Amelioration and restoration of Scots pine stands close to the Harjavalta Cu-Ni smelter in SW Finland

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Abstract

Although emissions of SO_2 and heavy metals from the Cu-Ni smelter at Harjavalta, SW Finland, have been strongly reduced in recent years, the topsoil of neighbouring forested sites still contains 50 years' accumulation of a wide range of heavy metals. Scots pine stands growing close to the smelter are suffering from serious defoliation and growth retardation, and microbial activity and mineralization in the forest floor are strongly retarded. A severe shortage of plant-available nutrients in the organic layer is accompanied by relatively high levels of immobilised macronutrients (e.g. Ca, Mg) in the organic layer. In order to alleviate and counteract the detrimental effects of heavy metal accumulation on stand health and vitality, the Finnish Forest Research Institute has established a number of field experiments with correction fertilizers, liming and the addition of an organic mulch (municipal compost/wood chips) to the forest floor. The effects of the treatments on tree growth, needle biomass, needle nutrient status, litterfall, fine root formation, deposition chemistry, soil microbial activity and community structure, and soil and soil solution quality were monitored during the latter half of the 1990's. Correction fertilization with a mixture of slow- and fast-release mineral nutrients (excluding N) and liming had a positive effect on soil and soil solution chemistry, and on fine root growth, but relatively little effect on the growth and nutrient status of the tree stand. In most parts of the pine stands the forest floor is completely devoid of ground vegetation, and the addition of an organic mulch has had an extremely positive effect on recolonisation of the site. The organic mulch has also considerably increased the concentrations of macro- and micronutrients in the soil solution.

Background

Copper and nickel are produced at the Harjavalta smelter in SW Finland by the flash-smelting pyrometallurgical process in two separate smelters. Copper smelting first started in 1945, a sulphuric acid plant was built at the site in 1947, and nickel smelting started in 1959. The copper and nickel ores smelted at the plants contain sulphur, heavy metals and arsenic. Emissions during the period from the founding of the smelters up until the end of the 1980's were relatively high, and comprised gases (SO_2) and particulate material, mainly consisting of Fe, Cu, Ni, Zn, Pb, Cd and As. Although emissions of SO_2 and heavy metals from the Cu-Ni smelter at Harjavalta have decreased considerably since the early 1990's as a result of the construction of a new, taller stack and installation of modern filters, the topsoil of neighbouring forested sites still contains 50 years' accumulation of a wide range of heavy metals.

Scots pine stands growing close to the smelter are suffering from serious defoliation and growth retardation, the needles have low nitrogen and especially magnesium concentrations (Derome & Nieminen 1998), fine root mortality is high (Helmisaari et al. 1999) (Fig. 1), the understorey vegetation in the stands has been almost completely destroyed (Salemaa et al. 2001), there is considerable accumulation of undecomposed needle and other plant litter on the forest floor (Fritze et al. 1989), microbial activity and mineralization in the forest floor are strongly retarded (Fritze et al. 1996), the soil bacterial community has become adapted to the elevated heavy metal concentrations

(Pennanen et al. 1996), and there is considerable accumulation in the organic layer of Fe, Cu, Ni, Zn and a number of other heavy metals. Despite the considerable SO_2 emissions during the past 50 years, no signs of soil acidification have been found in the organic layer or uppermost mineral soil layers. However, there has been an increase in exchangeable acidity and Al deeper in the mineral soil (20-40 cm layer), presumably due to the displacement of H^+ and Al^{3+} from the overlying organic and mineral soil layers. The loss in base saturation in the organic and uppermost mineral soil layers is correspondingly due to the displacement of base cations (Ca^{2+}, Mg^{2+}) by Cu^{2+} and Ni^{2+}, and not to the effects of acidic deposition. The accumulation of Cu and Ni in the soil has resulted in a severe deficit of plant-available Ca, Mg and K in the organic layer, caused by partial inhibition of the mineralisation of these nutrients from litterfall, and the displacement of base cations from exchange sites (Derome & Lindroos 1997).

The amelioration and restoration experiments

Although the area suffering from severe damage is relatively small, and is restricted to the immediate vicinity of the smelter, stand growth and the understorey vegetation have been affected over a distance of up to 4 km from the smelter. The research carried out in the area has concentrated on two questions – the restoration of pine ecosystems in the immediate vicinity of the smelter by means of liming and fertilization treatments and revegetation measures, and the correction of pollution-induced nutrient imbalances in the pine stands at greater distances from the smelter through fertilization and liming.

