural activities may be harmful for the well-being of aquatic communities (Vuori *et al*. 1998, Vuorinen *et al*. 1999).

The aim of the present study is to clarify the potential role of forestry in metal contamination of headwater streams. We use aquatic moss species *Fontinalis antipyretica* as a sentinel organism measuring metal contamination. This moss forms dense tufts in stream bottoms and serves as an important source of food and shelter for benthic organisms. The moss vegetation also has a predominant role in the total metabolism and overall functioning of many boreal stream ecosystems (Naiman 1983, Triska *et al*. 1982, Vuori and Muotka 1999). Aquatic bryophytes have been commonly used in assessing metal pollution of streams. Bryophytes respond quickly to increases in ambient metal concentrations and retain the increased levels for several days, even weeks, after concentrations in water have decreased. This enables the monitoring of both chronic metal contamination and sudden discharges (Say and Whitton 1983, Wehr and Whitton 1983, Mouvet *et al*. 1993). Our study species, *Fontinalis antipyretica*, has been considered as an ideal sentinel organism for metal studies in streams because it is widely distributed and relatively tolerant to pollution (Say and Whitton 1983, Vanderpoorten 1999).

Often, a major problem in assessing the impact of watershed land use on aquatic ecosystems is the lack of reliable reference conditions. Impact assessment of forestry-induced metal contamination should be based on comparisons between streams affected by silviculture relative to those not subjected to such impact. Ideally, such comparative studies should be carried out within the same catchment in order to overcome problems of large scale variation in climatic, historical and geographical factors (Clenaghan *et al*. 1998). This is possible in the border zone of Finland and Russia, due to the historical differences in the land-use between the two countries (Vuori *et al*. 1999). Our study is based on comparative sampling of streams located within the same catchment but on different sides of the Finnish-Russian border.

Study area

The study was conducted in the Finnish and Russian parts of the catchment of the Ylä-Koitajoki (Fig. 1). Forestry is the dominant form of land-use in the catchment, but its intensity differs drastically between the two countries. On the Finnish side most of the wetlands and forests have been extensively drained and logged. On the other hand, on the Russian side only minor local loggings have taken place after World War II (1939–1945), and the wetlands are in a natural state. The total land area of the catchment is 2262 km^2 (522 km^2 in Finland) and the percentage of lakes in the area is 5.9%. The bedrock of the drainage area is formed mainly of gneiss–granite. The soil consist mostly of peat and moraine. Over 50 per cent of the land area of the basin is covered by peat lands, the rest of the area is mainly forest.

Moss and water samples were collected at 10 sites on the Russian side and 8 sites on the Finnish side of the catchment area (Fig. 1). The size of the streams ranged from 1st to 5th order streams. Significant correlations between stream order and moss metal concentration were not observed. In Finland, all of the sampling sites are located in the immediate vicinity of drained peat lands and logged forests. In Russia, only one station may be suspected to be subject to runoff from such activities. This is the station in the main channel of the Koitajoki (R7, Fig. 1). However, while this station is located some 15 km downstream from the Finnish border and it

