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Elevated Cd and Zn uptake by aspen limits the phytostabilization potential compared to five other tree species

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Abstract

Phytostabilization of metals using trees is often promoted although the influence of different tree species on the mobilization of metals is not yet clear. This study examined effects of six tree species on the soil characteristics pH, organic carbon (OC) content and cation exchange capacity (CEC) and on the redistribution of cadmium (Cd) and zinc (Zn) on a polluted sandy soil. Soil and biomass were sampled in 10-years-old stands growing on former agricultural land. The tree species included were silver birch (*Betula pendula*), oak (*Quercus robur* and *Q. petraea*), black locust (*Robinia pseudoacacia*), aspen (*Populus tremula*), Scots pine (*Pinus sylvestris*) and Douglas fir (*Pseudotsuga menziesii*). In the short period of ten years, only aspen caused significant changes in the soil characteristics. Due to accumulation of Cd and Zn in its leaf litter, aspen increased the total as well as the NH_4OAc-EDTA-extractable Cd and Zn concentrations in the topsoil compared to deeper soil layers and to other tree species. Also topsoil pH, OC content and CEC were significantly higher than under most of the other species. This caused rather low 'bio-available' $CaCl_2$-extractable concentrations under aspen. Nevertheless, given the risks of aboveground metal dispersion and topsoil accumulation, it is recommended that aspen should be avoided when afforesting Cd and Zn contaminated lands.

Keywords: phytostabilization, trees, trace metals, sandy soil, biomass

1. Introduction

Industrialization has effectuated significant emissions of trace metals into the global environment. Subsequent deposition of these metals and accumulation in the soil poses serious risks for ecosystems and public health through leaching of metals to groundwater and dispersion in the food chain (WHO, 2000). In the Campine region in northern Belgium, zinc (Zn) and lead (Pb) were refined from the end of the 19th century until the 1970s, resulting in an extended area of about 700 km² diffusely polluted by particularly cadmium (Cd) and Zn (Ceenaeme et al., 2004). The risk for metal leaching and dispersion is intensified by the dominant sandy texture of the soils in the region. Sandy soils are characterized by a low cation exchange capacity (CEC), low acid neutralizing capacity and low metal ion sorption ability (Andersen et al., 2002).

Conventional soil remediation is technically and financially not feasible in this area because of the spatial extent of the metal pollution and the relatively moderate contamination levels. Therefore, adequate soil management that accounts for the present contamination appears to be the only realistic option. A possible management strategy for such degraded soils is afforestation (e.g. Dickinson, 2000; Pilon-Smits and Freeman, 2006; Pulford and Watson, 2003). In the Campine region, the most polluted agricultural soils have been taken out of production and a part of them has been afforested during recent years, in order to immobilize the metals and avoid their introduction in the food chain. This technique is also called phytostabilization.

Plants, and trees in particular, play an important role in the biogeochemical cycling of nutrients and pollutants, and can therefore be considered as ecosystem engineers (Jones et al., 1994). However, species specific effects of plants on nutrient cycling are not yet adequately known (e.g. Hobbie et al., 2007; Reich et al., 2005), including the effect of trees on the biogeochemical cycling of metals. The main driving biogeochemical processes affecting metal mobility in soils are oxidation-reduction reactions, acidification, organic matter dynamics, and changes in base cations concentrations and the CEC (Andersen et al., 2002; McBride et al., 1997; Römkens and Salomons, 1998; Sauvé et al., 2000). The changes in the above-mentioned soil characteristics after afforesting agricultural land strongly depend on the tree species (Alriksson and Eriksson, 1998; Augusto et al., 2002; Hagen-Thorn et al., 2004; Nordén, 1994; Reich et al., 2005). Consequently, the distribution and fluxes of metals in biomass, litter and mineral soil will be species specific as well (Alriksson and Eriksson, 2001; Mertens et al., 2007; Watmough et al., 2005). Selecting appropriate tree species is thus crucial for achieving successful phytostabilization. Trees can potentially be very well suited for phytostabilization purposes due to their extensive root systems and high transpiration capacity (Pulford and Watson, 2003). On the other hand, tree growth might enhance metal leaching because of soil acidification and production of dissolved organic matter (Mayer, 1998). Hence, with respect to risk control, it is very important to select tree species for phytostabilization purposes that cause low soil acidification and a minimal translocation of metals to their leaves (Mertens et al., 2007).

Therefore, the aim of this study was to investigate the effects of six different tree species on (i) soil characteristics that influence metal mobility: pH, organic carbon (OC) content and CEC and (ii) on Cd and Zn compartmentalization after 10 years of tree growth on a sandy soil that was formerly under agricultural practice.

2. Materials and Methods

2.1 Site description

The study site was the forest 'Waaltjesbos' (51°13'23" N, 5°15'01" E) in Lommel (north-east Belgium), which covers an area of 203 ha. The site is situated in between two zinc smelters. Approximately 2 km north of the site used to be the former zinc smelter of Lommel, which was built in 1904 and was closed down and demolished in 1974. The zinc smelter of Balen is about 2 km south-west, the predominant wind direction, of the site. It was built in 1889 and remains operational to this day. Metal emissions from this factory have been controlled significantly and currently satisfy requirements according to European standards. Nevertheless, the study site has been exposed to metal pollution during several decades, mainly to Cd and Zn and to a lesser degree to Pb, copper (Cu), arsenic (As) and mercury (Hg). The historical soil pollution in the region has two major causes: atmospheric deposition on the one hand and application of zinc ashes for road construction on the other hand. Although these zinc ashes have been recently removed as part of decontamination policy in the region, possible remainders in the area cannot be excluded.

The soil is a well-drained nutrient poor sandy soil (Humic Podzol; FAO/UNESCO classification). The forest is located at the edge of the Campine plateau, which originates from a mixture of tertiary sands and gravel-rich sands deposited by the Meuse River. During the Pleistocene these sands were covered by aeolian sand deposits.

Due to the past agricultural practices, that have lasted at least several decades, there is a clearly visible (black) plough layer extending till 40 cm depth. The site was converted to public forest and was planted in 1996-1998. Different tree species were planted in blocks, so that the forest consists of mainly homogeneous stands of pedunculate oak (*Quercus robur* L.), sessile oak (*Q. petraea* (Matt.) Liebl.), silver birch (*Betula pendula* Roth), Scots pine (*Pinus sylvestris* L.), Corsican pine (*Pinus nigra* ssp. *Laricio* Maire), black locust (*Robinia pseudoacacia* L.), Douglas fir (*Pseudotsuga menziesii* (Mirb.) Franco), Japanese larch (*Larix kaempferi* (Lambert) Carr.), black alder (*Alnus glutinosa* (L.) Gaertn.), common ash (*Fraxinus excelsior* L.) and beech (*Fagus sylvatica* L.). Aspen (*Populus tremula* L.) was planted in rows together with silver birch and black locust. No shrub layer is present in the forest.

