

Effect of Mulching on Uptake of Copper and Nickel from Smelter-Polluted-Soil by Planted Tree Seedlings

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ABSTRACT

Our aim was to determine the long-term effect of a mulching treatment on copper (Cu) and nickel (Ni) uptake by tree seedlings (*Pinus sylvestris* L. and *Betula pubescens* Ehrh.) from smelter-polluted forest soil in southwestern Finland. A mulch cover spread onto polluted barren soil did not have a clear positive impact on the biomass production and it did not decrease metal uptake by planted tree seedlings during a ten-year study period. In contrast, the Cu uptake by the above-ground parts of birch was increased as a result of mulching, although there were weak indications of slightly reduced availability of Cu and Ni to roots in the case of both species. As Cu and Ni concentrations of foliage and bark have been shown to be strongly affected by surface deposited metal containing aerosols, only the woody compartments were used as indicators of metal uptake from soil. The Cu:Ni ratios of woody compartments were lower than those predicted by the Cu:Ni ratios of soil suggesting that the soil extraction method used gives an underestimation of available Ni in relation to Cu. The lower soil Ni_{exch} concentrations on the mulched plots compared to the controls were in agreement with the slightly lower root Ni concentrations in the mulch treatments.

Keywords: Bioavailability; Biocompost; Birch; Forest Soil; Pine; Restoration; Wood Chips

1. Introduction

Several methods have been developed to ameliorate site conditions and enhance plant establishment and growth on metal contaminated environments. Different remediation agents, such as lime and fertilizers [1], compost and beringite mixture [2] and application of mulch [3-6] have been used in environmental restoration approaches. In addition to providing nutrients into soil, the high organic matter content of biowaste composts improves the water-holding capacity, cation exchange capacity and nutrient availability of soil, which in turn improve tree growth [7].

Plants reveal different patterns in the uptake of trace elements [8,9]. The vascular plants take up elements mainly by their roots from the soil, even if the foliar uptake of gases and soluble elements may also take place [10]. Trees have been suggested as a low-cost, sustainable and ecologically sound solution to the remediation of heavy metal-contaminated land [11]. Benefits arise mainly from stabilization of the soil, but in many cases,

trees may be sufficient to provide clean-up of the soil. Before the beneficial effects can be obtained, the trees must become established on a site [12]. However, on highly contaminated soils, tree establishment may be inhibited by metal toxicity. In less contaminated soils, other factors may limit plant growth, such as macronutrient deficiencies [13] and physical conditions, especially those properties leading to poor water holding, aeration and root penetration [14].

The effects of mulching have been reported to vary greatly according to site, plant species and mulch types [15,16]. However, only few studies on long-term effects of mulching on plant metal uptake have been reported. It is known from an earlier paper based on our study fields that mulching favored establishment of transplants and enhanced natural recolonization by pioneer species [17]. The aim of this paper was to assess the long-term effect of mulching on biomass production and Cu and Ni uptake by seedlings of pine (*Pinus sylvestris* L.) and birch (*Betula pubescens* Ehrh.) grown for a ten-year-period on a metal-contaminated site.

2. Methodology

2.1. Study Site

The study site at Harjavalta (61°19'N, 22°9'E), south-western Finland has been subjected to a heavy pollution load from a large metallurgical complex for several decades. Smelting of copper started in the area in 1945 by Outokumpu Oy, while the nickel smelter and refinery were established in 1959. The surrounding heathland Scots pine forests are suffering from severe needle loss and growth retardation [18,19] and high fine root mortality [20]. The understory vegetation is almost completely degraded [21,22] and even though viable seeds have been found in the forest soil close to the smelters, no seedling rooting takes place [23].

The long term (1960-1990) mean annual temperature at a nearby weather station of the Finnish Meteorological Institute is +4.0°C and the annual precipitation 558 mm. The mean annual temperature and annual precipitation during the study period (1996-2005) were 5.3°C ± 0.6°C and 591 mm ± 62 mm, respectively. The metallurgical plants are located on a forested esker running in a NW-SE direction. The soil consists of sorted fine or sorted fine/coarse sand with no stones. The soil was classified as an orthic Podzol [24]. The uppermost part of the forest floor consists of a dark thick layer of undecomposed litter, as a result of strongly retarded microbial activity and impaired mineralization [25].

