Heavy-metal Pollution and Remediation of Forest Soil around the Harjavalta Cu-Ni Smelter, in SW Finland

Oili Kiikkilä

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Heavy metals and sulphur have been emitted from the Cu-Ni smelter at Harjavalta since 1945. This article reviews the work that has been published in scientific journals after 1975 concerning heavy metal deposition and the effects of pollution on forest ecosystem around Harjavalta. The pollution has had diverse effects on boreal forest ecosystem, e.g. vegetation, nutrient cycle mediated by microbiota and soil animals, herbivorous insects and pathogens, resistance mechanisms of vegetation, and birds. The deposition of heavy metals has increased up to 30 km distance from the smelter. At 8 km distance the ecosystem began to approximate an undisturbed ecosystem where only slight changes in the understorey vegetation were observed. At 4 km distance the species composition of different ecosystem components (vegetation, insects, birds, soil microbiota) had changed and the growth of trees was retarded. At 0.5–1 km distance, where the nutrient cycling was disturbed and only the most resistant organisms were surviving, the ecosystem had ceased to carry out its essential functions. Remediation through liming or mulching with organic matter, of forest soil has had some positive effects on the ecosystem.

Keywords copper, forest, ecosystem, Harjavalta, nutrient cycle, remediation, liming, mulch
Author's address Vantaa Research Centre, Finnish Forest Research Institute, P.O. Box 18, FIN-01301 Vantaa, Finland **E-mail** oili.kiikkila@metla.fi
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1 Introduction

The largest heavy metal polluted area in the northern hemisphere is situated in NW Russia in the Kola Peninsula where the area of forest death is 600–1000 km^2 (Oleksyn and Innes 2000). Another large area is found in Sudbury, Canada, where mining and smelting activities have created 170 km^2 of barren land (Winterhalder 2000). In Finland, the largest heavy metal polluted areas are situated around Tornio, NW Finland, and Harjavalta, SW Finland (Kubin et al. 2000), where the area of forest death is less than 1 km^2 (Salemaa et al. 2001).

The effects of heavy metals on ecosystems have been widely studied. Research in the Sudbury region started in 1970's (Hutchinson and Whitby 1974, Whitby and Hutchinson 1974). Recently intensive research have been done in the Kola Peninsula where the contamination (Lindroos et al. 1996, Nikonov et al. 2001), the effects on vegetation (Nöjd et al. 1996, Nöjd and Reams 1996) and insects (Kozlov et al. 2000, Kozlov and Whitworth 2002) have been studied. Numerous field experiments have been established to study the remediation of heavy metal polluted landscape. Remediation agents such as zeolites (Vangronsveld et al. 2000), lime (Mälkönen et al. 1999), sewage sludge (Kelly and Tate 1998), gravel sludge (Krebs et al. 1999), compost and beringite mixture (Vangronsveld et al. 1996, 2000) and compost and woodships mixture (Kiikkilä et al. 2001) have been found to ameliorate soil. The only large-scale attempt to remediate a heavy metal polluted landscape has been in Sudbury, where more than 30 km^2 of barren land has been revegetated after liming and fertilising (Winterhalder 1996).

The Harjavalta region is one of the most intensively studied heavy metal polluted areas. This article reviews the work on heavy metal deposition and the effects of pollution on the forest ecosystem around Harjavalta that has been published in scientific journals since 1975. First, studies on vegetation damage, and the effects on the nutrient cycle, mediated by microbiota and soil animals, are reviewed. Next, studies on herbivorous insects and pathogens, vegetation resistance mechanisms, and the effects of pollution on birds are reviewed. Finally, the remediation experiments are reviewed. The aim is to outline the extent to which the metal pollution affects the ecosystem.

2 Emissions, Deposition and Contamination

Harjavalta (61°19´N, 22°9´E) is part of the southern boreal coniferous zone. Harjavalta is situated on an esker that runs to the SE of the Cu-Ni smelter. The forest on the esker consists mainly of Scots pine, *Pinus sylvestris* L., and is situated on dryish, relatively nutrient-poor sites (Mälkönen et al. 1999). According to the Finnish forest site type classification (Cajander 1949), the forest along the esker varies from *Vaccinium* to *Calluna* type (Derome and Lindroos 1998b). According to Mälkönen et al. (1999) the soil is comprised of sorted glaciofluvial sediments and the texture of the mineral soil is classed as sorted fine or fine/coarse sand with no stones. The soil type is ferric podzol, with an E horizon ranging between 6 to 15 cm, a B_s horizon 26 to 39 cm, and an organic mor layer 1 to 3 cm, in thickness. Southerly winds prevail in the area and therefore the shape of the pollution field is an ellipse in the direction of the river valley from SE to

Table 1. Annual emissions from Harjavalta smelter. (Data from Outokumpu Harjavalta Metals Ltd).