No initial data were available on the soil characteristics and metal concentrations at the moment of the afforestation.

2.2 Sampling and sample analyses

Six tree species were selected that are typical for sandy soils in the study region and with potential to be used in future afforestations: oaks (*Q. robur* and *Q. petraea*), silver birch, black locust, aspen, Scots pine and Douglas fir. Those species are expected to affect the soil divergently in terms of acidification and metal uptake. Hence, the Cd and Zn concentrations in the different soil layers and tree compartments are expected to reflect many biogeochemical interactions that took place over the preceding 10 years.

Stem, branch and leaf litter biomass as well as different mineral soil layers were sampled in August 2007. At that time, the trees under study were 10 years old. For each of the considered tree species, three stands were selected throughout the forest to account for potential differences in soil characteristics, except for Douglas fir, for which only two stands were available. In each stand, two healthy trees with average diameter were selected for sampling and marked as sampling points. Hence, six replicates were obtained per species, except for Douglas fir (four replicates). The sampling trees were cut and soil was sampled within a radius of 1 m, by pooling two core samples. On each cut tree, a stem slice was taken at 1.3 m stem height, while branches were collected throughout the tree crown. Leaf litter was collected with litterfall traps that had a circular surface area of 0.24 m^2. In every stand, four litterfall traps were placed that were emptied monthly in the autumn of 2007 until the broadleaved trees were leafless, and during one year for the conifer species (September 2007 – August 2008). Stem, branch and leaf litter samples were dried at 70 °C in a forced air oven to constant weight. Samples were not rinsed before drying. Every dried leaf litter sample was sorted out by hand to separate the leaf litter of the considered tree species while discarding other litterfall fractions. Monthly leaf litter samples were pooled per trap before analysis. Subsequently the biomass samples were milled and analysed for Cd and Zn concentrations by inductively coupled plasma atomic emission spectroscopy (ICP-AES) after microwave destruction with an *aqua regia* solution (HNO_3/HCl).

Soil samples were taken with a soil auger at four fixed depths: 0-5, 10-20, 20-30 and 50-60 cm (below plough layer). The first three sampling depths were all entirely located in the Ap horizon, whereas the fourth sampling depth was located in the C horizon. Soil samples were dried at 40 °C in a forced air oven to constant weight. Aggregates were broken and soil was passed through a 2 mm sieve. Soil pH was determined in a 1:5 soil/KCl solution (1 M) with a glass electrode. The CEC was determined spectrophotometrically using a hexaminecobalt trichloride solution as extractant (ISO/DIS 23470). The OC content was determined spectrophotometrically by oxidation in a sulfochromic medium (ISO 14235). Pseudo-total Cd and Zn concentrations were determined by digesting the sediment for 2 h in an *aqua regia* solution under reflux (ISO 11466) and analysing the solution by ICP-AES. An estimation of the mobile Cd and Zn pool was made by determining their extractable concentrations in a 1:10 soil/$CaCl_2$ solution (0.01 M) and in a 1:10 soil/NH_4OAc-EDTA solution (0.5 M NH_4OAc and 0.02 M EDTA, set to pH 4.65) and subsequent ICP-AES analysis. The unbuffered $CaCl_2$ solution was used to extract exchangeable metals at ionic strength similar to that of the soil solution, whereas the NH_4OAc-EDTA solution was intended to extract a complexable metal fraction as well. Complexation by EDTA and acetic acid simulates the complexing behaviour by root exudates, whereas NH_4^+ ions are capable to desorb the exchangeable soil fraction and the adapted pH simulates rhizosphere acidity (Meers et al., 2007).

2.3 Estimation of aboveground biomass

Estimation of the amount of aboveground biomass is necessary for calculating aboveground metal stocks. Leaf litter biomass was estimated by weighing the leaf litter collected in the litterfall traps (precision of 0.001 g). By summing the monthly litterfall masses, mean yearly dry biomass per litterfall trap was determined and extrapolated to annual leaf litterfall per hectare.

Woody biomass of stems and branches was assessed using species specific allometric relations between tree diameter and dry stem or branch weight. The allometric relations were established for the studied forest stands according to the power function:

$$M = aD^b \qquad (1)$$

where M is the aboveground dry biomass per tree (kg), D is the stem diameter at 1.3 m height (cm), and a and b are allometric coefficients (Crow and Laidly, 1980; Zianis and Mencuccini, 2004).

To establish these relationships, *D* was measured for a random subsample of trees, spread over the three selected stands per species (n = 61 to 178), in order to define the diameter distribution per tree species (Table 1). Then, trees were divided into four classes on the basis of the diameter range. For each *D* class one sample tree, representative of the class median, was felled in March 2008. For each sample tree, the branches were separated from the stem and both compartments were separately weighed in the field by a portable scale with a precision of 10 g. Subsequently, a disc of approximately 10 cm thick was taken from the stem at 1.3 m height and weighed immediately on a portable scale to the nearest 0.1 g. A random subsample of the branches was weighed in the field as well (± 0.1 g). In the laboratory, all subsamples were dried at 90 °C in a forced air oven to constant mass, and weighed to recalculate the total dry biomass (*M*) of the stem and the branches of every sample tree. The established allometric relations (Table 1) were used to calculate the biomass of the stem and branches of every tree for which *D* was measured. Based on the stem numbers, total stem and branches biomass was estimated per hectare for each tree species.

The Cd and Zn amounts in the stem, branches and leaf litter were calculated by multiplying the Cd and Zn concentrations with the estimated biomass.

Table 1. Average diameter *D* (± st dev) of the measured trees (*n*), diameter range, and parameters of the allometric relations (see Eq. 1, *n* = 4)

Tree species	*n*	*D* (cm)	Range (cm)	Compartment	*a*	*b*	R^2
Silver birch	161	5.1 ± 2.2	1.6 - 12.8	Stem	0.0587	2.4881	0.996
				Branches	0.0225	2.4326	0.998
Oak	175	5.9 ± 1.9	2.0 – 11.6	Stem	0.1542	1.9986	0.982
				Branches	0.0376	2.4163	0.927
Black locust	170	9.3 ± 3.1	3.8 - 19.5	Stem	0.0849	2.4362	0.999
				Branches	0.0055	2.8291	0.994
Aspen	178	9.0 ± 3.2	3.2 – 17.7	Stem	0.12	2.1706	0.997
				Branches	0.0202	2.52	0.994
Scots pine	150	7.7 ± 1.8	2.3 – 13.1	Stem	0.1175	2.006	0.957
				Branches	0.0028	3.2158	0.991
Douglas fir	61	11.0 ± 2.5	4.4 – 15.9	Stem	0.1281	2.0278	0.966
				Branches	0.0017	3.3768	0.985

