Faculteit Landbouwkundige en
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Academiejaar 2003-2004

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Metals in dredged sediment-derived soils along the rivers Scheldt and Leie: geographical distribution, bioavailability and ecosystem effects

ir. Bart Vandecasteele

Thesis submitted in fulfilment of the requirements for the degree
of Doctor (Ph.D.) in Applied Biological Sciences

Proefschrift voorgedragen tot het bekomen van de graad van Doctor in
de Toegepaste Biologische Wetenschappen
Op gezag van

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Decaan:
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Het landschap komt als een kelner
op ons toe
met weiden op zijn handen.

Zo eenvoudig is alles.
of zie het sas: de vaart neemt
een boot op zijn schouder
en draagt hem de trappen op.

Herman de Coninck

WOORD VOORAF

De toestand is hopeloos, maar niet ernstig….

Laat mij eerst iedereen bedanken die mij geholpen hebben heeft om dit werk tot een goed einde te brengen. Ir. Bruno De Vos en ir. Jos Van Slycken wil ik heel speciaal bedanken voor hun vertrouwen en hun steun tijdens dit leerproces. Prof. dr. ir. Filip Tack las met veel geduld de manuscripten na en leerde mij het wetenschappelijke wereldje te relativeren. Prof. dr. ir. Bart Muys wil ik bedanken voor het nalezen van de teksten en het verfijnen van de doelstellingen van dit werk. Ir. Paul Quataert wist mij steeds in de goeie statistische richting te duwen en was dan ook onmisbaar voor het maken van dit proefschrift.

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3000 bodemstalen, 2000 bladstalen, 500 strooiselstalen, 200 stekken, 600 regenwormen en 100 bladhaantjes verzamel, pluk, vang, droog, maal, ontsluit en analyseer je niet alleen. Ik heb dan ook heel wat mensen letterlijk in contact gebracht met baggergronden. Bedankt aan iedereen die mijn voorliefde voor zwarte smurrie in allerlei kleuren en geuren gedoogd heeft, en die mij bijstond bij de staalnames, het drogen en malen van de bodem- en bladstalen, en de analyses. Vooral Carine Buysse, Els Mencke, Athanaska Verhelst, Anya De Rop, Ann Capieau, Koen Willems, Koen Vervaet en Mathieu Pieters waren hierbij onvervangbaar. Jürgen Samyn, Linda Meiresonne, Nathalie Cools en Véronique Delanote, de andere teamgenoten, hebben mij geholpen bij de kleine en grote vragen.

Beste Bart Cosyn en Raf Lauriks, het is dus zeker niet voor niks geweest. Wie had ooit gedacht dat er uit deze vuile bodems dergelijke mooie resultaten zouden komen? Alvast bedankt voor de ondersteuning. De andere IBW-collega's wil ik in de bloemen zetten voor de interesse en de fijne werksfeer in deze minder fijne (en helaas zeer onzekere) tijden.

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We willen ook de heren ir. Jan Strubbe, ir. Jozef Van Hoof, ir. Louis De Bisschop, ir. Jan Bal en ir. Ludo Plessers van de Administratie Waterwegen en Zeewezen bedanken voor de financiële ondersteuning van dit project. Het heeft moed gevergd van deze administratie om de onderzoeksopdracht te starten. Het onderzoek levert AWZ argumenten voor een discussie op basis van gezond verstand. Andere administraties en instanties kunnen hier alleen maar uit leren. Koen Mergaert, Nathalie Devaere en Rita Van Ham (AWZ, afdeling Beleid Havens, Waterwegen en Zeewezen) wil ik bedanken voor hun interesse, het nalezen van allerlei teksten, en de vlotte samenwerking. Wordt vervolgd?

De neveneffecten van het wetenschappelijk werk waren ook niet van de poes. 'Hij heeft vandaag een doctoraatshumeur' klonk meestal niet veelbelovend. Vraag dat maar aan Mieke, ouders, zussen, de familie Van Zele of aan 'mie moatn'. Aan jullie om uit te maken of het de moeite loonde. Ik ben alvast bereid de 'verloren' tijd in te halen …

De toestand is hopeloos, maar niet ernstig… Politiek draait vooral om perceptie, en daarbij worden problemen zwart-wit voorgesteld. Problemen verzwijgen omdat ze te complex zijn lijkt mij geen optie. Dit proefschrift toont echter aan dat veel problemen genuanceerd benaderd moeten worden en dat toegepast beleidsgericht onderzoek leidt tot nieuwe inzichten en mogelijke oplossingen.

Bart Vandecasteele

Merelbeke, april 2004

CONTENTS

ABBREVIATIONS

AAS	:	Atomic Absorption Spectrometer
AES	:	Atomic Emission Spectrometer
ALL	:	Alluvial Soil
ALLUV	:	Alluvial Soil
ANOVA	:	Analysis of Variance
BAS	:	Soil with Baseline Contamination Level
BCF	:	Bio Concentration Factor
DA:DW	:	Dry Ash to Dry Weight Ratio
DM	:	Dry Matter
DSDS	:	Dredged Sediment-Derived Soil
DSL	:	Dredged Sediment Landfill
DTPA	:	Diethylene-Triaminepenta-Acetate
DW	:	Dry Weight
EC	:	Electrical Conductivity
Eh	:	Oxidation-reduction potential
FTM	:	Freshwater Tidal Marsh
ICP-AES	:	Inductively Coupled Plasma Atomic Emission Spectrometer
ISL	:	Infrastructure Spoil Landfill
LOI	:	Loss On Ignition
LOAEC	:	Lowest Observed Adverse Effect Level
LOEC	:	Lowest Observed Effect Level
NA	:	Not assessed
NC	:	Not calculated
NOAEC	:	No Observed Adverse Effect Level
NOEC	:	No Observed Effect Level
OC	:	Organic Carbon
OM	:	Organic Matter
OSZ	:	Overbank Sedimentation Zone
PAH	:	Poly Aromatic Hydrocarbon
PCB	:	Poly Chlorinated Biphenyl
PCA	:	Principal Component Analysis
PEC	:	predicted environmental concentration
PNEC	:	predicted no effect concentration
R^2	:	Coefficient of Determination
RSD	:	Residual Standard Deviation
SDS	:	Sediment-Derived Soil
SOM	:	Soil Organic Matter
TOC	:	Total Organic Carbon
VLAREBO	:	Flemish Decree on Soil Sanitation

Introduction

Polluted sediments are transported through the Scheldt basin and finally accumulate on intersection points, tidal branches or in harbours. Especially in the Flemish region both sediment quality and quantity is problematic due to intensive land-use, high urbanisation and industrialisation, and poor sewage treatment. For several decades, periodical dredging of river sediments has been necessary to allow for shipping traffic on the river Leie and Scheldt, on the Diversion Canal, on the Canal Ghent-Bruges and in harbours. As the problem of sediment quality and quantity has historically evolved, it must be put in a historical context for an appropriate assessment.