Year	Dust	Cu	Ni	Zn	Pb	As
			t year^{-1}			
1984	1100	98	47	216	55	
1985	1100	98	47	216	55	
1986	1200	126	46	232	60	
1987	1800	140	96	162	94	
1988	1000	104	45	103	48	
1989	1000	80	33	190	70	
1990	960	80	31	160	80	
1991	640	80	14	90	45	
1992	280	60	10	12	9	
1993	250	50	7	13	6	11
1994	190	40	6	6	3	5
1995	70	17	1.4	1.7	0.5	0.2
1996	195	49	1.2	5.3	1.9	4.2
1997	360	70	3	14	4	10
1998	132	23	1.7	6.1	2.4	10
1999	48	6	0.8	4.2	1.0	1.8

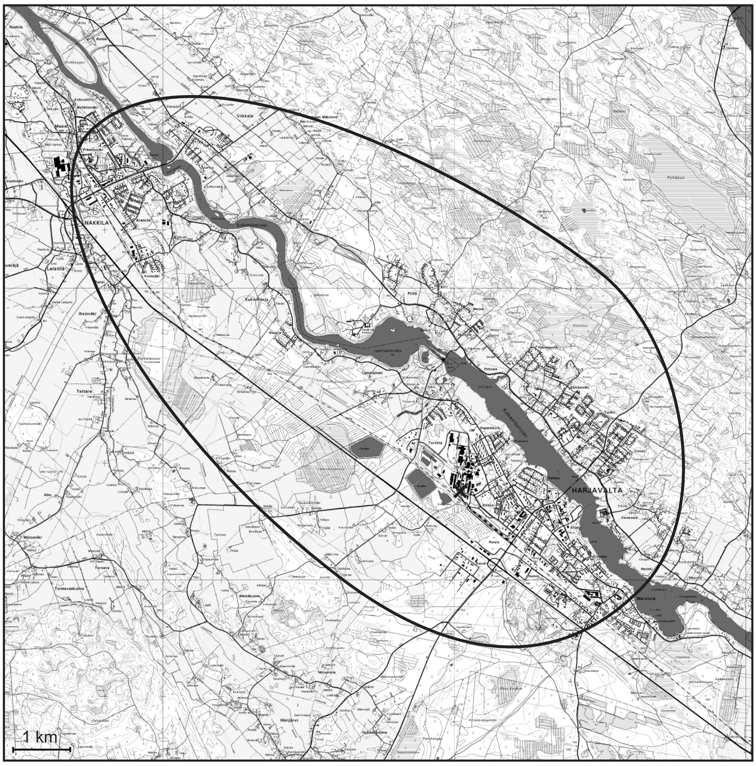

Fig. 1. The moderately polluted area. The ellipse indicates the area where the accumulation of copper in moss bags was over 5-fold compared to background areas in 1981–1982 (modified from Hynninen 1986).

NW (Laaksovirta and Silvola 1975, Hynninen and Lodenius 1986, Hynninen 1986) (Fig. 1). However, the pollution gradient which has often been examined in the studies runs to the SE of the Cu-Ni smelter.

A copper smelter started operating at Harjavalta in 1945 and a nickel smelter in 1960. In addition to copper and nickel, the emissions contain also zinc, lead, cadmium, arsenic, mercury, and sulphur (Table 1). Since the beginning of the 1990's there has been a considerable decrease in heavy-metal emissions as a result of technical modifications to the smelter complex and the construction of a taller smoke stack. This is reflected in the results of the heavy-metal moss surveys carried out in 1985, 1990 and 1995 (Kubin et al. 2000), which indicated a steep decrease in Ni concentrations in mosses and a clear, but less marked decrease in Cu concentrations, during the 10-year monitoring period. However, the bulk deposition near the smelter did not decrease between 1992 and 1996, probably because the dust from the degraded forest floor and slagheaps located nearby increased the deposition of metals (Nieminen et al. 1999).

Elevated Cu concentration in forest mosses was detected as far as 30–40 km from the smelter in 1995 (Kubin et al. 2000) (Fig. 2). With the moss bag method high airborne pollution of copper, nickel, zinc, lead, cadmium (Hynninen 1986) and mercury (Hynninen and Lodenius 1986) were observed in 1982 up to a distance of 9 km. Up to 4 km during 1992–1996 bulk deposition in open areas and stand throughfall, i.e. deposition inside the stand, was contaminated with sulphate and heavy metals (Derome and Nieminen 1998).

