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Metal uptake by young trees from dredged brackish sediment: limitations and possibilities for phytoextraction and phytostabilisation

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Abstract

Five tree species (*Acer pseudoplatanus* L., *Alnus glutinosa* L. Gaertn., *Fraxinus excelsior* L., *Populus alba* L. and *Robinia pseudoacacia* L.) were planted on a mound constructed of dredged sediment. The sediment originated from a brackish river mouth and was slightly polluted with heavy metals. This preliminary study evaluated the use of trees for site reclamation by means of phytoextraction of metals or phytostabilisation.

Although the brackish nature of the sediment caused slight salt damage, overall survival of the planted trees was satisfactory. Robinia and white poplar had the highest growth rates. Ash, maple and alder had the highest survival rates (>90%) but showed stunted growth. Ash, alder, maple and Robinia contained normal concentrations of Cd, Cu, Pb and Zn in their foliage. As a consequence these species reduce the risk of metal dispersal and are therefore suitable species for phytostabilisation under the given conditions. White poplar accumulated high concentrations of Cd (8.0 mg kg^{-1}) and Zn (465 mg kg^{-1}) in its leaves and might therefore cause a risk of Cd and Zn input into the ecosystem because of autumn litter fall. This species is thus unsuitable for phytostabilisation. Despite elevated metal concentrations in the leaves, phytoextraction of heavy metals from the soil by harvesting stem and/or leaf biomass of white poplar won’t be a realistic option because it will require an excessive amount of time to be effective.

Key words: heavy metals, phytoremediation, phytostabilisation, metal uptake, tree species

Introduction

Rivers and harbour docks are dredged on a regular basis to provide shipping traffic efficiency. In Flanders, the dredged material is for the most part polluted with both heavy metals and organic pollutants. Remediation techniques are often economically unacceptable because of the large volume of contaminated sediment (Förstner and Calmano, 1998). Traditionally, dredged sediment is hydraulically raised and placed in confined disposal facilities. To decrease the space required for storage, water can be removed from dredged material using a gravity-driven approach and the resulting material can be disposed into mounds. This technique results in a 50 to 114% increase in the amount of material that can be stored per unit area (Luyssaert et al., 2001).

Afforesting of polluted sites is part of a realistic, low-cost, ecologically-sound and sustainable reclamation strategy for bringing polluted sites into productive use (Dickinson, 2000). Planting trees on these sites initiates soil development and nutrient cycling and improves the visual appearance of the site (Glimmerveen, 1996). Trees are expected to be suitable for achieving extensive and long-term phytoremediation or phytostabilisation. Many authors postulate that trees have the ability to remove significant amounts of heavy metals from the soil (Landberg and Greger, 1996; Ernst, 1996; Dickinson, 2000). This technology, termed phytoextraction, removes heavy metals from the site by repeated coppicing of the trees. Willow and poplar are considered best suited for this task because of their strong nature to coppice, their high capacity for Zn and Cd uptake, and their high biomass production (Schnoor et al., 1996; Greger and Landberg, 1999; Robinson et al., 2000, Roselli et al., 2003).

Other authors state that the feasibility of phytoextraction by planting trees is doubtful (Felix, 1997; Salt et al., 1998; Robinson et al., 2003) since this technique may require an excessive amount of time and since no tree species are known to accumulate all of the most environmentally toxic metals such as Pb, Cd or As. At the same time, phytoextraction has environmental consequences, since heavy metals are brought into circulation via litter fall (Glimmerveen, 1996; Robinson et al., 2000), causing dispersal in the ecosystem.

Phytostabilisation is an alternative remediation technique, not aimed at extraction of the metals, but rather aimed at fixation of heavy metals in the soil. Tree growth stabilises contamination either by immobilisation or by preventing migration (Vangronsveld et al., 1995). Direct stabilisation of the soil is achieved by root growth and the establishment of a litter layer and a vegetation cover (Ross, 1994). The increased organic matter content increases soil aggregation and water holding ability which, together with increased interception and evapotranspiration, reduces leaching losses to the ground water (Schnoor, 2000; Pulford and Watson, 2003).