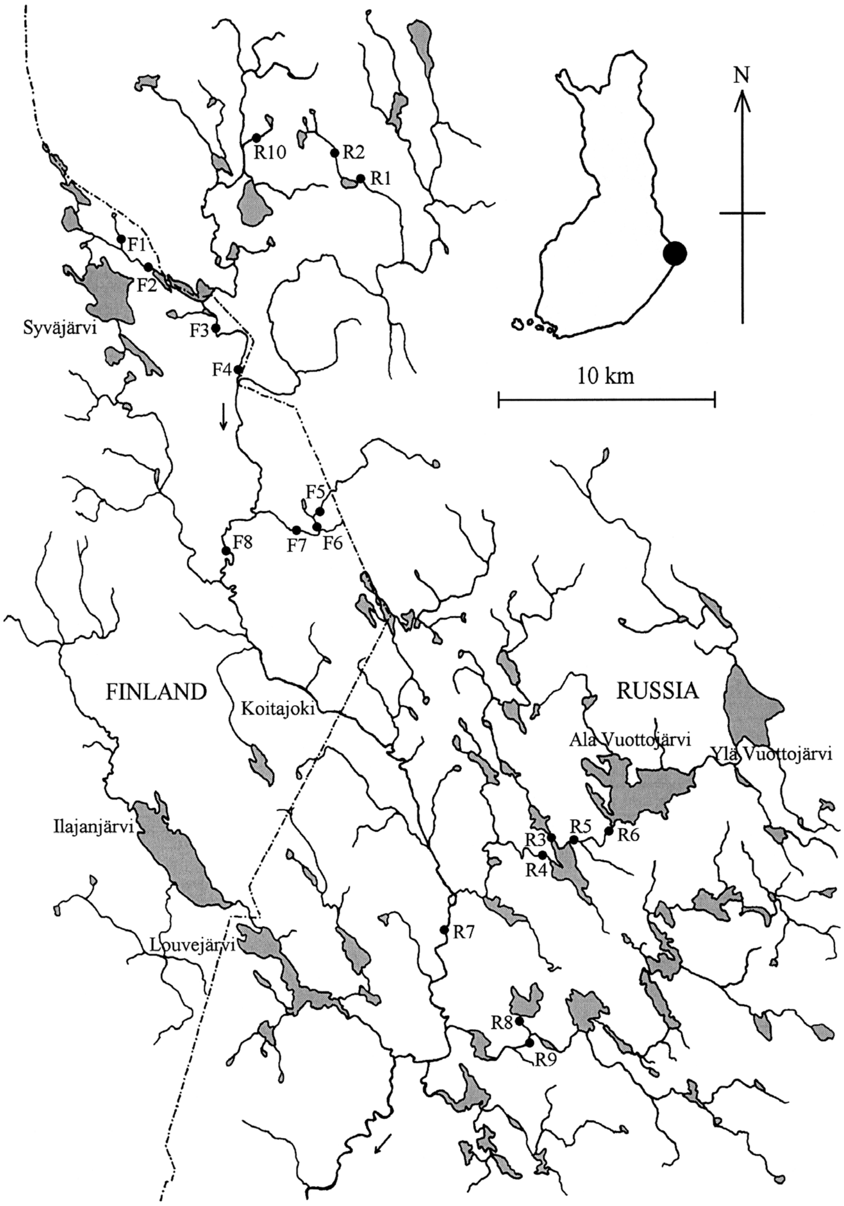

Fig. 1. Study area and sampling sites.

receives major runoff from the Russian tributaries, the impact from forestry might be considered minor relative to the Finnish stream sites.

Material and methods

Tufts of *Fontinalis antipyretica* Hedw. growing on stone surfaces in swift currents (50–60 cm s^{-1}) were sampled by hand in autumn 1996. The moss tufts were rinsed several times in stream water to remove excess organic and inorganic material and placed into acid-washed polyethylene containers. A volume of approximately 2 litres of moss tufts from three replicate samples were collected at each station. In the laboratory the mosses were deep frozen until the metal analysis stage.

Water samples to carry out analysis of chemical oxygen demand and total Al, Fe, Cd, Cu, Ni, Zn, Pb, Mn, Ca, Mg, nitrogen and phosphorus concentrations were taken once at each sampling site. Samples for metal analysis were taken in 125 ml acid-washed polyethylene bottles (Nalgene) and acidified with 0.5 ml conc. suprapurified HNO_3 in 100 ml samples. Transportation difficulties and long storage periods restricted our ability to carry out more extensive water quality sampling. The frozen moss samples were melted within six months of sampling. Five replicate samples of randomly chosen whole moss shoots from each sampling site were rinsed carefully in deionised water, dried in +105 °C for 6 hours, cooled in a desiccator and weighed with a Mettler AE200 analytical balance to the nearest 0.1 mg. The weighed moss shoots were transferred to 25 ml acid-washed quartz tubes and concentrated HNO_3 was added so that the sample-acid ratio was 1:10 (w/V). The tubes were heated at 50 °C for 2 h and then at 110 °C for 6 h. The sample was then made up to 20 ml with distilled water (Finnish Standards Association, SFS 5075). The metal concentrations of Al, Fe, Cd, Zn, Cu, Pb and Ni in the water and biotic samples were determined by atomic absorption spectrophotometer (Perkin Elmer 5000 Zeeman) according to the Finnish standards (Finnish Standards Association, SFS 3044, 3047 and 5502). An air-acetylene flame was used for Zn and an HGA 500 graphite furnace for Al, Fe, Cu, Cd and Pb analysis. Analytical accuracy was verified through the use of certified reference material (aquatic moss *Platihypnidium riparioides,* BCR No. 61).