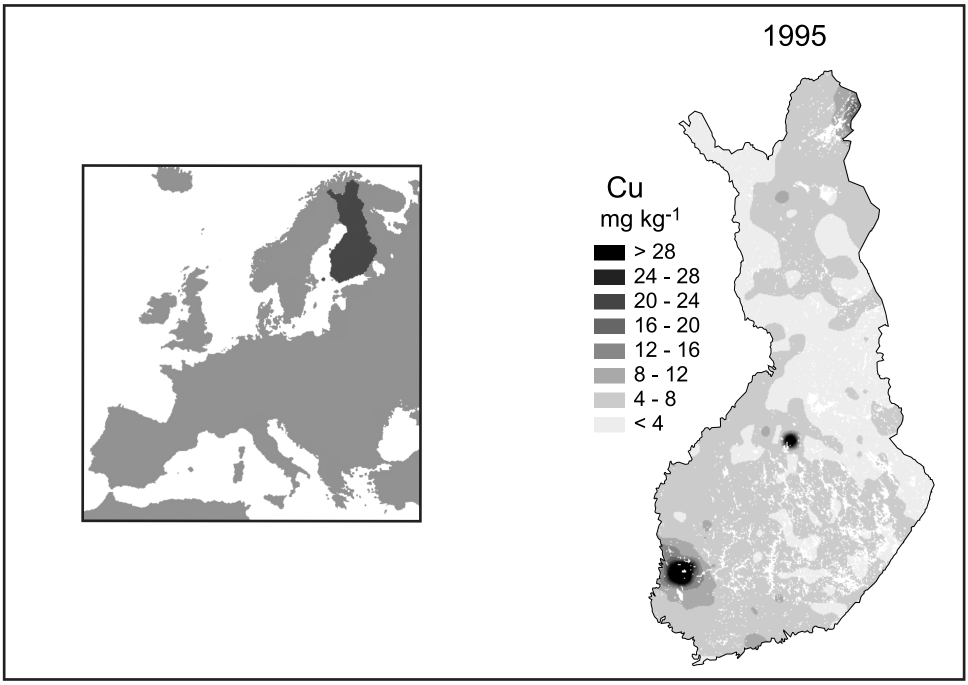

Fig. 2. The Cu concentration in forest mosses in Finland 1995 (reprinted from Kubin et al. 2000, with kind permission of Kluwer Academic Publishers).

The deposition of Cu was high close to the smelter the bulk deposition being 160 mg m^{-2} y^{-1}, and stand throughfall 360 mg m^{-2} y^{-1} (Derome and Nieminen 1998). At 8 km both the bulk deposition and stand throughfall were ca. 3 mg m^{-2} y^{-1}. The respective values for Ni close to the smelter were 70 and 140 mg m^{-2} y^{-1} and for Zn 20 and 40 mg m^{-2} y^{-1}, whilst at 8 km the values were ca. one tenth of these.

A clear, logarithmically decreasing gradient, studied at the distances of 0.5, 2, 4, and 8 km, was found in the soil for Cu, Ni, Zn, Cd, Fe, Pb, Cr and S (Derome and Lindroos 1998a and b). Elevated concentrations of heavy metals were also found in peatlands near the smelter (Veijalainen 1998, Nieminen et al. 2002) (Table 2). The total Cu concentration in the organic layer of the forest soil was 5800 mg kg^{-1} d.m. (dry matter) (Derome and Lindroos 1998b) and the exchangeable ($BaCl_2$ extractable) Cu concentration was 4700 mg kg^{-1} d.m. at the distance of 0.5 km from the smelter (Derome and Nieminen 1998). The respective values for Ni were 460 and 420 mg kg^{-1} d.m., and for Zn 520 and 130 mg kg^{-1} d.m. (Table 2). Total Fe concentration in organic soil was 18 600 mg kg^{-1} d.m. Uhlig et al. (2001) found extremely high total Cu concentration in organic soil under *Empetrum nigrum* patches, 49 000 mg kg^{-1} d.m. at 0.5 km and 12 000 mg kg^{-1} d.m. at 4.0 km, respectively. At 4 km the total Cu concentration was 660, Ni 124, Zn 137 and Fe 3200 mg kg^{-1} d.m. At the vertical scale, leaching of Cu, Ni, Zn and SO_4-S down to 40 cm depth in the soil profile was observed, Zn being the most mobile element and Cu being strongly bound to organic layer (Derome and Nieminen 1998).

No signs of soil acidification were found (Derome and Lindroos 1998b). The pH of the organic layer was 3.5 at 0.5 km and 3.6 at 8 km distance from the smelter. The respective values for exchangeable acidity were 91 and 85 meq kg^{-1} d.m.

In general Cu, Ni, Zn and Cd concentrations at different trophic levels have been reported to be

Table 2. Total concentrations of Cu, Ni, Zn and Cd in soil near the smelter (0.5–1 km). The concentration in the reference area (at 8 km or further) is in parentheses.

	Cu	Ni	Zn	Cd	Reference
		mg kg^{-1} d.m.			
Peat					Veijalainen 1998
Surface	3600 (170)	470 (50)	460 (86)	3.7 (0.9)	
0–10	1200 (75)	240 (16)	240 (16)	3.7 (0.9)	
10–20	180 (10)	75 (0)	110 (29)	1.5 (0.1)	
Peat					Nieminen et al. 2002
2 cm	4400 (6)	870 (3)	560 (53)		
14 cm	45 (5)	260 (7)	570 (60)		
Soil					
Organic layer	5800 (150)	460 (40)	520 (60)	4.6 (0.7)	Derome and Lindroos 1998b
Organic layer [a)]	4700 (120)	420 (37)	130 (47)		Derome and Nieminen 1998
Mineral soil [a)]					
0–5 cm	270 (1.5)	25 (0)	10 (1)		Derome and Nieminen 1998
5–10 cm	27 (0.4)	5.4 (0)	2.9 (0.4)		
10–20 cm	16 (0)	3.2 (0)	1.8 (0)		
20–30 cm	12 (0.2)	2.1 (0)	1.4 (0)		
Percolation water					
5 cm	0.6 (0.02)	0.5 (0.01)			Derome and Lindroos 1998a
20 cm	1.1 (0.01)	0.9 (0)			

[a)] Exchangeable ($BaCl_2$) metals

high near the smelter. Cu has usually been found in higher concentrations than Ni or Zn throughout the food chain although Zn has been emitted more than Cu until 1992 (Table 1). Elevated heavy metal concentrations have been reported in understorey vegetation (Helmisaari et al. 1995), pine (Derome and Nieminen 1998), spruce (Heliövaara and Väisänen 1991), birch (Koricheva and Haukioja 1995), insects (Heliövaara et al. 1990), spiders (Koponen and Niemelä 1995), and birds (Eeva and Lehikoinen 1995) (Table 4).