Important sources of metal pollution in the catchment of the river Scheldt are the transboundary industrial fluxes from Northern France. Transboundary influx of polluted water is screened yearly at the Flemish border (VMM, 1997; VMM, 2000a). Approximately 90% of Cd and Cr in the Flemish surface water are estimated to originate from transboundary pollution (VMM, 2000b).

Landfilling still is the mostly adopted handling option for sediments dredged from inland waters in Belgium. While sediments historically were disposed at convenient locations anywhere along the shore, they now are disposed off in centralised confined disposal areas. To estimate the geographical extent and the environmental consequences of historical sediment disposal on land along rivers in Flanders, a detailed survey was initiated in 1997 on behalf of the Flemish Authorities.

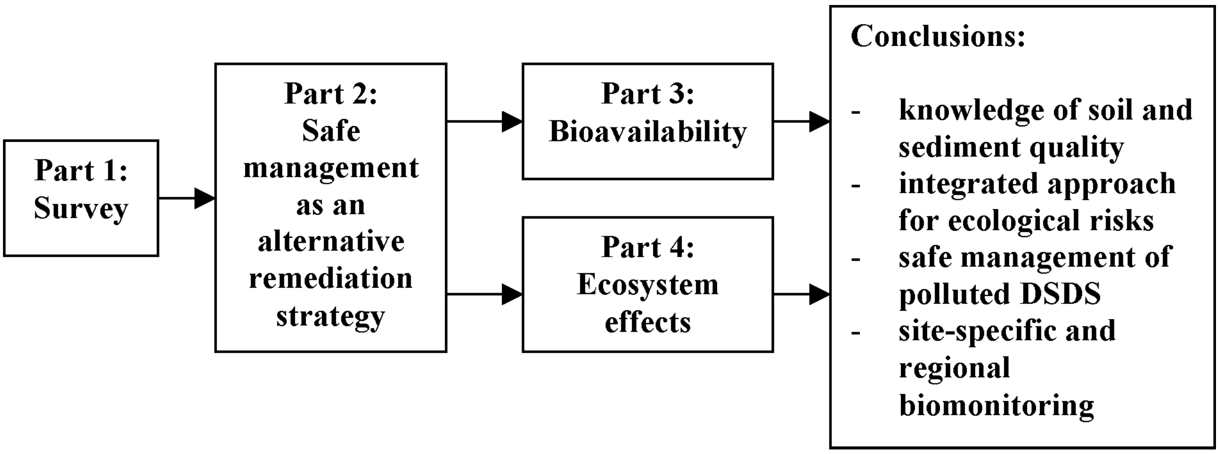
The first goal of our study (Part 1) is to survey the alluvial plains of the Leie and the Derivation Canal, the Canal Ghent-Bruges and the Upper and Sea Scheldt for the presence of dredged sediment-derived soils (DSDS), and to measure and appraise the metal contamination at these sites. Results on geographical impact for the Leie river are presented in Chapter 1.1. An overview of the geographical results for the study area is given in Chapter 1.2.

In Part 2, characteristics and ecological risks of polluted DSDS in the study area are summarised. A concept of safe land use of polluted areas has been developed for uncovered dredged sediment-derived soils in nature conservation areas. Two frequent endpoints are selected for further research: an upland soil type afforested with appropriate tree species, and a wetland soil type with dominance of willow shrubs.

Bioavailability of metals for plants is the topic of Part 3 of the study. Foliar samples were collected for several current land uses on DSDS (maize monoculture, willow forests, poplar plantations and other forestations) for assessment of metal bioavailability. Site and species effects on metal availability for *Salix alba* and *Salix cinerea* is focussed upon in Chapter 3.1, 3.2 and 3.3.

Effects of pollutants on organisms and processes for contaminated soils might vary considerably. In Part 4, research on ecosystem effects of metal pollution for forest floor cycling and soil-forming processes (Chapter 4.1) and earthworm populations (Chapter 4.2) are presented.

Conclusions about four issues are drawn from the results of this study: (1) knowledge of soil and sediment quality, (2) an integrated approach for ecological risks, (3) the safe management of polluted DSDS and (4) site-specific and regional biomonitoring. Results from Part 3 and Part 4 have consequences for site-specific ecological risk assessment of polluted DSDS. However, ecological risk assessment is a complex issue. Future research needs are formulated in the synthesis of this study.



When landfilled sediments are not capped after upland disposal, they gradually transform to a soil substrate allowing for plant rooting and soil invertebrate community development. Therefore, In this study, dredged sediment landfills are referred to as dredged sediment-derived soils (DSDS).

Objectives of this work are:

- Study the influence of former dredging activities along the river Leie on the alluvial plain quality. A soil survey and an archive query for reconstructing the history of dredging operations were conducted simultaneously in Chapter 1.1.
- Summarising ecological risks, opportunities for sanitation and safe management of dredged sediment-derived soils polluted with metals (Part 2).
- Examine for a range of elements whether site or species effect is dominant in determining leaf metal concentrations for volunteer willows on sediment-derived soils and soils with baseline contamination levels (Chapter 3.1.).
- Determine whether the hydrological circumstances, in particular the duration and intensity of submersion, of a polluted dredged sediment-derived soil affect the foliar and bark metal concentration for *S. cinerea*, a typical wetland willow species (Chapter 3.2.).
- Evaluate the bioavailability of Cd, Mn and Zn on a contaminated dredged sediment-derived soil with variable duration of submersion by measuring metal concentrations in the wetland plant species *Salix cinerea* in field conditions during the whole growing season (Chapter 3.3.).
- Assess forest floor decomposition rates and soil forming processes, and evaluate the use of an uncontaminated cover topsoil based on metal concentrations in earthworms after afforestation of a calcareous upland dredged sediment landfill with prevailing aerobic conditions in the upper soil horizons (Chapter 4.1.).
- Assess earthworm biomass and colonisation rate on dredged sediment-derived soils and compare results with observations for the surrounding alluvial plains, and evaluate if differences in earthworm biomass between sites should be accounted for in ecological risk assessment (Chapter 4.2.).
- Draw conclusions from research on geographical distribution, bioavailability and ecosystem effects of metal pollution on dredged sediment-derived soils for integrated ecological risk assessment, safe management and site-specific biomonitoring of polluted dredged sediment-derived soils.