Whenever phytostabilisation or phytoextraction is chosen as remediation technique, the risk of pollutant dispersal into the environment should be minimized. According to Ross (1994), the input of metals into the food web is potentially more harmful than metal leaching into the groundwater. Recycling of heavy metals through litter fall and litter decomposition can thus be an important pathway for input into the food web (Ross, 1994).

The aim of this preliminary study was to determine which tree species are able to grow on mounds constructed from dredged sediment and to evaluate the possibilities and limitations for phytoextraction and phytostabilisation of these mounds.

Materials and methods

Site description

In Antwerp, Belgium, dewatered dredged sediment from the river Scheldt was used to construct a mound of 9 m height in 1995. In March 1997, a rectangular area of 20 by 90 m on the flattened top of the mound was planted with 5 tree species: two pioneer species, black alder (*Alnus glutinosa* L. GAERTN.) and white poplar (*Populus alba* L.); two climax species, sycamore maple (*Acer pseudoplatanus* L.) and common ash (*Fraxinus excelsior* L.); and one fast growing exotic species, black locust

(*Robinia pseudoacacia* L.). The experimental area was divided in 18 connected plots of 10 by 10 m. Two-year old seedlings were planted in March 1997, 2 years after construction of the mound. For each tree species, seedlings were planted at a triangular spacing of 1.5 m in three randomly chosen plots. The remaining three plots were left unplanted.

Sediment analysis

Samples of the dredged sediment were taken in May 1997 at two soil depths (0-15 cm and 30-45 cm) at 36 points regularly spread over the planted area to permit physical and chemical characterisation. The samples were dried at 45°C until constant weight. The pH-KCl of a 1/5 sediment/KCl solution was determined with an ion-specific electrode (Loeppert and Suarez, 1996). The percentage of organic carbon in the sediment was determined with the method of Walkley and Black (Kalra and Maynard, 1991), total N with the modified method of Kjeldahl (Bremner, 1996), $CaCO_3$ with the simple titrimetric procedure (Loeppert and Suarez, 1996). Electrical conductivity (EC) was measured potentiometric on a 1/5 sediment/water solution (Kalra and Maynard, 1991) and total P colorimetric after colouring the extract using Scheel reagent (Van Ranst et al., 1999). The elements K, Cd, Cu, Fe, Mn, Pb, and Zn were extracted from the sediment sample by a digestion in aqua regia (ISO 11466). Subsequently the element concentrations of the solute were determined using flame atomic absorption spectrophotometry. Beside chemical characteristics, the percentage of sand was determined for six samples of the upper soil layer (0-15 cm) by wet sieving after destruction of $CaCO_3$, organic matter, FeO_2 and MnO_2 and after chemical dispersal to separate the individual sediment particles.

Tree sampling

Tree height, height growth and survival rates were determined in November 1998 after the second growing season. The experiment was untimely stopped in the winter of 1999, owing to the removal of the experimental area as a consequence of harbour expansion. Therefore the planned follow-up of the experiment is not available. Tree foliage on all planted plots was sampled in August 1998. Two bulk samples of five trees each were taken at each plot. Consequently the foliar composition of each tree species was represented by 6 bulk samples. Leaf samples were taken from all leaves of the upper third of the crown. The leaf samples were dried at 45 °C until constant weight and ashed for 4 h at 450°C.During ashing the temperature was gradually raised to avoid Cd volatilisation. Afterward, the ash was digested in concentrated HNO_3. The solute was then analysed for Cd, Cu, Fe, Mn, Pb and Zn concentration by flame atomic absorption spectrophotometry.