3 The Effects of Pollution on the Forest Ecosystem

3.1 Vegetation

The Scots pine forest stand close to the smelter has clearly suffered from pollution. The tree growth has been extremely poor (Mälkönen et al. 1999) and the understorey vegetation have drastically changed (Salemaa et al. 2001). The total coverage and the number of species has decreased towards the smelter and vegetation was almost absent up to a distance of 0.5 km from the smelter (Salemaa et al. 2001) (Table 3). On the most polluted sites, *Empetrum nigrum*, *Arctosthaphylus uva-ursi*, and *Vaccinium uliginosum*, clonal dwarf shrubs, have survived in small patches. Vigorous regrowth and phenotypic plasticity have improved the survival of *A. uva-ursi* and *V. uliginosum* (Salemaa et al. 1999). *E. nigrum* possesses an internal heavy metal tolerance (Monni et al. 2000a). However, decreased chlorophyll and organic acid, and an increased abscisic acid concentration in stems and leaves indicated a reduction in the physiological activity of *E. nigrum* near the smelter (Monni et al. 2000c). The tolerance mechanisms of *E. nigrum* may include accumulation of heavy metals in older tissues, the restriction of the metal transport to the green leaves (Uhlig et al. 2001), localisation of metals in certain cell compartments (vacuoles, cell walls, cytoplasm), possible detoxification of metals by phenolics (Monni et al. 2002), and accumulation and immobilisation of metals in the litter beneath *E. nigrum* patches

Table 3. The change in species abundance close to the smelter, – damaged, + benefit. The number in the parentheses refers to the distance where the point frequency % is more than 0.02. + in parentheses refers to the benefit in the moderately polluted area. The year in the parentheses refers to the study year.

Species		Reference
Vascular plants		
Arctostaphylus uva-ursi (L.) Sprengel	– (2)	Salemaa et al. 2001 (1993)
Calluna vulgaris (L.) Hull	– (4)	
Carex clobularis L.	(1) a)	
Empetrum nigrum L.	– (1)	
Pinus sylvestris	– (0.5)	
Vaccinium uliginosum L.	– (1)	
Vaccinium vitis idaea L.	– (1)	
Mosses		
Dicranum polysetum Sw.	– (8)	Salemaa et al. 2001 (1993)
Dicranum scoparium Hedw.	– (8)	
Cerantodon purpureus (Hedw.) Brid.	– (1)	
Polytrichum juniperum Hedw.	(1) a)	
Pleurozium schreberi (Brid.) Mitt.	– (8)	
Pohlia nutans (Hedw.) Lindb.	– (0.5)	
Ground lichens		
Cetraria islandica (L.) Ach.	– (2)	Salemaa et al. 2001 (1993)
Cladina rangiferina (L.) Nyl.	– (3)	
Cladina arbuscula (Wallr.) Hale&W.L.Club	– (3)	
Cladina stellaris (Opiz) Brodo	– (4)	
Cladonia spp.	– (2–3)	
Epiphytic lichens		
Hypogymnia physodes L.	– (4)	Fritze et al. 1989 (1987)
Pseudevernia furfuracea (L.) Zopf	– (4)	
Usnea hirta (L.) Wigg.	– (7)	
Bryoria fuscescens (Gyelnik) Brodo & Hawksw	– (7)	
Platismatia glauca (L.) Culb&Culb	– (7)	
Epiphytic algae		
Scoliciosporum chlorococcum	+	Fritze et al. 1989 (1987)
Endophytic fungi		
Cenangium ferrucinosum Fr:Fr	–	Helander 1995 (1992)
endophtic fungi total	–	Lappalainen et al. 1999 (1993–94)
Hormonema sp.	+	
Fusicaldium sp.	–	
Gnomonia setacea (Pers.) Ces. and de Not	–	
Soil animals		
Enchytraeids	–	Haimi and Siira-Pietikäinen 1996 (1993–94)
Microarthropods	–	
Collembolans	–	
Nematodes	–	
Bark bug		
Aradus cinnamomeus Panzer	– (+)	Heliövaara and Väisänen 1990a (1987–89)
Tortricid moths		
Retinia resinella L.	– (+)	
Rhyacionia pinicolana Doubleday	– (+)	
Blastesthia turionella L.	– (+)	
Blastesthia posticana Zetterstedt	– (+)	

Table 3 continued.