Study area

The study area for this research was the Flemish part of the Upper Scheldt and the Leie, the Derivation Canal between Deinze and Schipdonk, the Canal Ghent-Bruges between Schipdonk and Ghent, the Ring Canal between the Sea Scheldt and the Canal Ghent-Bruges and the section of the Sea Scheldt downstream of the city of Ghent to Temse (Fig. 0.1). The catchment area of the river Scheldt covers about 21.600 km^2 in the north-west of France, the west of Belgium and the south-western part of the Netherlands. The river can be divided in three parts, the Upper Scheldt river, the Sea Scheldt river and the Scheldt estuary. The Upper Scheldt is defined as the upstream part of the river which is not submitted to tidal influence. It comprises the section between the source in Saint-Quentin (France) and the city of Ghent. The Sea Scheldt river, between Ghent and the city of Rupelmonde, is the freshwater zone subjected to tidal influence. The Sea Scheldt is characterised by an unequality of the flow rate at high and low tide resulting in sediment accumulation in the upstream section. The Upper Scheldt was already strongly regulated in the 19th century (e.g. dike constructions) to allow for shipping transport and winter irrigation of the lower parts (van Strydonck & de Mulder, 2000). From 1957 on the Upper Scheldt was canalised in several phases to allow for shipping transport up to 1350 ton. The Leie is a major tributary of the Scheldt river, joining the Scheldt at Ghent. The catchment area of the Leie covers about 2600 km^2 in north France, and 3700 km^2 in the western part of Belgium. In the Leie section between the French border and Deinze, the first major canalization works were executed and sluices were built in the 19th century. In this period the Leie Diversion Canal was dug for a better drainage of the catchment. Between 1965 and 1985 meanders were cut short and the river profile upstream Deinze was broadened to allow for shipping traffic up to 1350 tons and to prevent cities from floodings.

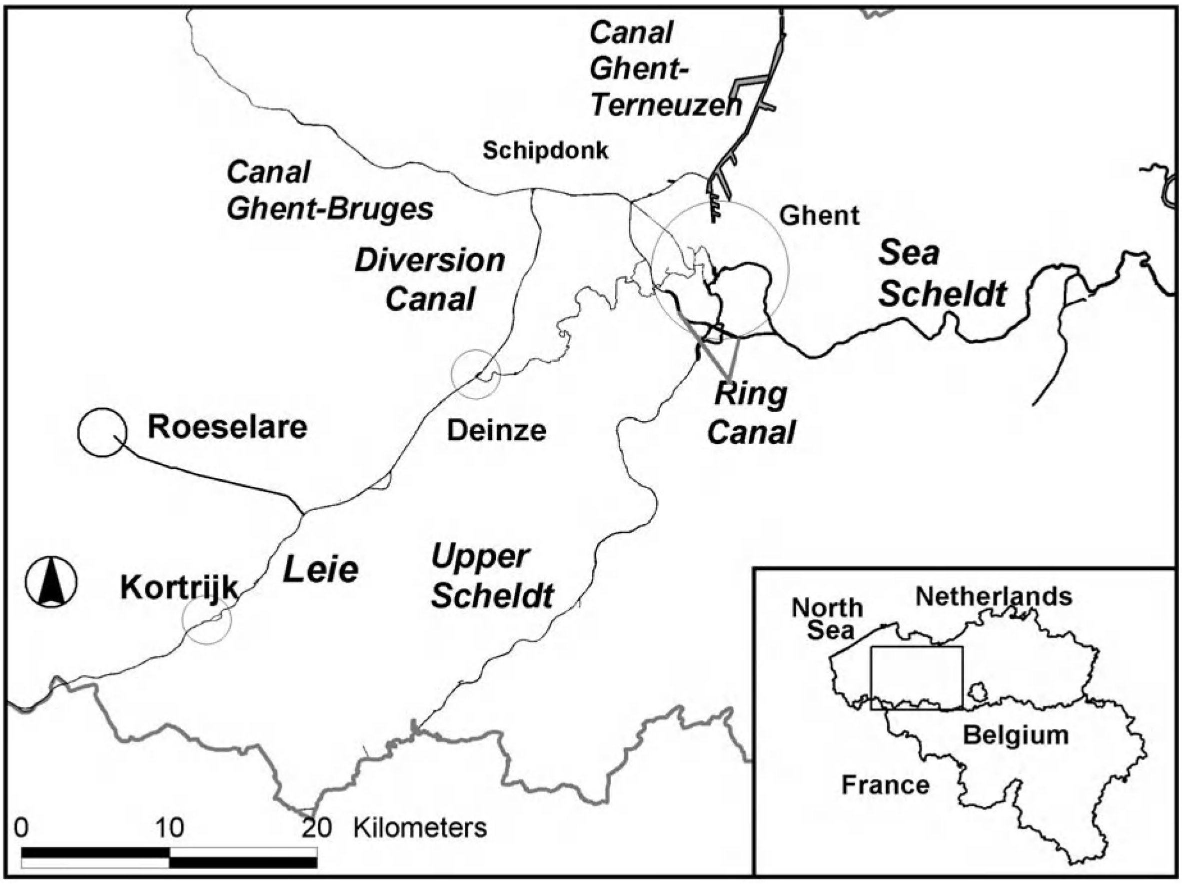


Fig. 0.1. Study area

Part 1. Geographical distribution

Chapter 1.1. Assessment of the pollution status of alluvial plains: a case-study for the dredged sediment-derived soils along the Leie river*

The concept of an integrated multifunctional river management is gaining importance. For major rivers, restoring the contact between rivers and alluvial plains is an important goal as riparian areas have a specific role for several riverine processes. However, former and current human activities are an obstruction or a limitation for river restoration. We studied the influence of former dredging activities along the river Leie on the alluvial plain quality. A soil survey and an archive query for reconstructing the history of dredging operations were conducted simultaneously. The geographical impact expressed as topographical changes and covering of the original soil profile and related processes and biota was large. The pollution status of dredged sediment-derived soils was found to be far from negligible: concentrations of Cd, Cr and Zn were, in 10% of the cases, higher than 20, 480 and 2800 mg kg^{-1} dry soil, respectively.