Results

Sediment characteristics

The concentrations of most elements were similar at 0-15 cm as at 30-45 cm ($p<0.05$; Table 1 and 2). The similarity of the soil characteristics reflects the high uniformity of the dredged sediment and indicates that distinct soil horizons are not yet present. The high electrical conductivity and high Na concentrations of the dredged material are explained by the estuarine origin of the sediment. In addition, Na and EC were the only variables which were significantly lower ($p<0.05$) in the top layer (0-15) than in the deeper layer (30-45 cm) of the sediment. The gradient is likely caused by leaching of the salts out of the top layer.

The dredged sediment also had a high pH and a high nutrient stock. According to reference data for heavy metals (Kabata-Pendias and Pendias, 1992; Table 2), Cu concentrations were in the normal range, Pb and Zn concentrations were relatively high, and Cd concentrations were more than threefold higher than the upper range. The Cd, Cu, Pb and Zn concentrations were below Flemish legislation limits for re-use of waste material (Table 2).

Growth and survival rate

Two groups could be depicted when survival rates and height growth were considered. More than 90% of the alder, ash and maple seedlings were still living two years after planting (Table 3), but these species showed stunted growth. The second group, existing of Robinia and white poplar, showed higher mortality but also higher growth. Mortality of these two species was lower than 30% and considered to be still acceptable. The average height growth of Robinia and white poplar measured more than 50 cm (Table 3). On almost 31 % of the maple and almost 80% of the ash trees necrosis of the leaf margins was observed, a typical symptom of salt stress (Munns, 2002). The foliage of the other species did not show visual symptoms of salt stress.

Foliar element concentrations

Foliar heavy metal concentrations were measured at the end of the second growing season (Table 4). The observed concentrations were within the normal range which was compiled from different literature references (Table 5), except for white poplar. White poplar foliar concentrations of Cd and Zn exceeded the normal range (Table 4 and 5) and it was the only species in which bio-accumulation occurred. The foliar

Cd and Zn concentrations for white poplar were higher than the Cd and Zn concentrations in the sediment.

Discussion

Tree growth and survival

Despite high sodium concentrations in the sediment, mortality of the planted seedlings was limited to at most 30%. Elevated salt concentrations in soils are reported to cause reduced growth rate and reduced photosynthetic leaf area of the plant due to necrosis of the leaf margins (Munns, 2002; Kozlowski, 1997). No specific literature was found for the planted species, but for *Prunus salicina*, Catlin et al. (1993) reported a salt tolerance threshold for fruit yield of 26 mS m^{-1}, markedly different from the reported EC-values (154 ± 81 mS m^{-1}). Because salts wash out of the soil over time, it is hypothesized that seedling growth will improve during subsequent years. Soil washing might cause salts to leach to the groundwater. As a consequence the location of the mound should be chosen carefully or appropriate measures should be taken to prevent leaching to the groundwater.

Phytostabilisation

The risk of environmental harm will be reduced by choosing tree species that do not accumulate heavy metals, because access to the contaminating metals will be reduced (Glimmerveen, 1996; Pulford and Watson, 2003). Recycling of mainly leaf bound heavy metals through leaf fall should be avoided to minimize the risk of spreading heavy metals into the environment. This was shown by Mertens et al. (2001) and Beyer et al. (1990), both on heavily polluted disposal sites for dredged sediment. On sites covered with non metal-accumulating reed (*Phragmites australis*

[Cav.] Trin. Ex Steud), Beyer et al. (1990) found no elevated metal concentrations in living biota. Elevated concentrations of Cd were found in small mammals living on disposal sites covered with willow (Mertens et al., 2001). Cadmium was thought to be introduced into the food web by willow litter fall. Alder, ash, maple and Robinia showed low foliar heavy metal concentrations and might therefore be suited for phytostabilisation purposes on the experimental site. Also Roselli et al. (2003) found alder and maple to display low metal concentrations in their above-ground tissues on a site with heavy metal contaminated composted sewage sludge. Although the mobility of heavy metals can be increased by the formation of chloride complexes when high amounts of chloride ions are present (Bauske and Goetz, 1993), no elevated concentrations in the leaves of these tree species were found.