Results

Water quality

There were clear differences in the water quality of the Russian and Finnish sampling sites (Table 1). Low concentrations of Al (46–120 μg l^{-1}), Fe (480–1000 μg l^{-1}) and organic matter (reflected by COD, 7.5–19 mg O_2 l^{-1}) were typical for almost all Russian streams. The only exceptions were Station R10 with somewhat elevated levels of COD and Al, and Station R7 with an elevated iron level, 1500 μg Fe l^{-1}. Station R7 locates in the main channel of the Koitajoki and is affected by the Finnish catchments (Fig. 1). Elevated Al (130–190 μg l^{-1}) and Fe (1200–1300 μg l^{-1}) concentrations characterized Finnish streams affected by forestry. Concentrations of all other metals were below their detection limits. Relatively high COD levels (23–50 mg O_2 l^{-1}) in all Finnish streams indicated an increased load of organic matter, probably due to heavy drainages of the catchment peat lands. Concentrations of Cd, Cu, Pb, Zn and Ni were below the detection limit in both Russian and Finnish stream waters, with the only exception at site F6 with a somewhat elevated Zn concentration (6 μg l^{-1}). Detection limits were 0.1, 1.0, 1, 2 and 1 μg l^{-1} for Cd, Cu, Pb, Zn and Ni, respectively.

Metals in mosses

There was a general tendency, excluding lead, for significantly higher whole-plant metal concentrations in the Finnish moss samples when compared to the Russian ones (Fig. 2).

Concentrations of aluminium in mosses ranged

Table 1. Water quality of the Russian and Finnish sampling sites. Site numbers refer to Fig. 1.

Russian sites	**R1**	**R2**	**R3**	**R4**	**R5**	**R6**	**R7**	**R8**	**R9**	**R10**
COD, Mn mg O_2 l^{-1}	14	18	19	13	19	15	13	7.5	15	25
Tot. N μg l^{-1}	–	–	–	–	–	–	–	–	–	–
Tot. P μg l^{-1}	12	36	14	23	29	23	20	11	20	14
Fe μg l^{-1}	720	860	610	980	1000	970	1500	620	480	660
Al μg l^{-1}	46	55	85	86	120	88	84	66	78	200
Mn μg l^{-1}	18	15	15	30	34	35	21	20	20	48
Ca mg l^{-1}	2.5	2.1	1.1	1.2	1.1	1.2	2.0	1.0	1.2	1.3
Mg mg l^{-1}	0.9	0.9	0.5	0.4	0.4	0.4	0.7	0.4	0.4	0.4
Finnish sites	**F1**	**F2**	**F3**	**F4**	**F5**	**F6**	**F7**	**F8**		
COD, Mn mg O_2 l^{-1}	23	23	28	28	32	45	50	48		
Tot. N μg l^{-1}	440	440	460	450	420	540	530	530		
Tot. P μg l^{-1}	26	26	19	19	15	13	14	14		
Fe μg l^{-1}	1200	1300	1200	1200	1300	1400	1300	1300		
Al μg l^{-1}	130	140	140	140	140	190	180	190		
Mn μg l^{-1}	41	34	39	33	53	59	56	59		
Ca mg l^{-1}	1.5	1.5	1.7	1.8	1.8	1.8	1.8	1.8		
Mg mg l^{-1}	0.5	0.5	0.6	0.6	0.6	0.6	0.6	0.6		