2.2. Remediation Experiment

In June 1996, tree seedlings were planted at a distance of ca. 500 meters from the main stack of the Cu-Ni smelters. Seedlings of two native species, *Betula pubescens* Ehrh. (1-year-old containerized downy birch seedlings) and *Pinus sylvestris* L. (2-year-old containerized Scots pine seedlings), were each planted on six replicate plots (5 × 5 m²) as 49 seedlings per plot. Three of the plots were totally covered with a 5 cm-thick layer of mulch, and the other three were left uncovered to serve as controls. In addition, six replicate plots (5 × 5 m²) without any transplants were established to serve as reference sites for soil characteristics. Three of them were covered with a 5 cm-thick layer of mulch, and the other three were left uncovered. The location of the experimental plots was randomized. One of the uncovered plots was unintentionally destroyed when slag was spread over it.

The pine and birch seedlings were planted in soil pockets (2 L, depth about 20 cm) containing mulch. Planting the seedlings in the mulch pockets penetrating down into the less contaminated soil was considered to be essential for their initial survival [17].

The mulch consisted of a mixture of household biocompost and woodchips (1:1, volume). The biocompost was 14 months old and had been produced in outdoor

windrows at the Ämmässuo Waste Handling Centre, Espoo, Finland by mixing kitchen and garden waste from the Greater Helsinki area and coarse woodchips (diameter ca. 50 mm). The mulch was prepared one week before spreading by mixing the biocompost with woodchips (diameter < 20 mm) of Scots pine and Norway spruce (*Picea abies* Karst.) stemwood [26]. The pH of the mulch was 6.3 and the carbon:nitrogen ratio 16:1 [26]. The average Cu and Ni concentrations in the Ämmässuo biocompost were 60 and 3 mg·kg⁻¹ as dry weight [26]. The mulch was spread directly on the layer of undecomposed litter with a plotwise dose of biocompost (excluding the woodchips) of 5.4 kg·m⁻² as dry weight. The input of C through mulching was 2 kg·m⁻² [26].

2.3. Harvest of the Seedlings and Soil Sampling

After a 10 year period, 3 seedlings from each experimental plot (3 replicate plots for both control and mulching) were harvested in August 2005. One of the seedlings was chosen among the tallest individuals, the second to represent the smallest ones, and finally the third one to represent the medium size. After removing the foliage, the youngest shoots (formed in 2005) and all roots were separated from the seedlings, and bark was carefully peeled away from the remaining part to obtain 5 compartments: foliage, young shoots, bark, wood, roots. The pine needles were grouped according to the year of their formation: Current, current + 1 year, current +2 and current +3 + older needles.

The soil samples were collected with an auger (diameter 58 mm) at the same time as seedlings were harvested. Three cores per plot were taken and after removing the organic layer the mineral soil was divided in layers of 5-cm-thickness (0 - 5, 5 - 10, 10 - 15, 15 - 20, 20 - 25, and 25 - 30 cm).

2.4. Analytical Methods

The plant samples were dried and weighed. After weighing the three replicates of each compartment from each plot were bulked together to give one composite sample of each compartment per plot, thus resulting in three replicate composite samples per treatment. The organic layer samples of the soil cores were dried and milled to pass through a 1 mm sieve, and thereafter they were divided into two parts for the total and exchangeable analysis. Total Cu and Ni concentrations from the organic soil and plant samples were determined, following microwave assisted wet digestion in HNO₃ and H₂O₂, by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES).

Exchangeable Cu and Ni were determined by extraction with 0.1 M BaCl₂ + 2% EDTA, (7.5 g of mulch or 15 g of mineral soil/150 ml extractant, shaking for 2

hours) followed by filtration and analysis by ICP-AES.

2.5. Statistical Analyses

We used the two way t-test to study the effect of the treatment on biomass and metal concentrations and the between-species variation.

3. Results

3.1. Biomass Production

There were no statistically significant differences in biomass production during the ten-year period between the mulch and control treatments, although the mean biomasses of both pine and birch tended to be slightly higher on the mulched plots (**Figure 1**).