Species		Reference
Leaf- miners		
Eriocrania solitary species	–	Koricheva 1994 (1992–93)
Eriocrania cicatricella Zetterstedt	+	
Geometrid moth		
Epirrita autumnata Bkh.	–	Ruohomäki et al. 1996 (1990)
Aphids		
Cinaria pini L.	+	Heliövaara and Väisänen 1990a (1987–88)
Pineus pini Gmelin	+	Heliövaara and Väisänen 1989b (1987)
Schizolachnus pineti Fabricius	+	
Diprionid		
Diprion pini L.	– (+)	Heliövaara et al. 1990
Ants	+	Koponen and Niemelä 1995 (1992)
Formica fusca L. or *F. lemani* L.	+	Koricheva et al. 1995 (1993)
Beetles		
Xylechinus pilosus Ratzb.	+	Heliövaara and Väisänen 1991
Tomicus piniperda L.	+	
Pityogenes chalcographus L.	–	
Ground living beetles	–	Koponen and Niemelä 1995 (1992)
Coccinella septempunctata L.	+	
Mites		
Aceria leionotus Nalepa	–	Koricheva et al. 1996 (1993)
Aceria longisetosus Nalepa	–	
Acalitus rudis Canestrini	–	
Spiders		
Xerolycosa nemoralis	+	Koponen and Niemelä 1993
Alopecosa aculeata	–	
Oedothorax apicatus	+	
Erigone atra	+	
Agyneta rurestris	+	
Zelotes petrensis	–	
Tapinocyba pallens	–	
Silometopus elegans	–	
Walckenaeria antica	–	
Walckenaeria atrotibialis	–	
Birds		
Parus major L.	–	Eeva and Lehikoinen 1996 (1993)
Ficedula hypoleuca Pallas	–	

[a] Rare, except at the distance in the parentheses

(Uhlig et al. 2001). Of the dwarf shrubs, *Calluna vulgaris*, growing first at 1.2 km to the NW of the smelter, proved to be least resistant to Cu (Monni et al. 2000b). Although germinable seeds of *C. vulgaris, Betula pubescens*, *Pinus sylvestris* and *V. uliginosum* were found in the most contaminated soil, seedlings of trees and dwarf shrubs were absent close to the smelter (Salemaa and Uotila 2001).

With regards to moss, *Pohlia nutans* and *Ceratodon purpureus* were the only moss species surviving in small patches on the most contaminated site (Salemaa et al. 2001) (Table 3). Although the frequency of *Pleurozium schereberi* and *Dicranum* spp. began to increase at 8 km (Salemaa et al. 2001) the Cu concentration in their tissues were considerably higher than those in background areas (Helmisaari et al. 1995). The reindeer lichens (*Cladina* spp.) appeared to be more tolerant than forest mosses, they increased in frequency at 4 km (Salemaa et al. 2001). Epiphytic lichens were absent up to 2 km, on an area of 8.8 km^2, in 1970 (Laaksovirta and Silvola 1975) and up to 4 km in 1987 (Fritze et al. 1989).

3.2 Nutrient Cycling

Inhibition of nutrient cycling and the displacement of base cations from cation exchange sites by Cu and Ni cations has resulted in a decrease of base cation (Ca, Mg, K) concentrations in the organic layer (Derome and Lindroos 1998b). Trees have not been able to utilise the nutrient pools in the mineral soil presumably due to the toxic effects of Cu and Ni in the plant fine roots, including ectomycorrhizal root tips (Helmisaari et al. 1999) since Mg, Ca, and Mn concentrations in Scots pine needles were low (Derome and Nieminen 1998). In contrast, trees obtained sufficient K from the soil, since despite K leached from the needle tissues close to the smelter, the needle K concentrations were relatively high (Nieminen et al. 1999). Autumnal nutrient retranslocation, i.e. transport of nutrients from the senescing needles to the remaining organs for overwinter storage, of P and K in Scots pine was less efficient close to the smelter than at 8 km (Nieminen and Helmisaari 1996). The retranslocation of nutrients was suggested to be inhibited by non-pathogenic endophytic fungi by Ranta (1995). However, endophytes seemed not to be a reason for the decreased nutrient retranslocation since the number of endophyte infected needles was lower close to the smelter than further away (Helander 1995).

3.2.1 Soil Decomposer Community

The number of soil animals has clearly decreased and their community structure strongly altered close to the smelter (Haimi and Siira-Pietikäinen 1996). Since at 2 km the number of soil animals has only slightly decreased, soil animals appeared to be quite resistant to heavy metals. An indication of increased Cu resistance of the enchytraeid worm, *Cognettia sphagnetorum*, Vejdovsky, usually the only abundant enchytraeid species found in northern coniferous forest soils, has been found near the smelter (Salminen and Haimi 2001). It seems that the presence of patches of lower metal concentrations was mitigating the effects of the metals on worm populations (Salminen and Haimi 1999).

The overall microbiological activity in the soil has decreased drastically near the smelter. Microbial respiration activity, physiological groups of bacteria (Fritze et al. 1989), and microbial and fungal biomass (Fritze et al. 1996) decreased towards the smelter. The toxicity of soil to *Photobacterium phosphoreum* increased towards the smelter (Vanhala and Ahtiainen 1994). At 4 km distance the structure of the microbial community had changed and the bacterial community was resistant to heavy metals but the microbial activity was on the level of unpolluted sites (Pennanen et al. 1996, Fritze et al. 1997). The fungal part of the microbial biomass was more sensitive to heavy metals than bacterial part (Pennanen et al. 1996). The decreased microbial activities have been reflected in a decreased rate of litter decomposition which could be seen as a changed structure of the humus layer (F + H) and as a 6–8 cm thick layer of accumulated brown needle litter on the top of the forest floor near the smelter (Fritze et al. 1989). The rate of litter decomposition has been influenced by the accumulation of Cu, Ni and Zn in brown needle litter and root litter, collected at the site (McEnroe and

Helmisaari 2001, Nieminen and Saarsalmi 2002). The accumulation of metals into decomposing unpolluted needle litter was also observed thus retarding the decomposition rate near the smelter (Ohtonen et al. 1990).