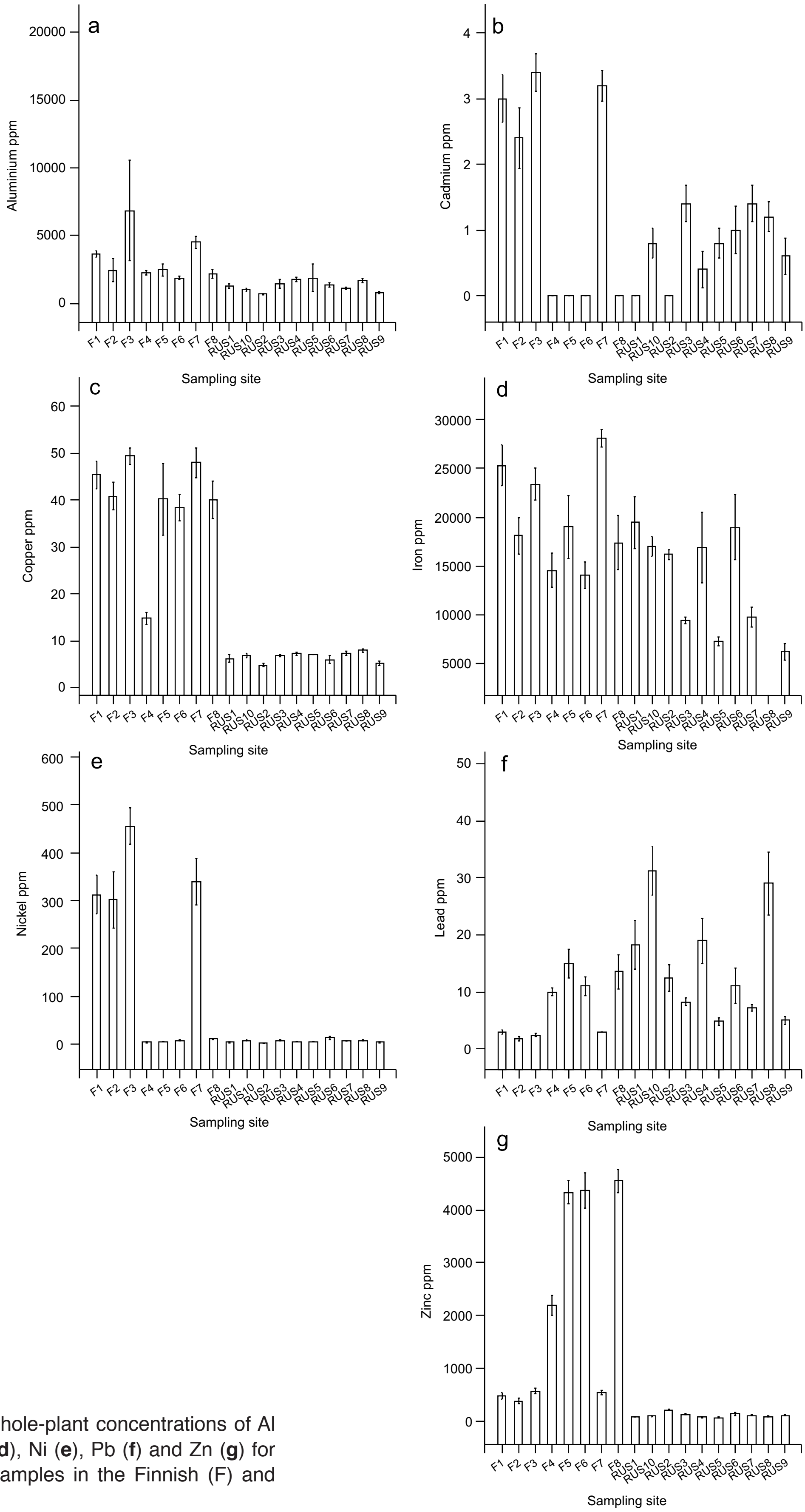

Fig. 2. Mean (±1 SE) whole-plant concentrations of Al (**a**), Cd (**b**), Cu (**c**), Fe (**d**), Ni (**e**), Pb (**f**) and Zn (**g**) for *Fontinalis antipyretica* samples in the Finnish (F) and Russian (RUS) streams.

between 1235–19904 μg g^{-1} and 557–5494 in the streams of Finland and Russia, respectively. The respective averages were 3312 and 1315 μg g^{-1}, with statistically significantly higher values in the Finnish mosses (Mann-Whitney U-test, $p <$ 0.001). Considerable variation in aluminium concentrations occurred between the Finnish stations, while variation was negligible in Russian (Fig. 2a). Recoverage of aluminium in AAS-analyses was poor, 28% to 46% of the certified values, which should be noted when interpreting the results.

Cadmium concentrations in mosses averaged 1.54 and 0.76 μg g^{-1} in Finland and Russia, which was a statistically significant difference (Mann-Whitney U-test, p = 0.004). There was greater inter-stream variability in the Cd results in Finland than in Russia (Fig. 2b). In Finland, four sites had elevated Cd concentrations (around 3 μg g^{-1}), while in others the concentrations were quite low. However, the concentrations were more even and mostly under 1 μg g^{-1} in Russian streams. High recoverages, 92.3%–124.2%, of the certified values were reached in Cd measurements.