3.2. Metal Concentrations

3.2.1. Foliage

The Cu and Ni concentrations of pine needles tended to increase with age but no statistically significant differences could be found between the treatments (**Table 1**). In case of birch the Cu and Ni concentrations of the leaves were slightly higher in the mulch treatment, but the differences were not statistically significant (**Table 1**).

3.2.2. Bark

The highest Cu and Ni concentrations of all the sampled compartments were those of pine bark from the control plots (**Figure 2**). The t-test showed that the between-treatment difference was statistically significant for the Cu concentrations ($t = 4.039$; $p = 0.016$).

The mean Cu concentration of the birch bark was slightly higher on the mulch plots, but the difference was not statistically significant. The corresponding Ni con-

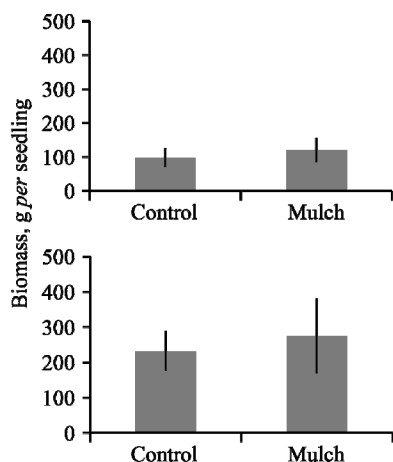


Figure 1. Mean total biomass of the sampled tree seedlings by treatments, $n = 9$ for both species. The bar indicates the standard error of the mean.

Table 1. Mean (\pm standard error of the mean) Cu and Ni concentrations of pine needles by needle age classes (C = current needles, C + 1 one-year-old needles, C + 2 two-year-old needles and C + 3 three-year-old needles), $n = 3$ in treatments.

Pine needle age class		Copper ($\text{mg}\cdot\text{kg}^{-1}$)		Nickel ($\text{mg}\cdot\text{kg}^{-1}$)	
		Control	Mulch	Control	Mulch
C + 3	mean	323	237	71.1	61.0
	st err	± 133	± 24.0	± 27.5	± 6.95
C + 2	mean	435	393	73.7	78.23
	st err	± 70.4	± 76.3	± 9.02	± 8.49
C + 1	mean	292	325	66.6	76.03
	st err	± 19.1	± 68.9	± 5.15	± 10.19
C	mean	116	124	46.0	42.3
	st err	± 3.18	± 18.0	± 1.54	± 2.90
Birch leaves	Mean	467	712	127	156
	St err	64.6	104	23.0	14.7

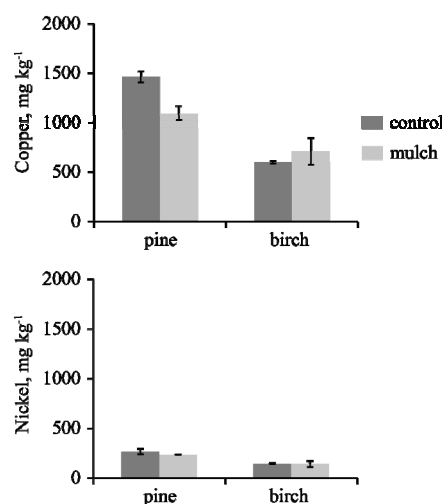


Figure 2. Mean Cu and Ni concentrations in bark of pine and birch by treatments, $n = 3$. The bar indicates the standard error of the mean.

centrations were about the same for both treatments (**Figure 2**).

3.2.3. Above Ground Woody Compartments and Roots

The Cu and Ni concentrations of the youngest shoots were much lower than those of bark, but there were no statistically significant differences between the treatments (**Figure 3**). The Cu and Ni concentrations in pine wood appeared to be slightly higher on the control plots, but the differences were not statistically significant (**Fi-**

gure 4).

The mean Cu and Ni concentrations in both pine and birch roots were slightly higher on the control plots compared to the mulch treatment, but the differences were not statistically significant (Figure 5).

3.2.4. Soil Concentrations

The Ni_{exch} concentrations in the mineral soil layers tended to be lower in the mulched plots compared to the control, while the differences between the treatments in the Cu_{exch} concentrations of the mineral soil were small (Figure 6). The Ni_{exch} concentrations were much lower than the Cu_{exch} concentrations, roughly ten times lower.