2.4 Data analysis

Differences between tree species were first tested using nonparametric analysis of variance (Kruskal-Wallis). Then, Mann-Whitney U tests were used to test the pairwise differences between the species. Moreover, soil data were subjected to an additional analysis to account for the spatial variation of the site. After all, both the historical atmospheric deposition as well as the possible remainders of the zinc ash roads may have contributed to a non-homogeneous metal pollution over the terrain before afforestation. The soil data indeed indicated an apparent spatial variation in soil characteristics and metal concentrations (e.g. total concentrations at 20-30 cm depth of 0.2 – 4.1 mg Cd kg^{-1} and 6.3 – 520 mg Zn kg^{-1}). On the other hand, the former agricultural use of the site has resulted in a strong (vertical) homogenization of the upper soil layers, due to many years of ploughing. Consequently we assumed a more or less uniform vertical distribution of the soil characteristics and metal concentrations in the soil profile until 40 cm depth at the moment of afforestation. After 10 years of tree growth, trees probably affected soil properties mostly in the upper soil layers. Therefore we considered the soil characteristics and metal concentrations in the 20-30 cm soil layer as being most representative for the initial site situation. Hence, to take into account the spatial variation, for each tree species the upper three soil layers (0-5, 10-20, 20-30 cm) were compared pairwise by means of a two-related-samples test (Wilcoxon signed-rank test). Correlations between pH, CEC, OC content, total and $CaCl_2$-extractable metal concentrations were calculated for each soil layer by means of Spearman correlation coefficients.

One of the Scots pine sampling points was marked as an outlier, with exceptionally high values for all measured soil variables (e.g. pH-KCl 5.8, total concentrations 4.3 mg Cd kg^{-1} and 490 mg Zn kg^{-1} in the 0-5 cm layer), and was excluded from all data analyses. All statistical analyses were performed in SPSS 15.0. Following Moran (2003), the p-values were not corrected for multiple comparisons. Therefore data analysis should be interpreted with care.

3. Results

3.1 Aboveground biomass: production and metal concentrations

Annual leaf litterfall was highest for Scots pine and aspen and lowest for silver birch and black locust (Table 2). Although no significant differences were found for the woody biomass estimates, black locust showed high stem wood production per ha. Silver birch produced low total aboveground biomass.

Table 2. Woody biomass estimates M_{stem} and $M_{branches}$ (ton DM/ha), annual leaf litterfall $M_{litterfall}$ (ton DM/ha/yr) and average wood production per year $Prod_{stem}$ and $Prod_{branches}$ (ton DM/ha/yr) for each tree species in 2008 (± st dev); values with the same letter did not differ between species (p < 0.05)

Tree species	M_{stem}	$M_{branches}$	$M_{litterfall}$	$Prod_{stem}$	$Prod_{branches}$
Silver birch	19.0 ± 9.8 [a]	6.6 ± 3.3 [a]	1.4 ± 0.6 [a]	1.9 ± 1.0	0.7 ± 0.3
Oak	26.2 ± 5.9 [a]	14.3 ± 3.9 [a]	3.7 ± 0.7 [b]	2.6 ± 0.6	1.4 ± 0.4
Black locust	92.3 ± 28.2 [a]	15.6 ± 5.5 [a]	2.1 ± 1.0 [ac]	9.2 ± 2.8	1.6 ± 0.6
Aspen	76.5 ± 27.7 [a]	29.7 ± 12.3 [a]	4.0 ± 1.0 [de]	7.7 ± 2.8	3.0 ± 1.2
Scots pine	41.7 ± 4.2 [a]	13.3 ± 2.2 [a]	4.9 ± 1.1 [e]	4.2 ± 0.4	1.3 ± 0.2
Douglas fir	56.6 ± 9.7 [a]	21.6 ± 4.1 [a]	3.1 ± 1.3 [bcd]	5.7 ± 1.0	2.2 ± 0.4

Significant differences between the six tree species were found for both Cd and Zn concentrations in the aboveground biomass compartments (Table 3). Aspen clearly took up more Cd than the other species, with the Cd concentration in the leaf litter being about 7 times higher than that of the other tree species. Zn concentrations in all the compartments were significantly higher in silver birch and aspen. Among the other species, only minor differences in Cd and Zn concentrations were found.

Table 3. Average Cd and Zn concentrations (± st dev) in the biomass compartments (mg/kg DM); per compartment, values with the same letter within a column did not differ between species (p < 0.05)

	Cd	Zn
	Stem	
Silver birch	0.83 ± 0.38 (3) [a]	239 ± 75 [c]
Oak	< 1.0 [a]	53 ± 31 [a]
Black locust	< 1.0 [a]	56 ± 15 [a]
Aspen	2.72 ± 1.50 (1) [b]	146 ± 60 [b]
Scots pine	1.03 ± 0.49 (2) [a]	63 ± 34 [a]
Douglas fir	< 1.0 [a]	41 ± 15 [a]
	Branches	
Silver birch	1.88 ± 0.81 [b]	701 ± 161 [b]
Oak	< 1.0 [a]	171 ± 53 [a]

Black locust	< 1.0 [a]	180 ± 30 [a]
Aspen	7.02 ± 2.66 [c]	497 ± 181 [b]
Scots pine	1.70 ± 0.22 [b]	177 ± 59 [a]
Douglas fir	0.87 ± 0.43 (2) [ab]	144 ± 23 [a]
	Leaf litter	
Silver birch	1.72 ± 0.44 [b]	1 540 ± 341 [c]
Oak	< 1.0 [a]	287 ± 106 [a]
Black locust	1.22 ± 0.40 (1) [bc]	427 ± 100 [b]
Aspen	13.0 ± 6.5 [d]	2 354 ± 494 [d]
Scots pine	1.08 ± 0.34 (1) [c]	321 ± 56 [a]
Douglas fir	< 1.0 [a]	295 ± 22 [a]

<: If all values were lower than the determination limit, the determination limit was given, preceded by ‘‘<’’
(1), (2), (3): The number between brackets is the number of values lower than the determination limit of 1.0 for Cd. The mean was calculated by equalling those values to the half of the determination limit.

The amount of Cd and Zn in the aboveground biomass of aspen was clearly higher than for the other tree species (Table 4). Silver birch, on the other hand, showed high Zn concentrations in its biomass (Table 3), but the aboveground Zn pools did not remarkably differ from the other species due to its low biomass production (Table 2).