Both agriculture and nature rehabilitation on dredged sediment-derived soils can only be accepted after profound risk assessment, and management should focus on ecological risk reduction. Results indicate the importance of soil quality assessment in alluvial plains for an integrated river management, rather than *a priori* assuming pristine soil conditions. The collected 'off-line' sediment data can be used as a reconstruction of past sediment pollution, especially when long-term sediment monitoring programs are not available.

1. Introduction

The concept of an integrated multifunctional water management is gaining importance. Restoring the contact between rivers and alluvial plains is an important element in this. Riparian areas have several important functions. They constitute a sink for nutrients (Reddy and D'Angelo, 1994), a reserve for water storage during high discharge events and a habitat for organisms. Former and current human activities have affected the alluvial plains of rivers and are a constraint for river restoration projects. On several locations along the rivers,

* *This chapter is based on a manuscript accepted on November 28th, 2003 for Archives of Environmental Contamination and Toxicology.*

dredged sediments have been disposed. This affected the topography along the river shores and eventually constituted a source of pollution with organic micropollutants and metals.

Sediments act as a sink and a carrier for pollutants and therefore provide time integrated data about geographical and temporal trends in pollutant emission to the river system (Regnier and Wollast, 1993; Sternbeck and Östlund 2001). However, these trends (Petersen et al., 1997, Bordas and Bourg, 2001) may be discerned with much difficulty from samples taken directly from the river (Mellor, 2001). Temporal trends may also be affected by diagenetic processes (Zwolsman et al., 1993). Besides the use of sediment and suspended matter samples taken directly from the river bed (Ollivon et al., 2002; Belzunce et al., 2001), sediment cores from floodplain lakes (Winter et al., 2001), landfilled dredged sediments (Vandecasteele et al., 2002a) and salt marshes (Zwolsman et al., 1993; Christiansen et al., 2002) were used to identify temporal trends in sediment quality.

This research was concerned with the areas affected by dredged sediment disposal along the river Leie in Flanders, Belgium. Their pollution status with respect to metals was assessed. An attempt was made for temporal analysis of sediment quality trends. A comparison of different groups of landfills was based on the dredging history of the river. Based on the results, general recommendations are discussed.

2. Materials and methods

2.1. Study area

The study area for this research was the Flemish part of the Leie (Fig. 1.1). The Leie is a major tributary of the Scheldt river, joining the Scheldt at Ghent. It is not subjected to tidal influence. The catchment area of the Leie covers about 2600 km² in north-west France, and 3700 km² in the western part of Belgium. The river is characterized by a low slope, at 11 cm km^{-1}. In the downstream part between Deinze and Ghent, the slope decreases to near zero and there is a strong meandering. Water pollution of the river already starts close to the French border, for example by the Deûle river coming from the agglomeration of Lille and Doaui (Vallée et al., 2000).

Since the industrial revolution, the river Leie has been strongly regulated. In the Leie section between the French border and Deinze, the first major canalization works were executed and sluices were built in the 19th century because of the economical importance of coal transport (Defoort and Roggeman, 1996). In this period the Leie Diversion Canal was dug for a better drainage of the catchment. Between 1965 and 1985 meanders were cut short and the river profile upstream Deinze was broadened to allow for shipping traffic up to 1350 tons and to prevent cities from floodings. The section downstream Deinze, which is not used for shipping traffic, largely retained the original meandering pattern. The dredged pure sandy or loamy soil materials resulting from the broadening works were used for levelling the lower parts of the alluvial plain (Defoort and Roggeman, 1996). For the section between Deinze and Ghent, human influence by infrastructure works on the river bed was minor, especially because this section lost its shipping transport function since 1970.