In copper analysis, the recoverage percentage ranged between 105% and 112% of certified values indicating high validity of results. In Finland mosses had significantly higher average copper concentrations than in Russia (Mann-Whitney U-test, $p <$ 0001). The concentrations were 12–60, and 4–9 μg Cu g^{-1} in Finland and Russia, respectively. Inter-stream variation in copper concentrations was considerable for the Finnish streams, while the Russian streams exhibited only minor variation (Fig. 2c).

The average iron concentrations in mosses were 10 421–31 346 and 4413–30 462 μg g^{-1} in the Finnish and Russian streams, respectively. The average iron concentration was significantly higher in the Finnish streams (20 053 μg g^{-1}) than in the Russian ones (13 457 μg g^{-1}) (Mann-Whitney U-test, $p <$ 0.012). In both countries, iron concentrations varied largely between the sites (Fig. 2d). In the Finnish streams, however, the average concentrations exceeded 20 000 μg g^{-1} at several sites, while concentrations in Russian mosses were always clearly below this level. Certified iron values are lacking, but the repeatability of the analysis was good and measures can be considered valid.

Also in nickel analysis the validity of the results is reflected by good recoverage values: 89%–111% of the certified reference values. Very high variation of the moss Ni concentrations was observed in Finland, whereas variation was very low in Russian streams (Fig. 2e). Concentrations were 3–551, and 2–22 μg g^{-1} in Finnish and Russian mosses, respectively. The average value in Finnish streams (184 μg g^{-1}) was statistically significantly higher (Mann-Whitney U-test, $p <$ 0.001) than in the Russian streams (5.8 μg Ni g^{-1}).

Lead measurements yielded 82%–96% recoverage of the certified values. Contrary to other metals, lead values were significantly higher in Russia than in Finland (Mann-Whitney U-test, $p <$ 0,001). The average values were 14.6 and 7.3 μg Pb g^{-1} in Russian and Finnish streams, respectively. Variation between sites was considerable in both countries (Fig. 2f). Zinc measurements yielded 101%–107% recoverage of the certified values. Concentrations were 263–5025, and 64–247 μg Zn g^{-1} in Finland and Russia, respectively. The Finnish zinc average (2117 μg Zn g^{-1}) was significantly higher (Mann-Whitney U-test, $p <$ 0.001) than the Russian one (116 μg Zn g^{-1}). This large difference was mainly due to the highly elevated concentrations at four of the Finnish stations, although average concentrations were always higher in the Finnish streams when compared to Russian streams (Fig. 2g).

Discussion

In general, metal concentrations in surface waters are affected by natural soil and bedrock sources as well as by anthropogenic loading from atmospheric deposition and various point and non-point sources (Förstner and Wittman 1979). In our study area, northern Karelia, atmospheric deposition and point loading are insignificant as sources of metals, while geochemical sources and land-use activities are more important (Tarvainen *et al.* 1997, Skjelkvåle *et al.* 1999). There was a general tendency for significantly higher metal concentrations in Finnish streams than in Russian ones. We suggest that three hypotheses may explain this phenomena: natural geochemical variability generates the differences, intensive forestry has enhanced mobilization and leaching of metals into the streams in Finland, or the combination of the differences in geochemistry and silvicultural practices causes the differences. According to the Finnish geochemical atlas, there are some metal anomalies in the bedrock of the study area. Especially zones of archaean supracrustal rocks, such as mafic greenstone, are found in the study catchment (Koljonen 1992). Greenstones contain high amounts of many metals, including Al, Cu, Fe, Ni and Zn, and they contribute to the elevated concentrations of these metals in soils and surface waters (Koljonen 1992, Tarvainen *et al*. 1997). Since increased metal mobilization is commonly observed in the areas where drainages disturb mineral soils and lands containing metal anomalies (Edén *et al*. 1999, Heikkilä 1999), it is possible that intensive forest drainages and the construction of the forest road network have enhanced leaching of the metals from the greenstones into the streams in our study area. Unfortunately, geochemical information from the Russian part of the catchment was not available to us. Hence, the role of geochemistry in stream metal contamination needs to be clarified in future studies.

Metal concentrations in *Fontinalis antipyretica* were highly variable even in a small restricted area within the Finnish side of the catchment. The three hypotheses mentioned above may also explain this variation, but should be further evaluated with a detailed GIS-analysis on the catchment properties, including bedrock geochemistry and the land-use history. Although we did not have such information available in this screening study, it is known that clear cuttings, scarcifications, drainages, fertilization and the construction of the forest road network have been conducted mainly during the last four decades with varying intensity and timing in different parts of the catchment (Mononen *et al*. 1990).