Table 4. Average Cd and Zn amounts (± st dev) in woody biomass and annual leaf litterfall of the six tree species (g/ha)

	Cd	**Zn**
	Stem	
Silver birch	16.2 ± 7.6	4 662 ± 1 533
Oak	< 26.7	1 422 ± 852
Black locust	< 91.2	5 124 ± 1 427
Aspen	208.2 ± 115.6	11 188 ± 4 666
Scots pine	44.3 ± 23.2	2 723 ± 1 592
Douglas fir	< 57.4	2 338 ± 947
	Branches	
Silver birch	12.6 ± 5.5	4 688 ± 1 117
Oak	< 15.8	2 701 ± 1 395
Black locust	< 15.5	2 775 ± 570
Aspen	209.7 ± 82.7	14 845 ± 5 649
Scots pine	23.2 ± 4.8	2 409 ± 896
Douglas fir	18.7 ± 9.9	3 086 ± 769

	Leaf litter	
Silver birch	2.4 ± 1.3	2 159 ± 1 093
Oak	< 3.7	1 063 ± 438
Black locust	2.5 ± 1.5	893 ± 492
Aspen	52.4 ± 29.5	9 461 ± 3 146
Scots pine	5.3 ± 2.0	1 562 ± 452
Douglas fir	< 3.1	910 ± 402

3.2 Soil

3.2.1. pH, OC and CEC

Only under aspen the pH in the topsoil (0-5 cm) was significantly increased compared to the deeper soil layer (Table 5). The other species induced a slight but not significant decrease in topsoil pH. Consequently, aspen showed a higher pH than the other species in the topsoil. At 10-20 cm depth the pH under silver birch was lower than under aspen, Scots pine and Douglas fir. With respect to the OC content in the topsoil, two groups could be distinguished, namely aspen and black locust in the higher range and the other species with lower OC contents. Aspen showed a significantly higher OC content in the topsoil compared to deeper layers. The CEC in the upper 5 cm of soil under aspen was higher than for the other species, whereas the lowest CEC values were found under silver birch and oak. Aspen had induced a CEC increase in the topsoil compared to 10-20 cm depth, whereas the CEC under silver birch at 10-20 cm depth was lower than at 20-30 cm depth. In each soil layer the CEC was significantly ($p < 0.05$) correlated with both pH (Spearman's $\rho = 0.381 - 0.692$) and OC content ($\rho = 0.341 - 0.687$), while pH and OC content were not significantly correlated ($p > 0.06$, $\rho < 0.374$). Pairwise comparison revealed that pH, OC and CEC in the deepest soil layer (50-60 cm) were significantly lower than at 20-30 cm depth (significances not shown).

Table 5. Soil characteristics pH-KCl, OC and CEC at four different depths (average ± st dev); values followed with the same letter did not differ, first capital letters denote species effects and should be read vertically ($p < 0.05$), second small letters denote differences between soil layers and should be read horizontally ($p < 0.05$)

	0 – 5 cm	**10 – 20 cm**	**20 – 30 cm**	**50 – 60 cm**
		pH-KCl (-)		
Silver birch	4.5 ± 0.3 $^{A\ a}$	4.3 ± 0.2 $^{A\ a}$	4.5 ± 0.1 $^{A\ a}$	4.4 ± 0.2 ^{A}
Oak	4.4 ± 0.2 $^{A\ a}$	4.5 ± 0.3 $^{AB\ a}$	4.6 ± 0.3 $^{A\ a}$	4.7 ± 0.3 ^{A}
Black locust	4.4 ± 0.3 $^{A\ a}$	4.5 ± 0.3 $^{AB\ a}$	4.7 ± 0.3 $^{A\ a}$	4.5 ± 0.1 ^{A}
Aspen	5.4 ± 0.6 $^{B\ a}$	4.7 ± 0.3 $^{B\ b}$	4.8 ± 0.5 $^{A\ ab}$	4.8 ± 0.4 ^{A}

Scots pine	4.5 ± 0.3 $^{A\ a}$	4.7 ± 0.3 $^{B\ a}$	4.8 ± 0.4 $^{A\ a}$	4.5 ± 0.4 ^{A}
Douglas fir	4.4 ± 0.4 $^{AB\ a}$	4.7 ± 0.1 $^{B\ a}$	4.7 ± 0.2 $^{A\ a}$	4.4 ± 0.2 ^{A}
OC (g C/kg DM)				
Silver birch	19 ± 5 $^{A\ a}$	14 ± 6 $^{A\ a}$	14 ± 6 $^{A\ a}$	7 ± 4 ^{A}
Oak	17 ± 1 $^{AB\ a}$	18 ± 5 $^{A\ a}$	16 ± 4 $^{A\ a}$	11 ± 8 ^{A}
Black locust	32 ± 9 $^{BC\ a}$	15 ± 8 $^{A\ a}$	18 ± 8 $^{A\ a}$	6 ± 3 ^{A}
Aspen	29 ± 6 $^{C\ a}$	18 ± 9 $^{A\ b}$	17 ± 7 $^{A\ b}$	13 ± 7 ^{A}
Scots pine	20 ± 7 $^{AB\ a}$	16 ± 7 $^{A\ a}$	14 ± 8 $^{A\ a}$	5 ± 4 ^{A}
Douglas fir	17 ± 4 $^{AB\ a}$	19 ± 7 $^{A\ a}$	14 ± 1 $^{A\ a}$	12 ± 12 ^{A}
CEC (cmol/kg DM)				
Silver birch	2.8 ± 1.2 $^{A\ ab}$	2.2 ± 0.8 $^{A\ a}$	2.9 ± 1.1 $^{A\ b}$	1.7 ± 0.4 ^{A}
Oak	2.8 ± 0.6 $^{AC\ a}$	2.7 ± 0.7 $^{AC\ a}$	3.1 ± 0.7 $^{AC\ a}$	2.8 ± 1.7 ^{AB}
Black locust	5.8 ± 1.0 $^{BCD\ a}$	4.5 ± 0.9 $^{B\ a}$	4.6 ± 0.7 $^{B\ a}$	3.4 ± 0.6 ^{B}
Aspen	8.1 ± 1.7 $^{D\ a}$	4.8 ± 1.8 $^{BC\ b}$	4.9 ± 1.5 $^{BC\ ab}$	4.0 ± 1.8 ^{B}
Scots pine	4.7 ± 0.7 $^{B\ a}$	4.6 ± 0.8 $^{B\ a}$	4.2 ± 1.1 $^{AB\ a}$	2.8 ± 0.7 ^{B}
Douglas fir	4.6 ± 1.0 $^{AB\ a}$	3.9 ± 0.2 $^{B\ a}$	3.7 ± 0.4 $^{AB\ a}$	2.8 ± 1.5 ^{AB}

3.2.2. Cd and Zn concentrations in the soil

Total Cd and Zn concentrations in the topsoil were highest under aspen and differed significantly from the lower Cd and Zn levels under the other species, except for Scots pine (Table 6). Aspen induced a declining depth profile for the total metal concentrations, the topsoil values being significantly higher than those in deeper layers. The tree species effect was most pronounced in the upper 5 cm, as no differences between species were found in the deeper soil layers.