An aspect that is often overlooked in the discussions on phytoremediation of contaminated sites with trees is the knowledge that forestry tends to mobilize pollutants through progressive soil acidification (Mayer, 1998). The heavy metals will be maintained in the sediment, until the acid neutralising capacity is used and the pH decreases, leading to an elevated mobility of the metals (Tack et al., 1996). Due to the high $CaCO_3$ concentration in the soil, the mound is not threatened by acidification. It will take 1150 years for the acid neutralising capacity of the upper 0.5 m to be completely depleted in case of aerobe soil conditions. This was calculated, accounting for internal and external acidification and for the acid neutralising capacity of CaCO3, clay and organic matter content (see Luyssaert et al., 2001).

The solubility of Zn in dredged sediment started increasing from pH lower than 6, for Cd it started beneath pH 4 and for Cu and Pb it started beneath pH 2 (Tack et al., 1996). Alder and Robinia are nitrogen fixing species, which will cause an accelerated decline in soil pH (Johnson and Lindberg, 1992). The acid production by N_2-fixing legumes was reported to range from 0.2 to 2.7 mol H^+kg^{-1} biomass produced (Tang et al., 1999). Assuming an acid production of 2.7 mol H^+kg^{-1} and a biomass production of 5 tons ha^{-1}, it could mean that the acid neutralising capacity is used in 570 years instead of 1150 years. It should hereby be stated that N_2-fixing will probably be low because N_2-fixing decreases for high nitrogen content of the soil (Tang et al., 1999). As a consequence, acidification is expected to be lower than estimated. On the short term acidification of the soil is not a threat on brackish sediment mounds.

Phytoextraction

The possibilities and limitations for heavy metal phytoextraction were estimated on the basis of leaf concentrations. Exact calculations require metal concentrations in stem-wood, bark and branches as well as the biomass of those components. For the extraction of metals from the soil by harvesting plant material, trees are needed that tolerate the pollution, coppice readily and take up high amounts of metals in the plant material. White poplar meets these three conditions, and high concentrations of Cd and Zn were found in the leaves of white poplar trees in this study.

It turned out however that white poplar is not suited to clean the soil of Cd and Zn pollution by harvesting tree biomass within a realistic length of time. When only leaves are harvested, an estimated leaf production of 2.5 ton DM/ha.y would yearly export 20 g Cd/ha and 1163 g Zn/ha. This is only about 0.04% of the total stock of both Zn and Cd in the upper 1 m of the soil. Cd concentrations in the wood of willows and poplars are reported to be 1 to 5 times lower than leaf concentrations, but woody biomass production is approximately double the production of litter (Riddell-Black, 1994; Schnoor et al., 1996; Greger and Landberg, 1999; Eriksson and Ledin, 1999; Robinson et al., 2000; Klang-Westin and Eriksson, 2003). This means that if woody biomass is harvested together with the leaves, at most 0.12% of the total Cd stock in the upper 1 m soil can be removed.

Moreover, the use of white poplar, and other poplar or willow species, for phytoremediation has the disadvantage that the soil will only be cleaned from Cd and Zn, not from other heavy metals. If trees are used, coppicing systems have to be developed that prevent recycling of metals in the system via annual leaf fall.

Conclusion

White poplar accumulated large amounts of Cd and Zn and might cause a risk because of recycling of Zn and Cd through litter fall. Ash, alder, maple and Robinia are suited for reclamation of dredged sediment mounds. The visual appearance is enhanced and the trees stabilise the waste. The risk of recycling of Cd, Cu, Pb and Zn in the ecosystem is minimal as these species do not accumulate elevated concentrations of these metals. Alder and Robinia will more rapidly decrease the pH because of nitrogen fixation but because of the high CaCO3 content of the soil it will still take 570 years before acid neutralising capacity of the upper 0.5 m is used. Alder, ash and maple displayed stunted growth because of high soil salt concentrations. Because salt leaches from the soil and because of the high survival rates, better growth is expected the forthcoming years.