In summary, silvicultural practices, as the dominant land-use forms prevailing on the Finnish side of the study area, are likely to play a significant role in metal contamination of the streams. Concentrations of many metals in *Fontinalis* on the Finnish side of the catchment clearly exceeded natural background concentrations and suggest major impact of land-use. Results from many earlier studies support this conclusion (*see* the references below). Forest drainage and clearfelling are known to be factors which increase the load of aluminium to water courses (Reynolds *et al* 1992, Vuori *et al*. 1998). The concentrations of aluminium in the Russian moss samples were mainly at the level typical for stream sites with minor anthropogenic impact. On the other hand, concentrations of Al in many Finnish stream sites were comparable to the Finnish streams affected by effluents from acid sulphate soils and forest drainages (Vuori *et al*. 1998b). Comparable aluminium levels have also been measured in *Hygrohypnum ochracheum* growing in acidified streams of the French Vosges Mountains (Claveri *et al*. 1995). The highest reported aluminium concentrations of lotic mosses exceed 25 000 μg g^{-1} measured by Engleman and McDiffett (1996) in streams impacted by acid mine effluents. However, we stress that the interpretation of Al results should be done cautiously, due to the difficulties in the AAS-analysis of aluminium from moss samples. Only a fraction of Al can be extracted with the digestion method utilized here.

Manifold increases in iron load to watercourses has been reported after the initiation of catchment silvicultural practices (Vuori 1995, Ahtiainen and Huttunen 1999). In this study, the moss iron concentrations in the Finnish streams were generally similar or higher than previously measured in the whole *Fontinalis* plants collected from streams impacted by forest drainages (Joensuu *et al*. 1997). However, iron contamination of the forest streams appears to be significantly milder than that of the streams receiving effluents from cultivated acid sulphate soils (Vuori *et al*. 1998b) or from mines (Engleman and McDiffett 1996). In addition, iron content in mosses varied largely between the sites in both countries (Fig. 2b). Reasons for this variation are not clear. It is known that the amount of humic matter is one of the key factors affecting concentration of iron in surface waters (e.g. Borg 1987, Verta *et al*. 1990), and forestry-induced humus loading (e.g. Ahtiainen and Huttunen 1999) may contribute to the variation in iron concentrations. In earlier studies, the background values of whole plant Cd content of *Fontinalis antipyretica* have been observed to be < 1 μg g^{-1} (Vuori *et al.* 1998b, Ukonmaanaho 1991). However, natural metal anomalies in the bedrock may result in elevated concentrations. In the study by Ukonmaanaho (1991), in the areas unaffected by air pollution in Lapland, the highest Cd values in *Fontinalis antipyretica* reached 4.9 μg g^{-1}. Claveri *et al.* (1995) reported relatively high concentrations (> 2 μg Cd g^{-1}) in mosses growing in acidified mountain streams. Much higher concentrations, up to 44–90 μg Cd g^{-1}, have been measured in the mosses from streams polluted by mine and industrial effluents (Wehr and Whitton 1983, Conçalves *et al.* 1994).

The highest zinc concentrations in Finnish streams can be considered exceptional, although much higher concentrations (> 10 000 μg Zn g^{-1}) have been measured in rivers polluted by industrial effluents (Wehr and Whitton 1983). Greenstone zones may cause elevated Zn concentrations in catchments (Lahermo *et al.* 1996), although Tarvainen *et al.* (1997) conclude that high Zn concentrations in headwater lakes and streams are seldom correlated with bedrock or till geochemistry. In addition, fertilizers increase the Zn load of catchments (Skjelkvåle *et al.* 1999).