In contrast to total soil Cd and Zn, the $CaCl_2$-extractable concentrations were not highest in the topsoil of aspen but of Scots pine, although no significant differences between the species or the soil layers were found. NH_4OAc-EDTA-extractable concentrations, however, followed the same trends as total concentrations, with significantly higher values in the topsoil under aspen compared to other species and to deeper soil layers. In addition, NH_4OAc-EDTA-extractable Cd under oak was significantly higher at 10-20 cm than in the topsoil.

Pairwise comparison revealed that both total and extractable Cd and Zn concentrations in the deepest soil layer (50-60 cm) were significantly lower than at 20-30 cm depth (significances not shown). This could be explained because the soil contamination was mainly caused by atmospheric pollution. Ploughing till approximately 40 cm depth redistributed the metals in the upper three sampling layers, but not deeper.

Table 6. Total and extractable soil Cd and Zn concentrations (mg/kg DM) at four depths (average ± st dev); values followed with the same letter did not differ, first capital letters denote species effects and should be read vertically ($p < 0.05$), second small letters denote differences between soil layers and should be read horizontally ($p < 0.05$)

	0 – 5 cm	10 – 20 cm	20 – 30 cm	50 – 60 cm
		Cd		
Silver birch	1.42 ± 0.86 (1) $^{AB\ a}$	1.50 ± 0.72 (1) $^{A\ a}$	1.67 ± 1.02 (1) $^{A\ a}$	0.42 ± 0.40 (4) ^{A}
Oak	1.45 ± 0.42 $^{A\ a}$	1.96 ± 0.92 $^{A\ a}$	1.90 ± 0.76 $^{A\ a}$	0.83 ± 0.98 (4) ^{A}
Black locust	1.70 ± 0.70 $^{AB\ a}$	2.15 ± 1.12 (1) $^{A\ a}$	1.82 ± 1.22 (1) $^{A\ a}$	0.43 ± 0.38 (4) ^{A}
Aspen	3.55 ± 1.44 $^{C\ a}$	2.20 ± 1.30 (1) $^{A\ b}$	1.96 ± 1.14 (1) $^{A\ b}$	1.15 ± 0.97 (2) ^{A}
Scots pine	2.34 ± 0.67 $^{BC\ a}$	1.88 ± 0.59 $^{A\ a}$	1.75 ± 0.93 $^{A\ a}$	0.33 ± 0.20 (4) ^{A}
Douglas fir	1.55 ± 0.29 $^{AB\ a}$	1.88 ± 0.72 $^{A\ a}$	1.88 ± 0.34 $^{A\ a}$	0.33 ± 0.26 (3) ^{A}
		Zn		
Silver birch	87 ± 52 $^{AB\ a}$	73 ± 44 $^{A\ a}$	77 ± 40 $^{A\ a}$	30 ± 19 ^{A}
Oak	86 ± 29 $^{A\ a}$	109 ± 68 $^{A\ a}$	110 ± 56 $^{A\ a}$	51 ± 47 ^{A}
Black locust	123 ± 62 $^{AB\ a}$	135 ± 70 $^{A\ a}$	120 ± 72 $^{A\ a}$	29 ± 23 (1) ^{A}
Aspen	267 ± 91 $^{C\ a}$	120 ± 72 $^{A\ b}$	114 ± 72 $^{A\ b}$	71 ± 69 (1) ^{A}
Scots pine	142 ± 38 $^{BC\ a}$	118 ± 23 $^{A\ a}$	101 ± 27 $^{A\ a}$	21 ± 13 ^{A}
Douglas fir	117 ± 69 $^{AB\ a}$	93 ± 34 $^{A\ a}$	98 ± 37 $^{A\ a}$	25 ± 10 ^{A}
		Cd ($CaCl_2$)		
Silver birch	0.56 ± 0.21 $^{A\ a}$	0.68 ± 0.31 $^{A\ a}$	0.60 ± 0.33 $^{A\ a}$	0.26 ± 0.20 ^{A}
Oak	0.67 ± 0.22 $^{A\ a}$	0.81 ± 0.24 $^{A\ a}$	0.65 ± 0.26 $^{A\ a}$	0.13 ± 0.13 ^{A}
Black locust	0.58 ± 0.18 $^{A\ a}$	0.64 ± 0.40 $^{A\ a}$	0.52 ± 0.31 $^{A\ a}$	0.16 ± 0.10 ^{A}
Aspen	0.45 ± 0.15 $^{A\ a}$	0.52 ± 0.21 $^{A\ a}$	0.49 ± 0.22 $^{A\ a}$	0.31 ± 0.20 ^{A}
Scots pine	0.87 ± 0.50 $^{A\ a}$	0.72 ± 0.38 $^{A\ a}$	0.56 ± 0.43 $^{A\ a}$	0.16 ± 0.13 ^{A}
Douglas fir	0.64 ± 0.15 $^{A\ a}$	0.57 ± 0.14 $^{A\ a}$	0.53 ± 0.16 $^{A\ a}$	0.14 ± 0.11 ^{A}
		Zn ($CaCl_2$)		
Silver birch	32 ± 11 $^{A\ a}$	28 ± 14 $^{A\ a}$	27 ± 13 $^{A\ a}$	11 ± 8 ^{A}
Oak	38 ± 11 $^{A\ ab}$	41 ± 19 $^{A\ a}$	35 ± 19 $^{A\ b}$	8 ± 8 ^{A}
Black locust	30 ± 9 $^{A\ a}$	34 ± 20 $^{A\ a}$	30 ± 19 $^{A\ a}$	6 ± 5 ^{A}
Aspen	33 ± 17 $^{A\ a}$	26 ± 13 $^{A\ a}$	22 ± 11 $^{A\ a}$	14 ± 11 ^{A}
Scots pine	43 ± 19 $^{A\ a}$	43 ± 34 $^{A\ a}$	28 ± 17 $^{A\ a}$	8 ± 7 ^{A}
Douglas fir	36 ± 15 $^{A\ a}$	26 ± 5 $^{A\ a}$	26 ± 9 $^{A\ a}$	4 ± 2 ^{A}
		Cd (NH_4OAc-EDTA)		
Silver birch	1.34 ± 0.77 $^{A\ a}$	1.29 ± 0.76 $^{A\ a}$	1.67 ± 1.12 $^{A\ a}$	0.39 ± 0.37 ^{A}
Oak	1.22 ± 0.47 $^{A\ a}$	1.76 ± 0.88 $^{A\ b}$	1.67 ± 0.95 $^{A\ ab}$	0.39 ± 0.71 ^{A}
Black locust	1.30 ± 0.64 $^{A\ a}$	1.72 ± 0.96 $^{A\ a}$	1.41 ± 0.87 $^{A\ a}$	0.27 ± 0.26 ^{A}
Aspen	3.26 ± 1.18 $^{B\ a}$	1.91 ± 1.10 $^{A\ b}$	1.78 ± 0.96 $^{A\ b}$	1.08 ± 0.93 ^{A}
Scots pine	1.96 ± 0.83 $^{AB\ a}$	2.09 ± 0.36 $^{A\ a}$	1.80 ± 1.11 $^{A\ a}$	0.29 ± 0.29 ^{A}
Douglas fir	1.84 ± 0.21 $^{AB\ a}$	2.10 ± 0.97 $^{A\ a}$	2.31 ± 0.96 $^{A\ a}$	0.25 ± 0.24 ^{A}
		Zn (NH_4OAc-EDTA)		
Silver birch	61 ± 35 $^{A\ a}$	43 ± 28 $^{A\ a}$	55 ± 32 $^{A\ a}$	15 ± 11 ^{A}