3.3 Herbivores and Pathogens on Trees

The adverse effects caused by forest pests increased with pollutant load as bark bugs, diprionids, tortricids, aphids, and bark beetles were abundant in the moderately polluted pine stands (Heliövaara and Väisänen 1990a), and near the smelter the Scots pines were heavily infested by aphids and bark beetles – *Xylechinus pilosus* being the most abundant bark beetle species in spruce and *Tomicus piniperda* in pine (Heliövaara and Väisänen 1991) (Table 3). Close to the smelter the cocoons of the defoliator species were smaller than further away (Heliövaara and Väisänen 1989a) but the smaller females produced more viable eggs (Heliövaara and Väisänen 1990a). Many insect species, however, suffered from severe pollution. *Pityogenes chalcographus*, which is one of the most common bark beetle species associated with spruce in Finland, was almost absent near the smelter (Heliövaara and Väisänen 1991). Also bark bugs, diprionids and tortricids, were scarce in the immediate vicinity of the smelter (Heliövaara and Väisänen 1990a). Insects such as a moth *Epirrita autumnata* (Ruohomäki et al. 1996) and a gall mite species on birch (*Betula pubescens* and *B. pendula*) (Koricheva et al. 1996) were also scarce near the smelter (Table 3). In contrast, densities of mites on European aspen (*Populus tremula* L.) were not affected by the pollution (Koricheva et al. 1996).

Great differences in metal concentrations between the insect species feeding on Scots pine were observed near the smelter (Table 4) (Heliövaara et al. 1987). The highest concentration was measured in a sap-feeding aradid bug (*Aradus cinnamomeus*), the Cu concentration being 800 mg kg^{-1}. The lowest Cu concentration was measured in a gall-forming tortricid moth (*Retinia resinella*), 40 mg kg^{-1} (Heliövaara et al. 1987). Metal levels were higher in the needles than in the insects *Neodiprion sertifer*, except in the case of Cd. Cd accumulated in the insects, the concentration in the adults was 2.6 mg kg^{-1} which is twice that in their food (1.3 mg kg^{-1}) and much higher than in their faeces (0.7 mg kg^{-1}) (Heliövaara and Väisänen 1990b). The low nutritional quality and high pollutant contents of pine needles increased the mortality of diprionids (Heliövaara and Väisänen 1990d) although outbreaks of diprionids were also common (Heliövaara et al. 1991). The susceptibility of *Neodiprion sertifer* to virus and other diseases increased near the smelter but the mortality of *N. sertifer* caused by parasitoids decreased.

Means of defence against herbivores for trees include the production of resin and the phenolics in the bark, phloem, and foliage (Kytö et al. 1998). Phenolics can also act as antidesiccation agents (Loponen et al. 1997). The resin flow decreased towards the smelter, indicating a decreased defence level, but the phenolic concentration increased, as a response to pollution, on Scots pine (Kytö et al. 1998) and on birch (Loponen et al. 1997). Compensatory growth, as a response to simulated herbivore, of two willow species, *Salix borealis* (Fries.) Nasar. and *S. caprea* L., was reduced near the smelter (Zvereva and Kozlov 2001). The endophytic fungal flora may affect their host plants positively by enhancing the resistance of the plant to pathogens (Butin 1992). Suppression of these non-pathogenic endophytes by air pollution did not promote the development of pathogenous *Gremniella abietina* (Lagerb.) Morelet, causing Scleroderris canker disease (Ranta et al. 1994).

Increased or decreased densities of leaf-miner species, which as pathogens are of minor importance, have been recorded around the smelter. The solitary *Eriocrania* species (Koricheva and Haukioja 1992) were found to be scarce whilst the gregarious *Eriocrania cicatricella* (former *E. haworthi*) was abundant near the smelter (Koricheva and Haukioja 1994). Several aspects which could be related to population density of the leaf-miners on heavy metal polluted areas were studied, such as: host plant quality (Koricheva and Haukioja 1992, 1995), larval parasitism (Koricheva 1994), ant predation of miners (Koricheva et al. 1995), and the densities of endophytic fungi (Lappalainen et al. 1999). Only host plant quality, i.e. heavy metal

Table 4. The concentrations of Cu, Ni, Zn and Cd in different plant species, cocoons of the insects, ants, spiders and faeces of birds near the smelter. The concentration in the reference area is in parentheses.