An estimate of the possibilities for phytoextraction suggested that none of the five investigated species was suited for phytoextraction of all soil metals. Even when using white poplar for the extraction of Cd and Zn, an excessive amount of time will be required to purify the sediment. The risk for recycling of the metals is an important issue that should be considered.

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Tables

Table 1
Mean and standard deviation of the soil characteristics (n=72 except for sand where n= 7)

depth (cm)	0-15	30-45
Sand (%)	30 ± 5	-
pH(KCl)	7.1 ± 0.1	7.1 ± 0.1
N (mg kg^{-1} DW)	2,019 ± 267	1,885 ± 286
P (mg kg^{-1} DW)	2,441 ± 412	2,209 ± 385
K (mg kg^{-1} DW)	9,155 ± 532	9,159 ± 840
Org. C. (%)	6.2 ± 0.8	6.2 ± 0.8
$CaCO_3$ (%)	9.0 ± 2.5	8.9 ± 2.9
Na (mg kg^{-1} DW)	457 ± 102	823 ± 205
$EC_{1:5}$ (mS m^{-1})	87.9 ± 33.8	219.3 ± 57.1
Fe (mg kg^{-1} DW)	54,202 ± 6,254	53,866 ± 6,061
Mn (mg kg^{-1} DW)	683 ± 77	650 ± 69

Table 2

Mean and standard deviation of the soil heavy metal concentrations (mg kg-1 DW, n=72), reference total concentrations of trace elements in surface soils calculated on the world scale (Kabata-Pendias and Pendias, 1992, mg kg-1 DW), and Flemish legislation limits for re-use of waste material in industrial zones (VLAREA)

Depth (cm)	0-15	30-45	Reference data	Legislation limits
Cd	5.7 ± 0.8	5.9 ± 0.7	0.08-1.61	10
Cu	54.2 ± 6.3	53.9 ± 6.1	4-100	375
Pb	75.2 ± 11.2	74.3 ± 8.1	1.5-70	1250
Zn	358 ± 40	359 ± 46	9-362	1250

Table 3

Survival rate and growth (cm) of the planted trees after the second growing season

	% survival	Growth (cm)	St dev	10% percentile	90 % percentile
Alder	92	5	31	-23	27
Ash	98	2	8	-3	8
Maple	99	5	11	-2	15
Robinia	83	53	39	9	97
White poplar	71	54	39	13	86

Table 4

Foliar heavy metal concentrations the second year after planting (mg kg-1 DW, n=6)

	Cd	*Cu*	*Pb*	*Zn*
Alder	<0.23*	5.8 ± 0.9	5.0 ± 0.5	65 ± 12
Ash	0.3 ± 0.3	12.4 ± 1.8	5.0 ± 1.2	26 ± 8
Maple	0.5 ± 0.3	5.9 ± 1.3	4.5 ± 1.6	74 ± 48
Robinia	<0.23*	8.3 ± 1.2	2.3 ± 0.3	45 ± 5
White poplar	8.0 ± 2.0	3.8 ± 0.4	3.3 ± 0.6	465 ± 125

*: Determination limit

Table 5
Reference data for foliar analyses (mg kg-1)

Species	Cd	Cu	Pb	Zn
Normal ranges in plants (1)	0.05-0.2	5-30	5-10	27-150
Normal ranges in plants (2)	0.1-2.4	5-20	0.2-20	1-400
Normal range in plant material (3)	0.2-0.8	4-15	0.1-10	8-400
Robinia pseudoacacia (6)	0.05	7.0	.8	30
Populus nigra (5)	0.25 ±0.07	5.1 ± 0.6	2.3 ± 0.7	
Populus spp. (4)		14.2		67.1

(1) Kabata-Pendias and Pendias (1992)
(2) Alloway (1995)
(3) Ross (1994)
(4) Reuter and Robinson (1997)
(5) Djingova et al. (1996)
(6) Bargagli (1998)