Oak	58 ± 24 $^{\mathrm{A\ a}}$	71 ± 48 $^{\mathrm{AB\ a}}$	71 ± 51 $^{\mathrm{A\ a}}$	20 ± 29 $^{\mathrm{A}}$
Black locust	59 ± 31 $^{\mathrm{A\ a}}$	72 ± 35 $^{\mathrm{AB\ a}}$	63 ± 32 $^{\mathrm{A\ a}}$	12 ± 10 $^{\mathrm{A}}$
Aspen	200 ± 49 $^{\mathrm{B\ a}}$	74 ± 47 $^{\mathrm{AB\ b}}$	70 ± 47 $^{\mathrm{A\ b}}$	48 ± 46 $^{\mathrm{A}}$
Scots pine	83 ± 28 $^{\mathrm{A\ a}}$	94 ± 47 $^{\mathrm{B\ a}}$	70 ± 23 $^{\mathrm{A\ a}}$	15 ± 16 $^{\mathrm{A}}$
Douglas fir	77 ± 21 $^{\mathrm{A\ a}}$	71 ± 27 $^{\mathrm{AB\ a}}$	82 ± 33 $^{\mathrm{A\ a}}$	8 ± 3 $^{\mathrm{A}}$

(1), (2), (3), (4): The number between brackets is the number of values lower than the determination limit of 0.40 for Cd or 5.0 for Zn. The mean was calculated by equalling those values to the half of the determination limit.

3.2.3. Correlation between soil characteristics and metal concentrations

Significant ($p < 0.01$) positive correlations were found between pH, CEC, OC and the total Cd and Zn concentrations in the topsoil (Figure 1). The three considered soil characteristics appeared to affect total metal concentrations significantly. The correlation coefficients with the $CaCl_2$-extractable metal concentrations were all negative but much weaker than those with the total concentrations. Only the influence of pH on extractable Cd was significant.

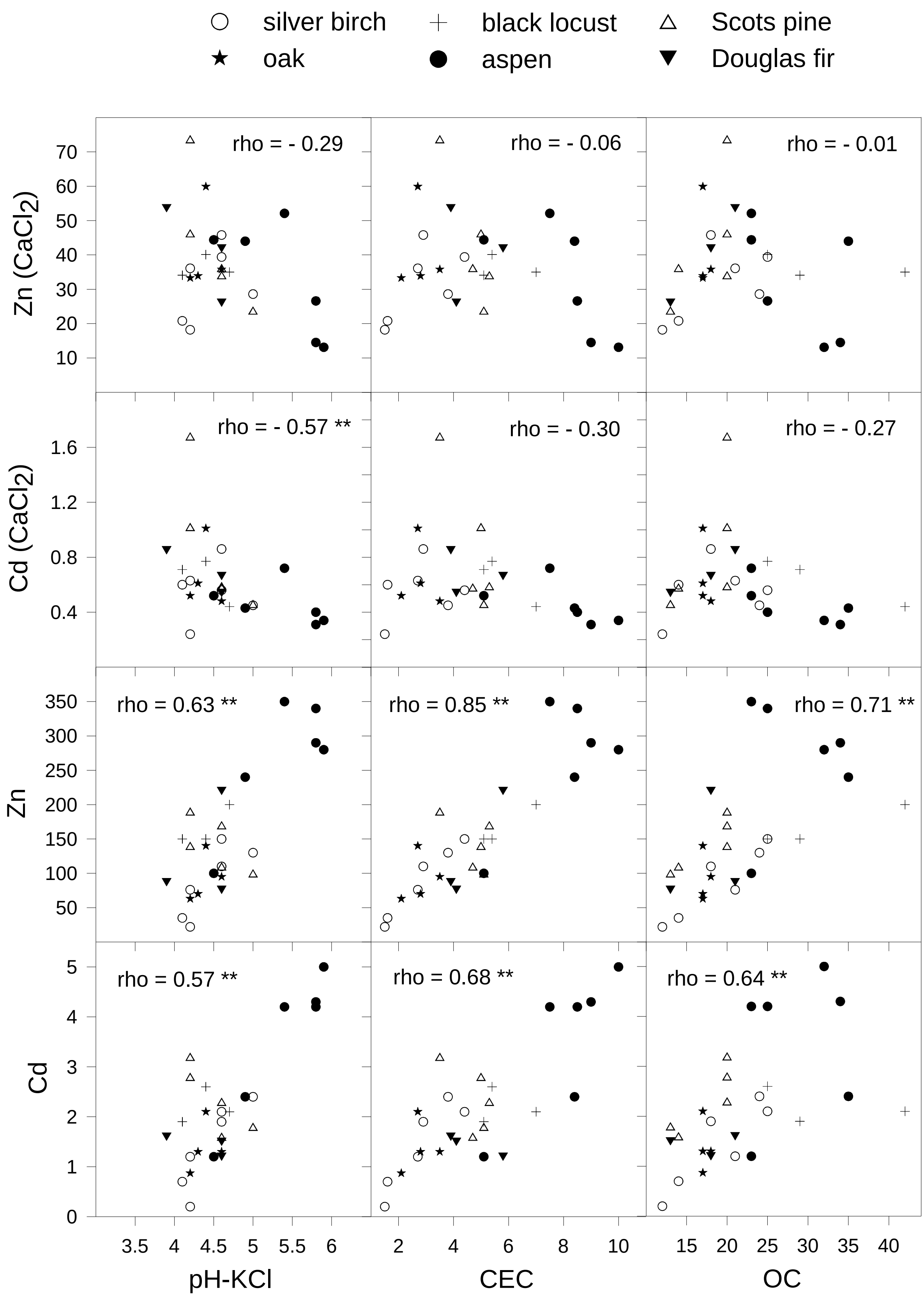

Figure 1. Correlations between the soil characteristics pH, CEC, OC and the total and $CaCl_2$-extractable Cd and Zn concentrations in the topsoil (0-5 cm) under the six tree species, with Spearman's rho correlation coefficients (p < 0.01)**

4. Discussion

The Cd and Zn concentrations in the different soil layers and tree compartments are the net result of a variety of biogeochemical and biophysical processes that took place during 10 years after afforestation. The most apparent tree species effects on the redistribution of Cd and Zn and the consequent implications for phytostabilization will be discussed.