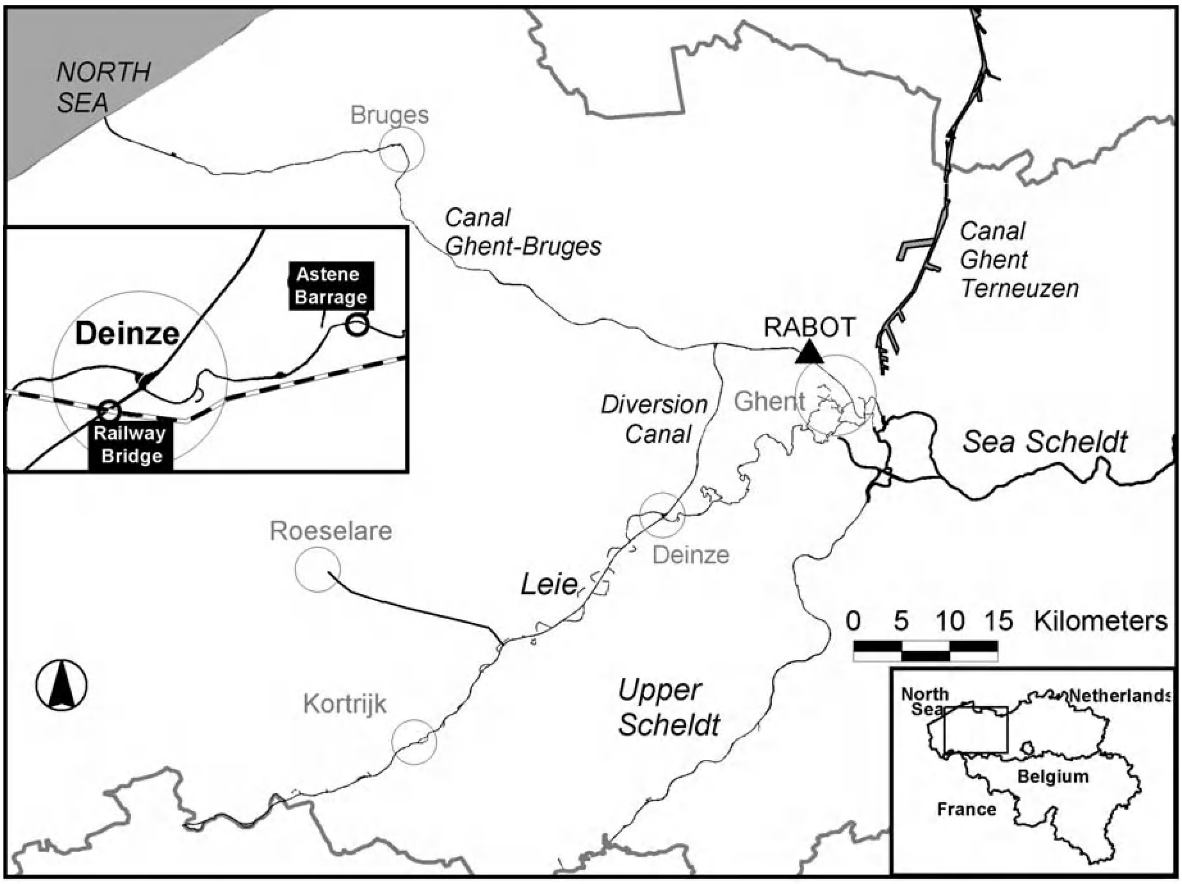


Fig. 1.1. The Leie and Scheldt river with the study area indicated.

The dredging history of the river was reconstructed based on the archives of the Flemish Government Administration. Dredged amounts are displayed as function of period and location of the dredging operation by means of a bubbleplot (Fig. 1.2).

2.2. Data of the sampled dredged sediment-derived soils

In this study we sampled the dredged sediment-derived soils (DSDS) in the Leie alluvial plains constructed before 1995. The procedures adopted to identify potentially levelled-up sites (Vandecasteele et al., 2001), the sampling strategy and criteria used to evaluate the pollution level are described in detail elsewhere (Vandecasteele et al., 2002b; Vandecasteele et al., 2003a). In the upstream part from Deinze, urbanization and industry execute a large pressure on the open areas, and mostly possible DSDS could not be sampled because they were already used for construction. The low [sampled area of the DSDS /dredged amount] ratio in the upstream part of Deinze is clearly illustrated when comparing Fig. 1.3b with Fig. 1.3c. For many of the sampled sites, their former use as a dredged sediment disposal could not be ascertained by historical records. These sites were identified based on field observations, comparative granulometric analyses and chemical analyses as outlined previously (Vandecasteele et al., 2002a). Briefly, criteria were developed, based on a comparison between reference data from soil samples of areas positively identified as affected by dredged sediment disposal and samples from undisturbed alluvial soils along the River Leie and the Upper and Sea Scheldt. In total, 48 sites with a total surface of 125 ha were identified as sites that have been affected by dredged sediment disposal. Observations of these DSDS were used for general pollution assessment and for studying the measured concentrations of metals in relation to location along the river Leie and time of landfilling.

Each DSDS was assigned to one of 4 groups of dredging works (Fig. 1.2). These groups were discerned based on the dredging history of the Leie river. Group (1) included the DSDS in the upstream part of the Gent-De Panne railway bridge (km 44 from the Flemish border crossing). Group (2) DSDS were constructed between 1960 and 1983 and are located between the Gent-De Panne railway bridge and the Astene barrage (km 52 from the Flemish border crossing). Group (3) comprises DSDS in the same section but constructed after 1983 and group (4) the DSDS in the downstream section from the Astene barrage constructed before 1970. The results from the sampled landfills were clustered according to these groups.

For a better detection of the temporal trend, we additionally sampled the Rabot site, the most recent landfill used for the 1994 dredging operation near Deinze but constructed outside the Leie alluvial plains (Fig. 1.1, Group 5 in Fig. 1.2). Group 2-5 all contain landfills constructed with sediments dredged in the most downstream part of the study area, thus comparison is allowed.

The proportion of DSDS and other levelled-up sites in the Leie alluvial plain were calculated based on a GIS-analysis including soil maps, river profile broadenings, constructed and levelled-up areas.

