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Influence of flooding and metal immobilising soil amendments on availability of metals for willows and earthworms in calcareous dredged sediment-derived soils

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Some soil amendments resulted in higher metal uptake by earthworms and willows than when the polluted soil was not amended but submersion of the polluted soil resulted in reduced Cd and Zn uptake in Salix cinerea.

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ABSTRACT

Soil amendments previously shown to be effective in reducing metal bioavailability and/or mobility in calcareous metal-polluted soils were tested on a calcareous dredged sediment-derived soil with 26 mg Cd/kg dry soil, 2200 mg Cr/kg dry soil, 220 mg Pb/kg dry soil, and 3000 mg Zn/kg dry soil. The amendments were 5% modified aluminosilicate (AS), 10% w/w lignin, 1% w/w diammonium phosphate (DAP, $(NH_4)_2HPO_4$), 1% w/w MnO, and 5% w/w $CaSO_4$. In an additional treatment, the contaminated soil was submerged. Endpoints were metal uptake in *Salix cinerea* and *Lumbricus terrestris*, and effect on oxidation–reduction potential (ORP) in submerged soils. Results illustrated that the selected soil amendments were not effective in reducing ecological risk to vegetation or soil inhabiting invertebrates, as metal uptake in willows and earthworms did not significantly decrease following their application. Flooding the polluted soil resulted in metal uptake in *S. cinerea* comparable with concentrations for an uncontaminated soil.

1. Introduction

In situ metal immobilization based on application of amendments has been proposed as an alternative to conventional remediation for contaminated soils (e.g. Adriano et al., 2004). The effects of inorganic additives on the speciation of trace metals depend on soil characteristics such as soil pH, organic matter content or concentrations of plant-available nutrient contents. The efficacy of certain amendments may at least partly be related to pH increases resulting in more alkaline conditions, increasing organic matter contents or nutrient levels. Good results with these approaches have mainly been reported for metal-contaminated acidic soils low in organic carbon or clay content (e.g. Vangronsveld et al., 1996). Next to organic residues (e.g. composts), lime and nutrients (e.g. phosphate compounds), aluminosilicates (zeolites, clay minerals, cyclonic coal ash, modified aluminosilicates), and Fe-oxide and Mn-oxide rich materials (e.g. steel shot) are regularly used for metal immobilization.

Significant areas in Northern Belgium exhibit moderate contamination with trace metals caused by disposal of contaminated dredged sediments (Tack and Vandecasteele, 2008 and references therein) or by the periodical deposition of suspended contaminated sediments in floodplains (Du Laing et al., 2009; Teuchies et al., 2008). Although high $CaCO_3$ contents in most of these soils limit the metal availability in upland situations (Du Laing et al., 2009), several plant species accumulate elevated levels of Cd and Zn in aboveground plant parts. Metal concentrations in soil dwelling organisms and small mammals are elevated compared to reference situations (Tack and Vandecasteele, 2008 and references therein). The aim of this paper is to study the feasibility of using amendments to reduce the bioavailability of metals in metal-contaminated calcareous dredged sediment-derived soils. Amendments previously proven to be successful for calcareous soils or soils with neutral pH were tested for their effects on metal uptake in the willow *Salix cinerea* and in the earthworm *Lumbricus terrestris* in an emerged polluted soil, and on the oxidation–reduction potential (ORP) of a submerged polluted soil.

We selected *S. cinerea* and *L. terrestris* for this experiment as both organisms are found in contaminated alluvial soils or may colonise dredged sediment landfills. *S. cinerea* is known to

Abbreviations: AS, modified aluminosilicate; ORP, oxidation–reduction potential; FW, fresh weight; DAP, diammonium phosphate; DM, dry matter.

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Table 1
Properties for the soils used in the experiments. Values in parentheses denote standard deviations for four replicates (TOC: total organic carbon).

	Cd	Cr	Cu	Pb	Zn	Mn	Ni	As	Clay	TOC	$CaCO_3$	pH_{CaCl_2}	C/N ratio
	mg kg^{-1} dry soil								%	%	%	–	–
Contaminated	25.7 (0.3)	2170 (23)	136 (2.3)	226.8 (55.3)	3044.8 (19.2)	599.2 (15.5)	46.7 (0.3)	48.4 (0.9)	49.0 (1.0)	5.3 (0.1)	8.4 (0.4)	6.7 (0.1)	11.4 (0.3)
Uncontaminated	0.6 (0.1)	50.3 (2.9)	13.5 (0.5)	24.5 (1.1)	104.8 (4.2)	570.2 (40.1)	19.1 (0.8)	8.6 (0.7)	30.2 (6.0)	5.1 (0.3)	3.7 (0.1)	7.1 (0.1)	12.9 (0.1)