4.1 Tree species effects on aboveground metal accumulation

Poplars (*Populus* spp.) and *Salix* spp. are known to take up Cd and Zn from polluted soils and to accumulate these metals in their biomass, including branches, leaves and wood, distinctly more than other tree species (Brekken and Steinnes, 2004; Hassinen et al., 2009; Laureysens et al., 2004; Mertens et al., 2007; Unterbrunner et al., 2007; Vandecasteele et al., 2003). The Cd and Zn concentrations measured in the biomass of aspen (*Populus tremula*) are in line with these reports. Our findings also confirm that birches (*Betula* spp.) do not take up Cd but are known as Zn accumulators (Alriksson and Eriksson, 2001; Rosselli et al., 2003). Furthermore, a clear compartmentalization between the three studied biomass fractions was observed, in particular for Zn. Tree species that take up Cd or Zn from the rooting zone obviously translocate those trace metals to their leaves, as reported before (Alriksson and Eriksson, 2001; Hammer et al., 2003; Meers et al., 2005; Mertens et al., 2007; Unterbrunner et al., 2007; Vervaeke et al., 2003).

Aboveground metal amounts (Table 4) represent the actual metal stock in the aboveground biomass and can be used to estimate metal fluxes by litterfall and wood immobilization. Considering the risk of metal dispersion within ecosystems, litterfall should be given principal attention as it brings the total foliar Cd and Zn stock to the upper soil layer every autumn, or continuously in case of the conifer species. The high Cd and Zn stocks in the leaf litter of aspen represent large Cd and Zn fluxes every autumn and generate consequently a pathway of aboveground metal dispersion. On the other hand, the large Cd and Zn stocks in aspen's woody biomass will have smaller implications with regard to ecotoxicological risks, since the woody compartment more slowly enters the decomposition cycle.

4.2 Tree species effects on soil pH, OC and CEC

It is generally known that plant species can differ in their effect on soil pH, organic matter content and CEC, and differences between tree species for these soil characteristics have been reported in the past (Augusto et al., 2002; Finzi et al., 1998; Hagen-Thorn et al., 2004; Mertens et al., 2007; Nordén, 1994; Reich et al., 2005). In the present study, all tree species except aspen, tended to decrease the pH slightly, but not (yet) significantly, in the topsoil compared to 20-30 cm (Table 5). A moderate acidification of the topsoil was expected since trees are generally known to lower the soil pH due to soil respiration, tree growth and decomposition of organic material (Nilsson et al., 1982; Reich et al., 2005). The fact that aspen increased the topsoil pH was thus remarkable, particularly given the short period of tree growth (10 years) and the nutrient poor sandy soil conditions. Sandy soils are characterized by a low CEC and a low acid neutralizing capacity, which makes them more vulnerable to acidification. The differences in topsoil pH between the species can probably be attributed to the chemical properties and the decomposition rate of the different leaf litter types (Hagen-Thorn et al., 2004; Mertens et al., 2007; Reich et al., 2005). Retarded litter decomposition leads to the production of organic acids and delays the return of base cations to the soil, which will result in lower pH values (Hagen-Thorn et al., 2004). Leaf litter from poplars is generally of high nutritional quality, facilitating rapid litter decomposition and nutrient release to soils (Cooke and Weih, 2005). In a common garden test with 14 tree species Reich et al. (2005) found that litter Ca concentration appeared to be a key driver of changes in soil properties and that soil pH increased with the Ca concentration of foliage litter. Our results showed that aspen indeed had the highest Ca concentrations in its leaf litter (unreported data). The ameliorating effect of the nutrient rich aspen litter on pH did not occur yet in the deeper soil layers. Black locust is a nitrogen (N)-fixing legume tree (Boring and Swank, 1984) and litter decomposition rates of N-fixing species are reported to be higher than those of non-N-fixing species (Aerts and Chapin, 2000). However, no increase of the topsoil pH under black locust was found, most likely because symbiotic N_2-fixation can lead to soil acidification as a by-product of increased nitrification rates (Johnson and Lindberg, 1992; Rhoades et al., 2001; Van Miegroet and Cole, 1985).

As CEC is influenced by the clay and organic matter content and the soil pH (Helling et al., 1964), differences in CEC at this sandy site will mainly be determined by variations in organic matter content. The CEC was indeed strongly positively correlated with OC in each soil layer. Due to input of organic material from litterfall, trees increase the amount of organic matter in the upper soil layers which will eventually lead to the formation of an ectorganic horizon (Emmer, 1995). No ectorganic horizon was discernible yet under this young forest. Nevertheless, the OC contents in the topsoil under aspen and black locust, i.e. those species with high litter decomposition rates, could be distinguished from the other species. This was also reflected in a higher CEC under aspen.

The values of the three examined soil characteristics in the deepest layer (50-60 cm) were significantly lower than at 20-30 cm depth. This could be attributed to the former agricultural use of the site, which aimed at maintaining a high pH and organic matter content in the plough layer by liming and fertilizing.

Effects of the different tree species on the soil characteristics after only 10 years of tree growth on a poor sandy soil were remarkable and important, considering the strong influence of pH, CEC and OC on metal mobility, as shown in Figure 1. Both the positive and negative relations between the soil characteristics and metal concentrations were expected and confirmed previous findings (Andersen et al., 2002; Römkens and Salomons, 1998; Sauvé et al., 2000; Watmough et al., 2005).

4.3 Tree species effects on metal redistribution in the soil

Input and output fluxes determine metal concentrations and metal amounts in the different soil layers. Output of metals from the soil layers is mainly controlled by uptake by plant roots and leaching. Input to the upper soil layer is determined by input of contaminated litter and atmospheric deposition. Input via atmospheric deposition was not accounted for in this study, since the metal emissions from the factory nearby decreased significantly before the site was afforested.

The most distinct species effect on metal redistribution was found for aspen, showing higher total Cd and Zn concentrations in the topsoil compared to the other species. Total Cd and Zn concentrations in the upper 5 cm were on average respectively 1.6 and 2 times higher than those at 20-30 cm depth and respectively 2 and 2.5 times higher than under the other species. Such a metal redistribution can be attributed to an accumulation in the topsoil or to leaching of metals from the deeper layers. Whether accumulation of metals via the litterfall causes an increase of the soil concentration depends on two conditions: (1) the concentration in the decomposed litter is higher than the soil concentration and (2) the input flux is greater than the output flux. Decomposition of litterfall slows down and stops at a ‘limit value’ (fraction of litter that does not decompose) of 0-55 % of the dry weight (Berg, 2000). As metals do not decompose, metal concentrations in the litter will at least double during decomposition from fresh leaves to decomposed litter. The Cd and Zn concentrations of the decomposed litter of the aspens thus are expected to increase to about 26 mg Cd kg^{-1} and 4700 mg Zn kg^{-1} dry weight (Table 3). They will therefore be significantly higher than the soil concentrations, and allow an increase of the topsoil concentrations to occur. However, this only holds true if output fluxes of metals do not exceed the input flux. In addition, the relatively high values for pH, OC and CEC found in the topsoil under aspen contribute to the adsorption of the metals on the soil’s exchange sites (Sauvé et al., 2000). This was reflected in the fact that (1) the $CaCl_2$-extractable Cd and Zn concentrations in the topsoil were not increased under aspen and (2) neither the total Cd and Zn concentrations in the 10-20 cm layer were increased compared to 20-30 cm depth (Table 6). Thus, the input flux of metals via the litterfall exceeded the output flux, resulting in a Cd and Zn accumulation in the topsoil under aspen. A similar accumulation of Cd and Zn in the topsoil of *Populus*, compared to other species, was reported on a polluted dredged sediment disposal site with 33-year-old trees (Mertens et al., 2007). Nevertheless, our results were striking as the trees were only 10 years old and sandy soils show faster dynamics in metal mobilization than calcareous dredged sediments.