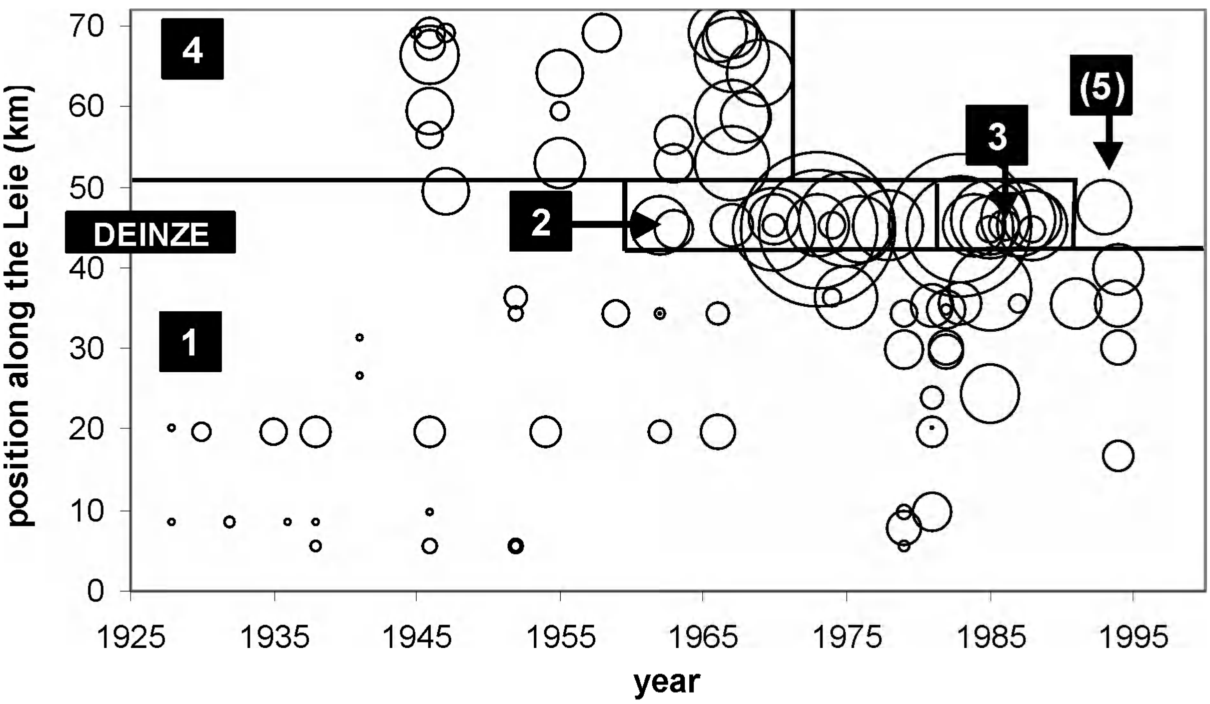


Fig. 1.2. Bubbleplot indicating the dredged amounts as function of both period and distance along the river. The distance is measured downstream from the French border. The circle surface displays the dredged amount ((1) DSDS in the upstream part of the Gent-De Panne railway bridge, (2) DSDS constructed between 1960 and 1983 and located between the Gent-De Panne railway bridge and the Astene barrage, (3) DSDS in the same section but constructed after 1983, (4) DSDS in the downstream section from the Astene barrage constructed before 1970).

2.3. Soil chemical and physical analyses

Soil pH and electrical conductivity (EC) were measured in a 1:5 soil to water suspension. $CaCO_3$ content was determined by back-titration with 0.5 M NaOH of an excess of H_2SO_4 added to 1 g air-dried sediment. Total organic carbon (TOC) in the soil and forest floor was measured with a TOC analyzer equipped with a solid sample module operated at 900°C (Shimadzu 5050A Solid Sample Module Analyser, Shimadzu, Kyoto, Japan). Organic carbon (OC) was determined by the method of Walkley-Black, assuming that this method measures 75% of the total organic carbon. The grain size distribution of the soil samples was determined using laser diffractometry (Coulter LS200, Miami, FL), with the clay fraction defined as the 0-6 µm fraction (Vandecasteele et al., 2002b). Total soil N (N_{soil}) was measured by a NH_4-N distillation method and then titrated with boric acid. Soil total concentrations of Cd, Cr, Cu, Ni, Pb, Zn, P and S are actually pseudo-total *aqua-regia* extractable concentrations measured with ICP-AES (Varian Liberty Series II, Varian, Palo Alto, CA). Digestion was performed using a microwave (Milestone 1200 MS Mega). The accuracy was checked by analysis of a reference sediment material (CRM 320: river sediment) for metals and CRM 100 (Beech leaves) and CRM 101 (spruce needles) for P and S.

2.4. Statistical methodology

Statistical processing started with an explorative principle components analysis (PCA) to detect the major relations between soil parameters. The general characteristics and the degree of pollution of the 4 groups of DSDS were compared with ANOVA. All variables were tested for symmetry and homoscedasticity. No transformation was necessary based on visual inspection of the data. Multiple comparison of means was performed according to the Sidak method with 95% simultaneous confidence intervals (Mathsoft, 1999). This is a conservative method which allows for comparison of groups with a different number of elements. The metal concentrations were compared as such but for temporal trend detection data also were compared after normalization for the average clay content. It is important to know if changes in metal concentrations in the sediments are related to varying sediment properties or not. Normalisation of metal concentrations towards the clay content is an appropriate technique for testing the robustness of the data. Statistical analyses were performed by Splus2000 (Mathsoft, Seattle).

3. Results

3.1. Archive data on dredging history

The bubbleplot (Fig. 1.2) shows dredged quantities as a function of location and period of the dredging operation. The marked increase in amounts with time near Deinze was caused by the works for straightening and broadening the river. This reduced the length of the Flemish part of the Leie upstream Deinze by 40%. It caused a higher fall and consequently an increased flow rate and a changed sedimentation pattern. Near Deinze, the flow rate decreases and part of the water is entering the Diversion Canal. The downstream part of the Leie has not been affected by broadening works during the last decades and continues its strongly meandering course. In the archive data, it is found that before the river broadening most sedimentation occurred in the section downstream Deinze. Fig. 1.3a reveals that dredged amounts increased sharply after 1960. The first large peak coincided with the winter floodings between 1965 and 1968, which resulted in a large sedimentation problem in the downstream part. After 1970, regular large dredging operations were necessary, especially around Deinze. At that time several parts of the river upstream were already straightened and canalized. Current sedimentation rates in the Leie catchment are estimated at 140.000 m^3 $year^{-1}$ (Flemish Government Administration, personal communication). The most recent dredging works were executed in 1994 and 2001. Between 1994 and 2001 no dredging was possible because of developments in environmental law and regulations.

3.2. Pollution assessment

From 125 ha of dredged sediment landfills representing 48 sites, 139 soil samples were collected at 92 sampling points (Fig. 1.3c). Metal concentrations and physico-chemical properties are shown in Table 1.1. Strong correlations of metal concentrations with clay, loam, organic carbon (OC) and $CaCO_3$ contents are clearly revealed by PCA (Fig. 1.4). No distinct clusters of samples were observed. Descriptive data for the sampled alluvial plain soils used as a reference data (not influenced by sediment disposal or overbank sedimentation) are given in Table 1.2. Cd and Zn levels on the DSDS are on an average 10 times higher than in the surrounding alluvial soils. Total metal levels displayed in Table 1.2 are slightly above normal ranges for soils of similar clay and organic carbon content in this region (Tack et al., 1997), while levels measured in the DSDS are considered to be very high. Values for P, $CaCO_3$ and EC are clearly higher for the DSDS than for the alluvial soils.