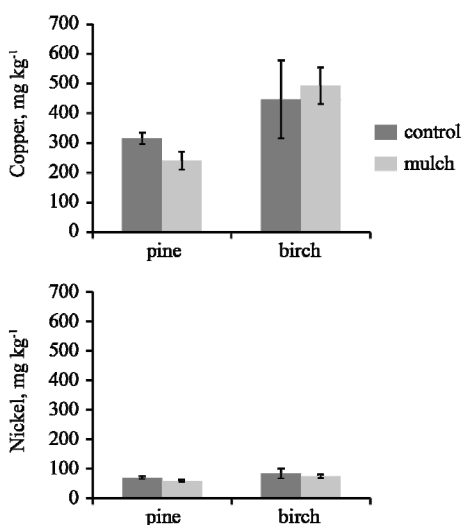


Figure 3. Mean Cu and Ni concentrations in the youngest shoots of pine and birch by treatments, n = 3. The bar indicates the standard error of the mean.

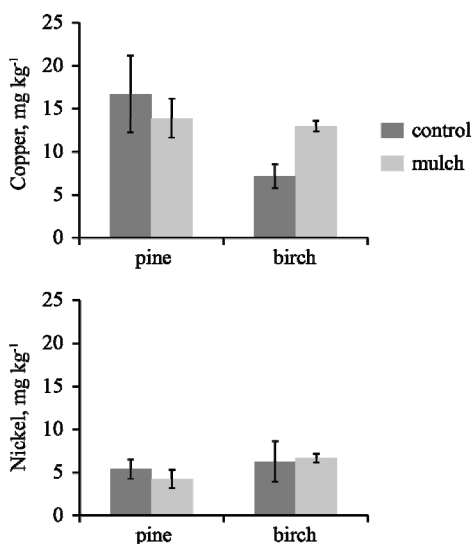


Figure 4. Mean Cu and Ni concentrations in wood (without bark) of pine and birch by treatments, n = 3. The bar indicates the standard error of the mean.

4. Discussion

4.1. Biomass Production and Metal Uptake

Both Scots pine and downy birch are considered as metal tolerant species, since they are able to survive in metal-polluted areas around smelters [17,27-29]. Restricted uptake of metals by roots and low translocation into foliage is the most common resistance trait [30]. The mulch cover is supposed to restrict the metal uptake of seedlings by orientation of their roots into this layer containing less metals and to protect plant roots from drought and to provide a source of nutrients [17,31]. However, our results are not in agreement with these

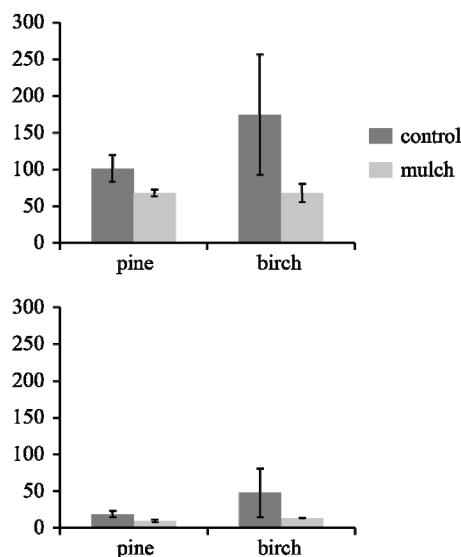


Figure 5. Mean Cu and Ni concentrations in roots of pine and birch by treatments, n = 3. The bar indicates the standard error of the mean.