In contrast to aspen, the high Zn concentration in the leaf litter of silver birch (Table 3) was not reflected in a considerable Zn accumulation in the topsoil compared to the 20-30 cm layer (Table 6). This can probably be attributed to the low leaf litter biomass production of silver birch (Table 2). The other tree species did not show any marked Cd or Zn accumulation in their aboveground biomass and consequently no metal accumulation in the topsoil could be observed. On the contrary, for some species we observed a slight decrease in the total topsoil concentrations compared to the deeper layers, although this trend was not significant. Leaching of the metals to deeper soil layers, due to soil acidification, might be one of the reasons for that decrease.

The total Cd and Zn concentrations were compared with the background concentrations and with the Flemish soil sanitation reference values (Vlarebo). These values are assessed as a function of the organic matter (2.71 %) and clay contents (3.33 %) of the soil (Table 7). In more than half of the sampling points, total Cd concentrations in the upper three soil layers exceeded the soil sanitation reference value, whereas for Zn this limit was not exceeded at the study site.

Table 7. Background concentration and soil sanitation norm (mg/kg) for Cd and Zn in Waaltjesbos, taking into account the percentage OM and clay

	Cd	**Zn**
background concentration	0.64	56
soil sanitation reference value (type I)	1.59	544

Unlike total soil metal concentrations, extractable metal concentrations allow an estimation of the mobile metal pools in the soil, thus reflecting a metal fraction that could potentially be taken up by adjacent plant roots, be detrimental to various soil biological organisms, or otherwise be leached from the soil and contaminate groundwater and surface water (Sauvé et al., 2000). Extractable metal concentrations thus offer a better criterion when it comes to evaluation of possible dispersion risks of the metals in the environment or in the food chain. Our results of the

$CaCl_2$-extractable Cd and Zn concentrations in the soil profile showed no significant tree species effects (Table 6). It was, moreover, particularly remarkable that aspen did not generate elevated $CaCl_2$-extractable concentrations in the topsoil. This could be explained by the higher pH and CEC under aspen (Table 5, Figure 1). This implies that the risks concerning aspen might actually be smaller than expected from the accumulation of total Cd and Zn in the topsoil. However, NH_4OAc-EDTA-extractable Cd and Zn concentrations in the topsoil under aspen were elevated compared to other species and to deeper soil layers. Being a very strong extractant, NH_4OAc-EDTA yielded almost ‘totals’ on this poorly buffered sandy soil. The implications of these findings with respect to risk control in phytostabilization projects are discussed below.

4.4 Implications for phytostabilization

Phytostabilization uses plants to minimize the mobility and bioavailability of pollutants in the environment, either by immobilizing them or by preventing their migration (Smith and Bradshaw, 1972; Vangronsveld et al., 1995). Consequently, tree species that accumulate metals in their leaves and that induce soil acidification should be avoided, as these processes might give rise to dispersion of metals in the environment via aboveground and belowground pathways.

Van Nevel et al. (2007) identified three main risks associated with the accumulation of metals in aboveground plant parts: (i) metals entering the food chain through herbivores, (ii) dispersion of contaminated plant material to adjacent environments and (iii) accumulation of metals in the topsoil. The topsoil is particularly vulnerable as it is the biologically most active part of the soil system and biological activity has been shown to be highly sensitive to metal pollution (Bergkvist et al., 1989). In this respect, aspen and *Populus* species in general should be avoided for afforesting Cd and Zn contaminated lands because they translocate high amounts of Cd and Zn into the foliage. Our results clearly demonstrate an accumulation of total Cd and Zn concentrations in the topsoil under aspen trees and this only after 10 years of tree growth. This accumulation pattern is expected to be continued in the future. Such a metal redistribution in the ecosystem is undesirable in phytostabilization projects. However, ‘bioavailable’ $CaCl_2$-extractable Cd and Zn concentrations appeared not to be elevated under aspen, due to the higher pH and CEC values in the topsoil of this species. As extractable ‘bioavailable’ concentrations should preferably be considered for risk analysis, one may argue that on the short term there are few concerns regarding the ecotoxicological risks linked with aspen. However, the NH_4OAc-EDTA-extractable Cd and Zn concentrations in the topsoil under aspen were increased compared to the deeper soil layers, as was also reported by Mertens et al. (2007) for *Populus* ‘Robusta’.

This indicates that in more acid conditions the accumulated Cd and Zn in the topsoil will yet become more mobile and thus ‘bioavailable’. This could for instance be the case when the aspen trees would be harvested and replaced by more acidifying species. Moreover, it should be stressed that all metals accumulated in the leaves might still pose a long-term risk to primary consumers and enter the food chain.

Although silver birch translocated considerable amounts of Zn to its leaves, its low leaf litter biomass production mitigated the risk of Zn dispersion in the ecosystem, as confirmed by the absence of Zn accumulation in the topsoil under birch. Aboveground dispersion of Zn from silver birches might, however, still occur via herbivory. Furthermore, as leaf litter biomass production will increase in time, Zn accumulation in the topsoil can potentially occur in the future.

Soil acidification might give rise to metal leaching to deeper soil layers and eventually to groundwater contamination. According to the present study, all tree species other than aspen or silver birch induced a slight decrease in pH in the topsoil. Continued tree growth in the future will most likely result in a more progressive acidification of the upper soil layers. Given the risk of Cd and Zn leaching, we recommend not to plant acidifying tree species, or else to mix them with other species that have high nutritional leaf litter quality, facilitating rapid litter decomposition and nutrient release to soils, e.g. European rowan (*Sorbus aucuparia*) or alder buckthorn (*Rhamnus frangula*).

The effects of the different tree species on the soil characteristics and on the metal redistribution in the soil profile will evolve during the next decades and will probably become more pronounced in the future. A next sampling campaign within 10 or 20 years is therefore essential.

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