Species	Cu	Ni	Zn	Cd	Reference
		mg kg^{-1} d.m.			
Pohlia nutans	1390 (270)				Helmisaari et al. 1995
Empetrum nigrum					
Last annual shoot	180 (20)				Helmisaari et al. 1995
	86 (22) a)	30 (13)	50 (16)	0.5 (0.1)	Uhlig et al. 2001
Older living parts	1500 (30)				Helmisaari et al. 1995
	340 (90) a)	120 (40)	220 (40)	1.1 (0.5)	Uhlig et al. 2001
Cladina arbuscula	160 a) (60)				Helmisaari et al. 1995
Picea abies (L.) Karsten					
Bark	600 (40)	100 (15)	300 (180)	1.2 (1.1)	Heliövaara and
Phloem	75 (6)	80 (6)	340 (170)	1.1 (1.2)	Väisänen 1991
Wood	6 (1)	8 (1)	40 (10)	0.2 (0.1)	
Pinus sylvestris					
Bark	1500 (30)	390 (9)	190 (21)	5.6 (0.5)	Heliövaara and
Phloem	66 (6)	35 (5)	120 (56)	5.1 (1.5)	Väisänen 1991
Wood	11 (3)	6.9 (1.2)	25 (7.8)	0.7 (0.2)	
Trunk wood	0.9 a) (1.1)	0.4 (0.2)	7.9 (5.2)	0.3 (0.3)	Harju et al. 1997
Needles	500 (10)	140 (10)		1.3 (0.2)	Heliövaara and Väisänen 1990b
Needles	210 (9)	44 (5)	83 (33)		Derome and Nieminen 1998
Stems (1–22 years)	2 (0.4)				Helmisaari et al. 1995
Fine roots	480 (75)				
Fine roots	590 (21)	110 (15)	70 (90)	2.1 (0.6)	Helmisaari et al. 1999
Betula pubescens Ehrh.					
Foliage	96 (10)	51 (10)	250 (210)		Koricheva and Haukioja 1995
Betula pendula Roth.					
Foliage	64 (10)	40 (10)	220 (180)		
Aradus cinnamomeus b)	800 (40)	110 (10)		13 (7)	Heliövaara et al. 1987
Retinia resinella	40 (5)	7 (2)		1.6 (0.2)	
Panolis flammea					
Denis and Schiffermüller	70 (10)	10 (1)		2 (0.1)	Heliövaara and
Bupalus piniarius L.	90 (10)	1.6 (0)		0.6 (0.1)	Väisänen 1990c
Diprion pini L.	70 (10)	8 (1)			Heliövaara et al. 1990
Gilpinia socia Klug	60 (20)	10 (2)			
Neodiprion sertifer					Heliövaara and Väisänen
Geoffroy	80 (20)	7 (1)		2 (0.5)	1989c
Gilpinia virens Klug	60 (10)	5 (0)		1 (0)	
G. frutetorum Fabricius	90 (20)	8 (2)		2 (0)	
Microdiprion pallipes Fallén	130 (20)	20 (2)		4 (1)	
Ground living ants b)	300 (30)			6 (4)	Koponen and Niemelä 1995
	180 (20)	30 (5)			Eeva and Lehikoinen 1996
Ground living spiders	2000 (800)			20 (20)	Koponen and Niemelä 1995
Parus major	320 (50)	45 (5)	550 (350)		Eeva and Lehikoinen
Ficedula hypoleuca	420 (70)	55 (5)	700 (250)		1996

a) 4–6 km distance, b) adults

concentrations in birch foliage, was found to correlate with the densities of the species which either increased or decreased towards the smelter (Koricheva and Haukioja 1992 and 1995). The authors suggest that *E. cicatricella* possess higher tolerance for pollutants than solitary species.

Some changes in the ground living arthropod fauna have also been reported. Beetles, except *Coccinella septempunctata*, were scarce near the smelter (Koponen and Niemelä 1993 and 1995). Differences in diversity and species composition of spiders, ants and bugs was observed along the pollution gradient although there were no differences in the total numbers (Table 3).

3.4 Birds

During 1991–1997, the survival (Eeva and Lehikoinen 1998) and behaviour (Eeva et al. 2000a) of two hole-nesting passerines, Pied Flycatcher (*Ficedula hypoleuca* Pallas) and Great Tit (*Parus major* L.) were studied around Harjavalta. *F. hypoleuca* was more susceptible to pollutants than *P. major*, the response of which was weaker in many aspects. The breeding success of *P. major* was below background levels up to 3–4 km from the smelter (Eeva and Lehikoinen 1996) whilst *F. hypoleuca* was affected severely only next to the smelter (ca 1 km) (Eeva and Lehikoinen 1995, 1996). No clear differences in the female condition (Eeva et al. 1997b), and in the density of ectoparasites in the nests (Eeva et al. 1994) of these two bird species in relation to the pollution were found. The different responses of these two bird species were probably due to their different diet (Eeva and Lehikoinen 1996).

The poor breeding success of *P. major* was suggested to be related to habitat changes that have taken place around the smelter, e.g. a scarcity of suitable insect food for nestlings (Eeva and Lehikoinen 1996). The proportion of green larvae in nestling diet was smaller (Eeva et al. 1997a) and the nestling were lighter (Eeva et al. 1998) in the vicinity of the smelter than further away. Air pollution was found to fade the yellow colour in plumage of the *P. major*. Pale plumage might affect mate choice, and predict reduced winter survival (Eeva et al. 1998). However, better wintering conditions next to human habitation may in general compensate for the possible detrimental effects of pollutants on the *P. major* population (Eeva and Lehikoinen 1998).

The low local survival rate of *F. hypoleuca* adult females was suggested to be caused by higher emigration from the low quality habitat (Eeva and Lehikoinen 1998). However, *F. hypoleuca* nestlings were directly affected by increased amounts of heavy metals and the low availability of calcium-rich food items in their diet near the smelter (Eeva et al. 2000b). The pollution related stress of *F. hypoleuca* was detected in biomarkers from blood and liver (Eeva and Lehikoinen 1998) and as growth abnormalities of legs and wings and changes in egg shell quality near the smelter (Eeva and Lehikoinen 1995, 1996). The authors suggest that heavy metals might accumulate more in ground living, mobile, often adult, prey items of *F. hypoleuca* than in foliage living, less mobile, often larval, prey items of *P. major*. The concentrations of Cu, Ni and Pb in ants were higher close to the smelter than further away (Table 4) and correlated positively with *F. hypoleuca* nestling faecal concentrations (Eeva and Lehikoinen 1996). Close to the smelter the heavy metal concentrations in ground living ants and spiders (Koponen and Niemelä 1995) were higher than the concentrations of defoliator species (e.g. Heliövaara and Väisänen 1990c) (Table 4).