Reducing heavy metal toxicity and correcting nutrient imbalances through liming and fertilization

In order to alleviate and counteract the detrimental effects of heavy metal accumulation on stand health and vitality, and to counteract nutrient imbalances in the pine stands, the Finnish Forest Research Institute (Metla) established four field experiments with correction fertilizers and liming in Scots pine stands on dry-ish, relatively nutrient-poor sites at distances of 0.5, 2, 4 and 8 km along a transect running to the SE from the smelter. The liming and fertilizer treatments were applied on 30 x 30 plots using a random design with three replications of each treatment. The soil type in all the experiments was ferric podzol, with an E horizon 6 – 15 cm thick and a Bs horizon 26 – 39 cm thick. The thickness of the organic layer varied from 1 – 3 cm. The correction fertilizer treatment consisted of a mixture of slow- and fast-release mineral fertilizers, and the stand-specific fertilizer treatment was formulated to correct excessive soil acidity and/or nutrient deficiencies as determined on the basis of soil and needle analyses (Table 1).

The treatments that included limestone (LT and SSF) strongly decreased bioavailable Cu and Ni concentrations in the organic layer up to a distance of 4 km from the smelter, and the LT treatment increased the exchangeable Ca and Mg concentrations (Derome & Saarsalmi 1999). These treatments had a corresponding effect on the soil solution collected at a depth of 20 cm. The liming treatments had only a very small pH-increasing effect on the organic layer and soil solution close to the smelter, presumably due to the neutralizing effect of the relatively high exchangeable Fe concentrations (>5% d.w.) in the organic layer (Derome 2000). Liming increased both the amount of microbial biomass and evolution of CO_2 in the organic layer (Fritze et al. 1996) and changed the community structure (Fritze et al. 1997), but the other treatments had no effect. All the treatments increased volume growth of the pine stand at distances of 2 and 4 km from the smelter, while the increase at 0.5 km was of minor importance because the growth on the control plot at this site was only a fraction of the "normal" level at 8 km. The LT and CT treatments had only marginal effects on the needle biomass at the different sites. However, the SSF treatment, which included nitrogen, increased the needle biomass rather strongly at distances of 0.5 and 2 km. The treatments had no significant effect on the extremely low needle Mg concentrations at 0.5 km, but the SSF treatment (including N) significantly increased the needle N concentrations at all sites (Mälkönen et al. 1999). The LT treatment clearly increased the growth and survival of fine roots at 0.5 km, which was the site where fine root mortality on the control plots was extremely high (Helmisaari et al. 1999).

The restoration experiments

The main aim of the restoration experiments was to promote the recovery of the pine ecosystems through the establishment of a functioning organic layer, and to revegetate the sites with seedlings of native tree and dwarf shrub species. Recovery of the ecosystems presupposed successful restoration of the following stages and processes: 1) seedling root development in pockets of unpolluted soil, 2) recovery of litter production and improvement in litter quality, 3) recovery of microbial activity, 4) initiation of mineralization and nutrient cycling, and 5) reducing the bioavailability, mobility and leaching of heavy metals. Individual studies were carried out on soil biota in both the field and the laboratory, as well as the chemical properties of the soil and soil solution, and the community structure and plant life histories of the vegetation.

The restoration experiment was established in a seriously damaged pine stand in the immediate vicinity of the smelter in summer 1996. A 5 cm-thick layer of mulch was added to half of the plots in the experiment. The mulch was spread directly over the layer of undecomposed plant litter on the forest floor. The mulch consisted of a mixture of municipal compost and woodchips (1:1, volume). The compost was 14 months old and had been produced in outdoor windrows from a mixture of organic household waste and coarse woodchips (diam. ca. 5 cm). The experiment was supplemented one year later on a number of new plots: the polluted organic layer was removed and the mulch was spread directly on top of the exposed mineral soil (Kiikkilä et al. 2001). Plate lysimeters were installed at depths of 20 and 40 cm in the experiments in order to monitor the effects of mulching on percolation water quality. In addition to the routine water analyses, copper in the samples was also fractionated into free copper (Cu^{2+}) and organically complexed copper (Kiikkilä et al. 2002a). In addition to the mulching treatment, a number of tree and dwarf shrub seedlings/cuttings (*Pinus sylvestris*, *Betula pubescens*, *Empetrum nigrum*, *Arctostaphylos uva-ursi*) were planted in mulch pockets on the mulched plots. Planting the seedlings in mulch pockets penetrating down into the underlying contaminated soil was considered to be essential for their survival.