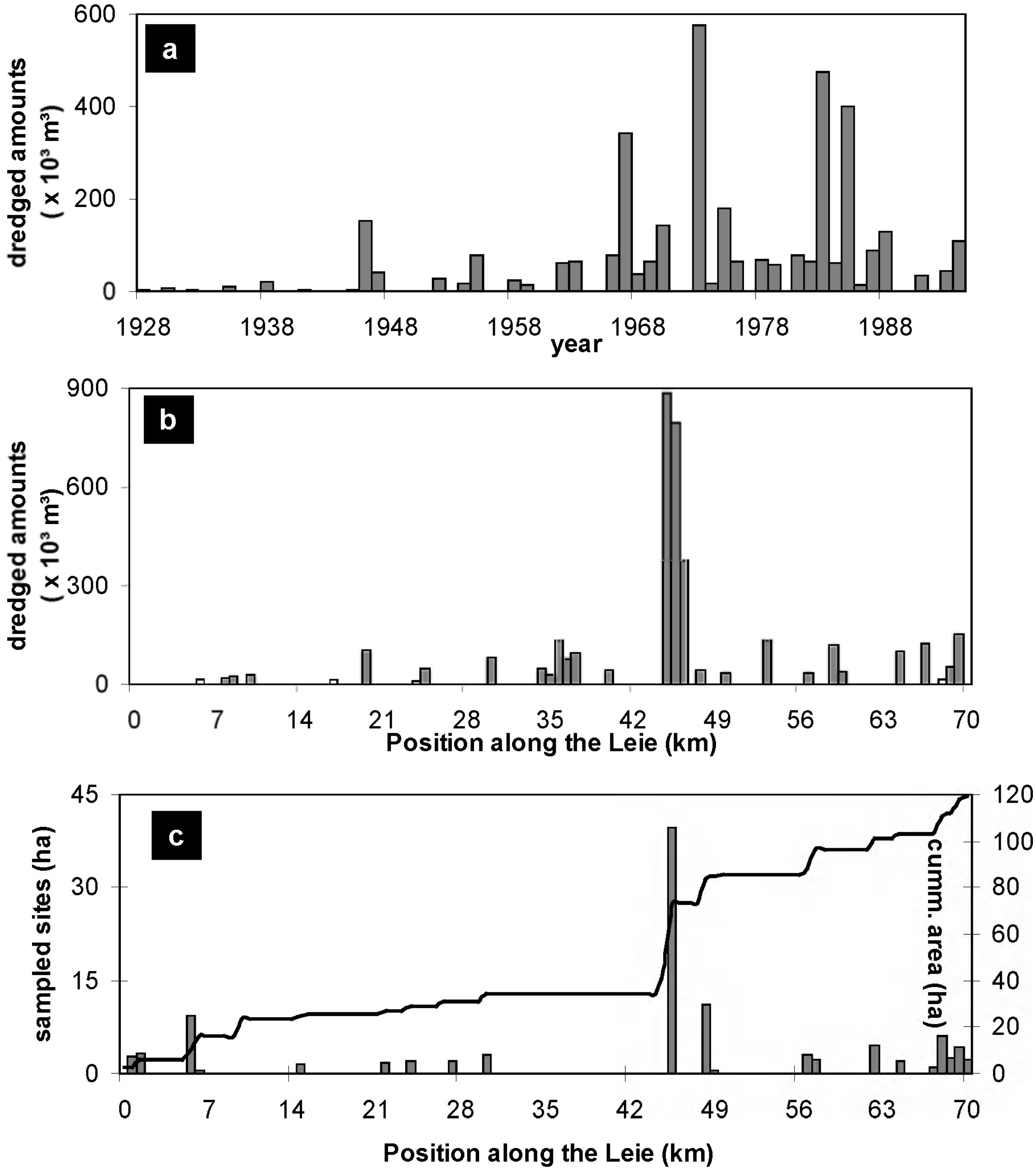


Fig. 1.3. (a) Total dredged amounts for all locations as a function of period and (b) total dredged amounts for all periods as a function of location of the dredging operation, and (c) differential and cumulative distribution of the area of sampled sites as a function of position along the Leie.

Table 1.1. Summary statistics for the dredged sediment-derived soils (139 soil samples)

	Mean	Median	Stdev.	Min.	10th Perc.	90th Perc.	Max.
Cd (mg kg^{-1} DM)	9.7	9.9	6.4	0.3	1.6	19.7	26.4
Cr (mg kg^{-1} DM)	269	232	209	35	93	484	1761
Cu (mg kg^{-1} DM)	190	177	89	18	87	310	512
Ni (mg kg^{-1} DM)	45	45	18	11	21	67	142
Pb (mg kg^{-1} DM)	397	344	249	35	140	703	1139
Zn (mg kg^{-1} DM)	1520	1395	846	77	509	2796	3808
% clay	38	39	12	12	24	53	65
% silt	45	45	9	16	34	56	60
% sand	16	12	16	0	1	41	70
P (g kg^{-1} DM)	2.8	2.8	1.0	0.5	1.4	4.2	5.7
S (g kg^{-1} DM)	3.2	2.3	2.5	0.4	1.0	6.2	13.6
N (g kg^{-1} DM)	3.9	3.8	1.6	1.3	2.3	5.3	12.4
% $CaCO_3$	6.9	6.9	1.5	3.8	5.0	8.8	13.1
% OC	4.1	4.0	1.3	1.8	2.3	5.8	7.4
pH-H_2O	7.5	7.5	0.2	6.9	7.2	7.8	8.0
pH-$CaCl_2$	7.1	7.1	0.2	6.5	6.9	7.4	7.6
EC (µS cm^{-1})	767	465	667	113	169	1844	2550