4 Remediation of Forest Soil

The aims in remediation have been to immobilise heavy metals, to improve the availability of nutrients, to promote decomposition of soil organic matter, and to stabilise nutrient cycling for a long period. Mälkönen et al. (1999) started a soil remediation experiment in 1992. Treatments consisted of liming, applying a slow release mineral mixture, and stand-specific fertilisation determined on the basis of needle and soil analyses (Mälkönen et al. 1999). Liming had positive effects on soil chemistry during the 5 study years. It increased exchangeable Ca and Mg concentrations (Derome 2000) and reduced exchangeable Cu and Ni concentrations in the soil (Mälkönen et al. 1999) and decreased leaching of metals down the soil profile (Derome and Saarsalmi 1999). Positive effects on tree growth and survival were also detected, liming being the most successful treatment. All the fertiliser treatments increased volume growth of Scots pine (Mälkönen et al. 1999) and liming increased the growth and survival of fine roots (Helmisaari et al. 1999), reduced the detrimental effects of heavy metals on experimental seedling survival (Salemaa and Uotila 2001), alleviated pollution stress of Scots pine assessed by needle fluctuating asymmetry (Kozlov et al. 2002), and increased the phenolic concentration of the phloem, indicating an improvement in the defence level against pathogens of Scots pine (Kytö et al. 1998). Liming did not affect the soil decomposer animal community (Haimi and Mätäsniemi 2002) but increased microbial respiration activity (Fritze et al. 1996). Liming also changed the structure and the metabolic profile of the microbial community (Fritze et al. 1997).

Another remediation experiment was started in 1996. Polluted forest floor was mulched with organic matter, a mixture of compost and woodchips, and seedlings of *Empetrum nigrum*, *Arctostaphylos uva-ursi*, *Betula pendula* and *Pinus sylvestris* were planted in pockets filled with mulch. The mulch was spread directly over the layer of undecomposed plant litter on the forest floor or on top of the exposed mineral soil following the removal of the polluted litter layer and organic soil layer. The chemical and microbial changes in organic soil during 3 growing seasons after mulching were reported by Kiikkilä et al. (2001). Mulching the polluted soil with the mixture of compost and woodchips converted copper into less toxic forms, which was detected as lower exchangeable Cu concentration in the soil, and a lower Cu^{2+} concentration in the soil water, as well as a decreased toxicity of soil water to bacteria. The microbial response to remediation was clear. Microbial activities increased and tolerance of the bacteria to Cu decreased in the organic layer. Mulching the forest floor after the removal of the polluted organic layer had a similar but greater influence on Cu speciation and the toxicity of percolation water (Kiikkilä et al. 2002). However, also the leaching of Cu down the soil profile was the highest for this treatment. The changes in soil chemistry and the success of revegetation during 4–7 years following mulching will be reported.

The decreased emissions is reflected in the decreased concentrations of heavy metals in forest mosses between 1990 and 1995 (Kubin et al. 2000). However, the decreased emissions were not reflected in the bulk deposition, soil solution, needle biomass, or radial growth of the trees between 1992 and 1996 (Mälkönen et al. 1999) probably because of the pools of accumulated metals in the ecosystem (Nieminen and Saarsalmi 2002). However, decreased emissions together remediation actions have probably benefited birds. The breeding success of *Parus major* and *Ficedula hypoleuca* has markedly improved in the vicinity of the smelter between the years 1991 and 1997, and the lead concentrations in nestling have decreased by about 90% during this time (Eeva and Lehikoinen 2000).

5 Conclusion

The effects and mechanisms of heavy metal deposition on the forest ecosystem are diverse. The deposition of heavy metals has increased up to 30 km distance from the smelter. At 8 km distance the ecosystem began to approximate an undisturbed ecosystem where only slight changes in the understorey vegetation were observed. At 4 km distance the species composition of different ecosystem components (vegetation, insects, birds, soil microbiota) had changed and the growth of trees was retarded. At 0.5–1 km distance, where the nutrient cycling was disturbed and only the most resistant organisms were surviving, the ecosystem has ceased to carry out its essential functions.

The main findings were i) copper was strongly bound to organic layer and seemed to be the main pollutant in the ecosystem while zinc was the most mobile element and did not accumulate to any specific part of the ecosystem, ii) the forest mosses and epiphytic lichens were the most sensitive plant species and the seedlings of the vascular plants that had survived near the smelter were absent, iii) there was a highly resistant soil decomposer community near the smelter although the activity of the soil animals and microbiota was low and their community structure altered, iv) many insect species also suffered from pollution although the adverse effects caused by forest pests increased with pollutant load, v) the low survival rate and breeding success of hole-nesting passerines near the smelter was caused by habitat changes and the quality of food, vi) the adverse effects seem to be to some extent reversible after decreased pollutant load or remediation actions.

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