Overall, the Cu concentrations of Russian moss samples are similar to values measured earlier in unpolluted arctic and boreal streams (Vuori 2002). The highest reported concentrations in *Fontinalis* have been measured in large polluted rivers (157–725 μg g^{-1}, Wehr and Whitton 1983, Conçalves *et al.* 1994). Concentrations in our study were well below this range also on the Finnish side of the catchment. However, the average Finnish values were comparable to the concentrations measured in the whole *Fontinalis* plants collected from streams polluted by runoff from acid sulphate soils and forest drainage areas (Vuori *et al.* 1998b). Lead was the only metal found at higher concentrations in Russian moss samples when compared to the Finnish ones. Similar and higher Pb values in whole *Fontinalis* plants have been measured both in unpolluted and polluted areas (Ukonmaanaho 1991, Vuori *et al.* 1998b). Claveri *et al.* (1995) reported higher Pb concentrations (88–189 μg g^{-1}) in aquatic mosses of acidified mountain brooks. Mine and industrial pollution have yielded much higher concentrations in *Fontinalis antipyretica* (228–17 800 μg g^{-1}, Dietz 1973, Wehr and Whitton 1983, Conçalves *et al.* 1994). High Pb values in surface waters are connected to high dissolved organic carbon concentrations, suggesting that peat lands act as sources of Pb in headwaters (Tarvainen *et al.* 1997). In natural peat lands *Sphagnum* mosses exhibit both strong heavy metal accumulation capacity and high preference for binding Pb (Breuer and Melzer 1990). We did not have exact data of relative proportions of different peat lands, but the wetlands in the Russian side are known to be predominantly in a natural state, while practically all peat lands in the Finnish side are drained. Hence, differences in the relative proportion of natural peat lands and those drained for silviculture may play a role in the observed difference in Pb concentrations.

Concentrations of nickel in *Fontinalis antipyretica* in Russian streams were at the level reported earlier from uncontaminated boreal and Arctic streams (Ukonmaanaho 1991, Vuori 2002). On the Finnish side of the catchment the concentrations of some streams can be considered exceptionally high, representing levels measured earlier mainly in streams contaminated by mine effluents (Wehr and Whitton 1983, Conçalves *et al.* 1994). Tarvainen *et al.* (1997) concluded that geochemical sources control regional patterns of Ni in lakes and streams in areas with low atmospheric deposition. They also concluded that such land-use factors as ditching affect concentrations, but are difficult to distinguish from geochemical sources. Ni anomalies in lakes in eastern Finland correlated with high Ni concentrations of glacial till (Tarvainen *et al.* 1997). Overall, our results suggest that silvicultural practices in combination with specific geochemical features may cause metal contamination of streams in areas otherwise considered unpolluted. Naturally, we cannot rule out other potential anthropogenic factors, such as local contamination sources from past or present military activities in the region. The area was the scene of relatively heavy battles during the Second World War and today the border regions are closed military areas. However, since the highest metal concentrations were observed in the areas heavily modified by silviculture and were located far away from border guard detachments and known historical battle fields, this alternative is not plausible.

The results raise the question of ecological risks of the elevated metal concentrations in mosses. In our study, the metal content was measured from the whole plants. Despite careful rinsing of the mosses, they will always contain some unknown amount of inorganic and organic particles firmly attached to the branches and folded leaves (*see* Johnson 1978). Hence, the results more probably describe some kind of total contamination of the benthic habitats than real bioaccumulation of metals within the plants and potential toxic impacts on them. At least no visual signs of damage in mosses were observed. Harmful impacts could also be anticipated on benthic invertebrates. Liljaniemi *et al.* (2002) studied habitat structure and zoobenthic assemblages at the very same sampling stations. They detected only minor differences in the species richness and community structure between the two countries, although habitats were strikingly dissimilar. The communities were dominated by stress-tolerant taxons, suggesting that natural acidity and humic waters of the area largely determine the community structure in streams. Nevertheless, the results of Liljaniemi *et al.* (2002) suggest that the observed metal contamination does not cause major ecological impacts, maybe due to the low bioavailability of the metals found in moss habitats. While moss plants appear to be of minor nutritional value and are seldom used as food by benthic invertebrates (Vuori and Muotka 1999), they are not likely to act as a significant route of metals to benthic macroinvertebrates or fishes, unlike that reported in some circumneutral streams (Dallinger and Kautzky 1985).

However, our results stress the importance of taking precautions against forestry-induced metal contamination in forested catchments. In particular, maintenance ditching of the drainage network and the construction of forest roads should include careful evaluation of the geochemical metal anomalies and the application of proper water protection measures, such as effective overland flow areas, in order to decrease leaching of metals into streams.

Acknowledgments: We thank Alexander Freindling, Antero Koikkalainen and Petri Liljaniemi for assistance in the field. The financial support of the Ministry of Environment, Finland, is gratefully acknowledged.

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Received 17 September 2001, accepted 10 September 2002