Addition of the organic mulch resulted in the transport of large amounts of organic matter down into the underlying contaminated mineral soil, and an initial increase in the leaching of nitrogen (NO_3), base cations (Ca, Mg and especially K) and also Cu, but <u>not</u> Ni or Zn. The increase in dissolved organic carbon (DOC) concentrations lead to an increase in the concentration of organically complexed Cu in the soil solution. Mulching was found to have no direct effect on the response of soil microbes to Cu toxicity in the laboratory microcosm experiment (Kiikkilä et al. 2002b). Mulching in the field promoted the conversion of soil solution Cu into forms (e.g. organic complexes) that were less toxic to soil microbes (Kiikkilä et al. 2002a). In addition, mulching increased the microbial activities and decreased the tolerance of bacteria to Cu in the organic layer.

Planting the cuttings and seedlings in mulch pockets on both the mulched and untreated plots was relatively satisfactory. All the *Empetrum nigrum* cuttings survived on the mulched plots, and the mortality on the untreated plots was only 2% during 1996-2000. The mortality of *Arctostaphylos uva-ursi* was higher (mortality 52 and 60%, respectively), probably due to the use of smaller cuttings. The mortality rate of the pine seedlings was very low, < 10%. The mortality rate of the birch seedlings on the mulched plots was higher than that on the control plots, 44% vs. 13%. However, this was due to hare damage, and in fact probably indirectly reflected a feeding preference (more leaves, higher palatability) for the birch seedlings on the mulched plots.

<u>Conclusions</u>

The results of these experiments indicate that the restoration of severely damaged pine ecosystems cannot be achieved through the use of fertilizers and liming alone, but that the addition of unpolluted organic material is essential to achieve the development of new seedling material and understorey vegetation, and to bring about the recovery of nutrient mineralization and cycling within the ecosystem. Such organic material provides a source of macro- and micronutrients for the vegetation, and decreases Cu toxicity through the formation of organic complexes. The addition of limestone below the mulch should be tested as a means of ensuring long-term immobilization of heavy metals. Dwarf shrubs appear to be very suitable for the revegetation of heavily polluted, relatively infertile soils owing to their low mortality in nutrient-poor soils, clonal growth habit that facilitates rapid spreading and coverage of the forest floor, dormant bud activation and rapid regrowth that increase resistance to heavy metals, and the fact that herbivorous animals usually tend to avoid such unpalatable species. The use of liming and correction fertilizers appears to be a promising method of reducing the bioavailability of heavy metals and providing a source of rapid and slow release nutrients (especially N and Mg) in stands that are not severely affected by the presence of heavy metals in the soil, but which are suffering from nutrient imbalances.

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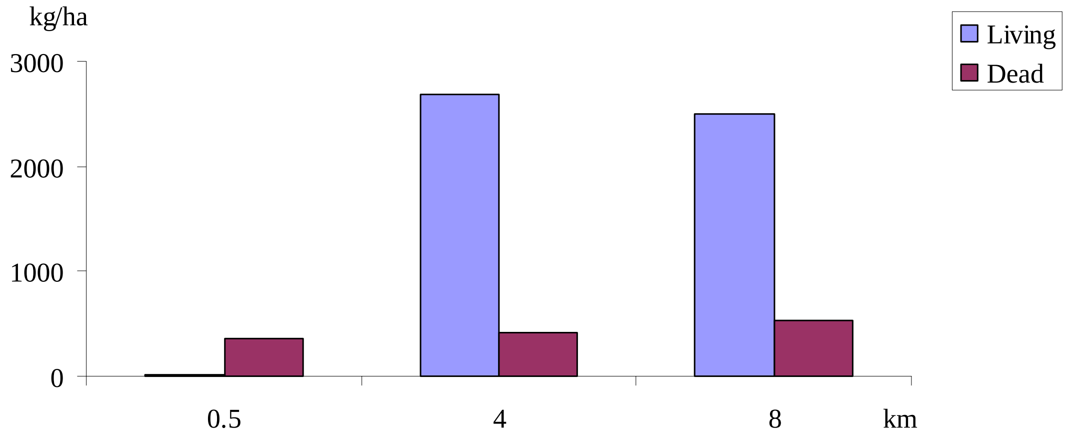

Fig. 1. Dead and living fine root mass in ingrowth cores (1994 - 1995) inserted in the soil at different distances from the smelter (Mälkönen et al. 1999).

Table 1. The liming and fertilizer treatments used in the experiments. MU = methylene urea, AN = ammonium nitrate. * = the SSF treatment included P fertilizer in the experiment at 4 km only.

Treatment	Nutrient application, kg/ha								
	N	P	K	Ca	Mg	S	B	Cu	Zn
Liming (LT)				420	100		2.0		
Correction fert. (CF)		31	56	98	61	37	1.3	0.8	0.8
Stand-specific fertilizer (SSF):									
0.5, 2 and 4 km	75 (MU) 75 (AN)	30*		315	50				
8 km	60 (MU) 60 (AN)								