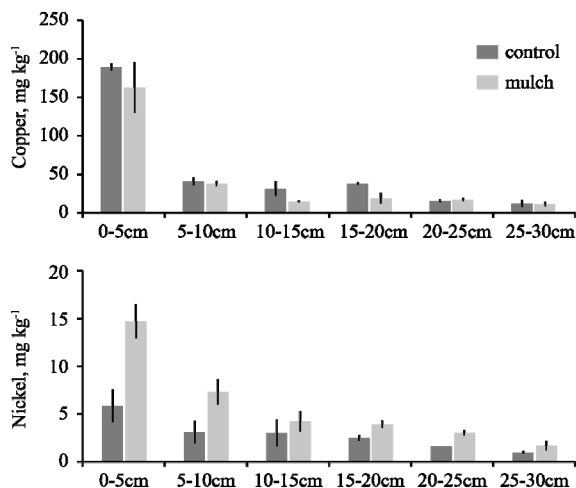


Figure 6. Vertical distribution of mean exchangeable Cu and Ni in the soil profile by treatments, n = 3. The bar indicates the standard error of the mean. Please, note the different scale of the vertical axis for Cu and Ni.

earlier findings. We found no enhancement of biomass production by mulching, and the mulch layer did not decrease the availability of Cu and Ni to the seedlings. In contrast, the Cu concentrations in birch wood increased by the mulch treatment. As the root Cu concentration did not increase, the results suggest increased root-to-shoot mobility of Cu due to mulching. It appears that some of the Cu compounds formed through complex formation by organic molecules supplied by mulching would be more readily translocated from birch root to shoot than the Cu forms at the control plots.

4.2. Soil Extraction as a Predictor of Cu:Ni Uptake Ratio

Clearly more Cu than Ni was taken up by pine, which is in agreement with the higher Cu concentrations in relation to the Ni concentrations measured from the soil samples. However, the soil Cu concentrations were almost 10 times higher than the soil Ni concentrations, while the wood Cu concentrations of pine were only 3 times higher than those of Ni. In birch, the Cu and Ni concentrations in the wood of the control seedlings were equal and in the mulch treated seedlings the Cu concentrations were only twice as high as those of Ni. Hence, the BaCl₂ + EDTA extraction schema used by us as soil extraction method appeared to give an underestimation of both the birch and pine available amount of Ni in relation to Cu. The BaCl₂ method is reported to give an indication of immediately exchangeable metals [32,33], while the use of EDTA has been reported to give a good estimation of potentially plant available metal fractions [34].

4.3. Surface Deposition Affected Cu and Ni Concentrations

The Cu and Ni concentrations in the compartments subjected directly to aerial deposition (bark, young shoots, and foliage) were clearly higher than those of wood and roots. A high proportion of these metal concentrations is caused by aerial deposition of dust that accumulates on the plant surfaces and do not penetrate into the living tissues [18,35,36]. Thus, high amounts of heavy metals on plant surfaces do not necessarily pose any acute toxic hazard to plant metabolism.

Tree bark is known to sorb and accumulate airborne contaminants and therefore, it has been largely used for monitoring of atmospheric pollution [37-39]. In our study the whole bark layer, including the living inner bark, was taken by peeling it completely from the tree shoots. The inner bark metal concentrations reflect the phloem sap flow. The Cu and Ni concentrations in the bark obtained in our study are roughly hundreds of times higher than the nationwide mean values (3.6 and 1.1 mg·kg⁻¹, respectively) reported by Lippo *et al.* [35]. Also Saarela *et*

al. [39] found lower metal concentrations (Cu 89 mg·kg⁻¹ and Ni 18 mg·kg⁻¹) than our values in Scots pine bark sampled during forest felling 6 kilometers northeast from the Harjavalta smelters.

Scots pine needle Cu concentrations ranging from 1.7 to 270 mg·kg⁻¹ and Ni concentrations from 1 to 262 mg·kg⁻¹ have been found in a 350 km-long transect extending from the Monchegorsk smelter complex, NW Russia, through Finnish Lapland to the Finnish-Swedish border [40-43]. We found even higher concentrations, Cu ranging from 100 - 600 mg·kg⁻¹ and Ni from 50 - 140 mg·kg⁻¹ in our study than the values reported from the Kola gradient.

5. Conclusion

The effect of adding a mulch layer on metal availability to tree seedlings was species dependent. The mulching had no clear effect on the Cu and Ni availability to pine, while Cu uptake by birch was enhanced on the mulch treated plots. In addition, although generally more Cu than Ni was taken up by the tree seedlings, the Ni uptake rate was higher than what could be predicted on the basis of the ratio of soil exchangeable Cu and Ni concentrations.

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