accumulate high concentrations of Cd, Zn and Mn in leaves and bark on contaminated alluvial soils in emerged situations (Vandecasteele et al., 2007b). Flooding conditions were found to largely influence the uptake of these elements (Vandecasteele et al., 2007a). *L. terrestris* is known as an accumulator of metals (Kennette et al., 2002). We used *L. terrestris* in the pot trial because this deep-burrowing anecic earthworm species is expected to have a greater value as indicator of changes in metal availability in soil than species like *Eisenia fetida* or *L. rubellus* that live in the litter at the soil surface. *L. terrestris* feeds mainly on organic materials on the soil surface, and incorporates the organic material in the soil. The hydrology of sediment-derived soils may vary from regularly submerged over occasionally flooded to permanent aerobic conditions (Bert et al., 2009), and generally has a clear effect on oxidation–reduction potential (ORP) in top soil and deeper soil (Du Laing et al., 2009). The ORP affects the metal availability in the soil, and indirectly the metal uptake by plants and soil organisms. Therefore the effect of submergence of the amended polluted soil on ORP was assessed as well.

2. Materials and methods

2.1. Plots used for collection of soil, cuttings and leaves

Soil and stem cuttings were collected from two plots in Northern Belgium. Site 1 is a polluted dredged sediment landfill near Destelbergen. Site 2 was an infrastructure spoil landfill near Deinze with baseline metal concentration levels. The 0–30 cm soil horizon was sampled on site 1 (contaminated soil = soil 1) and the 0–15 cm soil horizon on site 2 (uncontaminated soil = soil 2) (Table 1). Soil material was intensively mixed and homogenised, and sieved through a 10 mm sieve. Stones, plastics, twigs and branches were removed. On site 1, eight undisturbed 13 × 13 × 13 cm soil monoliths were taken.

2.2. Experimental design

2.2.1. Effects of amendments on metal uptake in willows

Time is expressed as the week number during the year. Willow cuttings were collected from two-year-old dormant stems from a single shrub at site 2 in week 7 (February 2005) and stored as 1 m stems for 1 week in the refrigerator at 4 °C. The willow stems were wrapped in plastic bags to prevent desiccation.

In total, 72 containers (nine treatments with eight replicates each) were used in the trial. Willow cuttings were 20 cm long and were planted in 13 × 13 × 13 cm containers filled with 1 kg wet soil. Moisture content was 66.6 ± 0.2% (m/m) for the polluted soil 1 and 69.3 ± 1.3% (m/m) for the uncontaminated soil (n = 4). Two cuttings with a total weight between 16.5 and 18.0 g were planted in each container. For assessing initial metal concentrations in willow cuttings, four pairs of cuttings were analysed as a whole and four pairs of cuttings were divided into bark and wood and analysed at the start of the experiment (Table 2). Treatment W1 was the blank treatment with contaminated soil from site 1. Treatment W2 was a reference treatment with uncontaminated soil from site 2. Contaminated soil in treatment W3, W4, W5, W6 and W7 was mixed with 5% w/w modified aluminosilicate (AS) (Metapro n.v., Gent, Belgium), 10% w/w lignin, 1% w/w diammonium phosphate (DAP, $(NH_4)_2HPO_4$), 1% w/w MnO, and 5% w/w $CaSO_4$ on a dry weight base, respectively (Table 3). Lignin is referring here to ground (<2 mm) wood (after removal of bark) of *S. cinerea* with low metal concentrations (1.1 ± 0.1 mg Cd/kg dry weight (DW), 1.7 ± 0.1 mg Cu kg^{-1} DW, 10.4 ± 0.3 mg Zn/kg DW, 34.3 ± 1.2 mg Mn/kg DW). Treatment W8 contained submerged soil from site 1. In treatment W9, undisturbed 13 × 13 × 13 soil monoliths from site 1 were used instead of mixed and sieved soil material.

All containers were placed in a greenhouse with natural day-night regime and temperature, and regularly irrigated with rain water. Each 'upland' replicate

Table 2
Metal concentrations in the cuttings of *Salix cinerea* used in the experiment on metal uptake in willows. Cuttings were split up in bark and wood. Values in parentheses denote standard deviations for four replicates.

	Cd	Cu	Zn	Mn
	mg kg^{-1} dry weight			
Cuttings	2.2 (0.2)	6.1 (3.0)	46.4 (26.2)	145.7 (7.0)
Bark	5.8 (0.4)	5.1 (2.2)	93.3 (15.0)	325.8 (5.3)
Wood	1.4 (0.3)	6.9 (3.6)	38.6 (17.6)	94.9 (25.8)