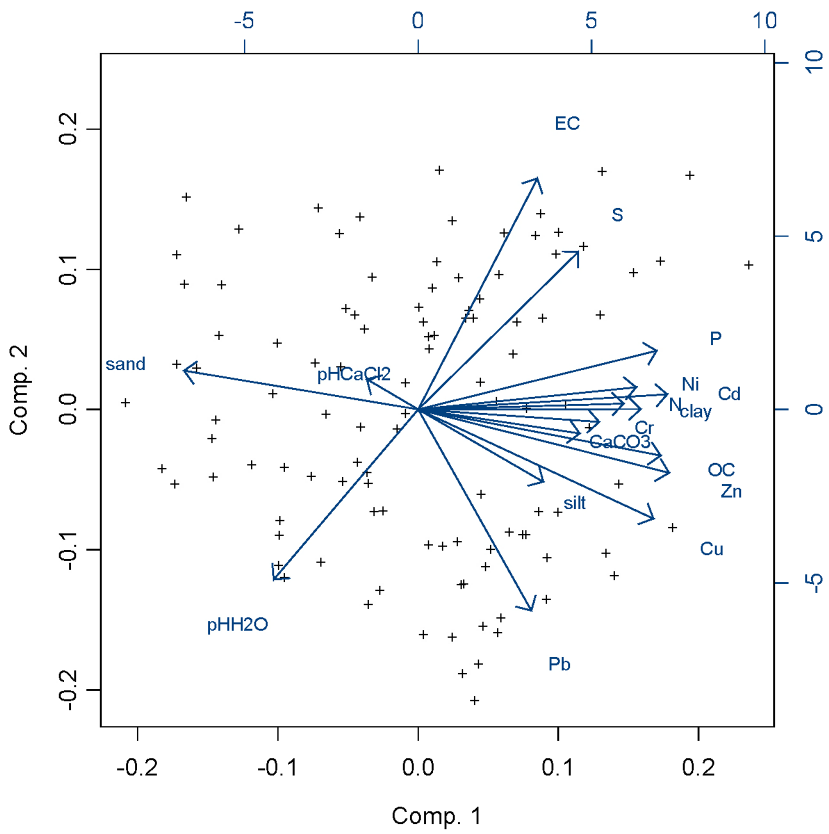


Fig. 1.4. Biplot with results of a PCA of the metal concentrations and other soil characteristics for the 139 soil samples from dredged sediment-derived soils, in which 59% of the variation is explained by the first two components.

From the 92 sampling points (95%) on DSDS, 87 or 95% were found to be polluted with at least one of the measured metals according to the criteria outlined by the Flemish Decree on Soil Sanitation (VLAREBO, 1996). Classification of the measured concentrations of metals according to the reference values (Fig. 1.5) clearly demonstrates severe pollution with Cr, Cd, Zn and Pb. Cu is a minor contaminant, while Ni is of less environmental concern in the area. In 99 of the 139 samples (72%) with soil pollution, Cr, Cd as well as Zn concentrations are above the pollution criterion for agriculture, nature and forestry. Cd pollution is lower than in the Upper Scheldt area (Vandecasteele et al., 2002b) and similar to the Sea Scheldt (Vandecasteele et al., 2003a). In comparison with the Upper and Sea Scheldt, Cr levels are lowest while the Zn, Cu and Pb levels are highest for the Leie DSDS.

Table 1.2. Summary statistics for the reference soil samples from the A horizon of 28 alluvial soils in the study area

	Mean	Median	Stdev.	90th Perc.	Max.
Cd (mg kg^{-1} DM)	0.9	0.9	0.5	1.5	2.3
Cr (mg kg^{-1} DM)	65	66	29	100	119
Cu (mg kg^{-1} DM)	27	23	16	47	78
Ni (mg kg^{-1} DM)	24	26	11	37	46
Pb (mg kg^{-1} DM)	65	57	38	105	192
Zn (mg kg^{-1} DM)	157	139	90	257	441
% clay	21	22	8	31	37
% silt	38	39	12	51	54
% sand	41	38	18	73	79
P (g kg^{-1} DM)	1.1	1.0	0.4	1.6	2.1
S (g kg^{-1} DM)	0.8	0.7	0.4	1.4	1.7
N (g kg^{-1} DM)	4.5	4.6	2.1	7.2	8.3
% $CaCO_3$	1.1	1.0	0.6	1.8	2.2
% OC	3.7	3.9	1.7	5.5	6.4
pH-H_2O	6.3	6.4	0.5	6.9	7.2
pH-$CaCl_2$	5.7	5.7	0.5	6.3	6.5
EC (µS cm^{-1})	130	142	58	210	234

3.3. Geographical impact and morphology of the DSDS

The area of the landfills varied between 0.25 and more than 10 ha and averaged to 2.6 ha. The average area for landfilling was 1.6 ha in the period before 1965, 2 ha between 1965 and 1980 and 6 ha after 1980. For 85% of the sampling points where pollution was found, the polluted sediment was on the surface. The thickness of the polluted layer was lower than 50 cm for 32% of the total area of contaminated sites, and between 50-100 cm for 27% of the area. For 41% of the area designated as contaminated, the thickness of the contaminated layer exceeded 1 meter.

Almost half of the landfill area (45%) was raised between 1965 and 1980, while 25% of the area was raised before 1965, and 30% after 1980. When the cumulative dredged sediment landfill area is plotted against the position downstream along the Leie (Fig. 1.3c), it is observed that 70% of the landfills are situated in the most downstream 1/3th of the study area. There is a distinct clustering of landfills near the city of Deinze. Several landfills of group 3 were constructed along the Diversion Canal, but were filled with sediments from the Leie and the canal outfall.

Table 1.3. Average concentration of metals (mg kg^{-1} dry soil) and descriptive soil properties for the dredged sediment landfills as a function of location (definition of the groups: see Fig. 1.2). Means that are not significantly different are denoted with the same letter (Sidak multiple comparison of means at the 95% level of significance). Background concentration (BC) for metals according to the Flemish Decree related to Soil Sanitation for the standard soil type (1% OC and 10% clay) are displayed too

	1	**2**	**3**	**4**	**BC**
Cd	5.9 a	14.0 b	9.5 a	8.8 a	0.8
Cr	145 a	210 b	152 ab	218 b	37
Cu	154 a	362 c	234 ab	280 b	17
Ni	38 a	54 b	49 ab	40 a	9
Pb	300 ab	350 b	196 a	580 c	40
Zn	1020 a	1935 c	1284 ab	1604 bc	62
clay (%)	31 a	46 c	39 bc	36 ab	
OC (%)	3.4 a	4.2 ab	4.1 ab	4.4 b	
P (g kg^{-1} DS)	2.4 a	3.4 b	3.2 ab	2.5 a	
EC (µS cm^{-1})	742 ab	935 b	1169 b	445 a	

3.4. Spatial trends in DSDS quality

The differences in sediment quality between the four groups of DSDS (Fig. 1.2) were tested with ANOVA. For all metals, phosphorus and clay content, differences were highly significant ($p < 0.001$). Differences were still significant for OC ($p < 0.05$) but not for silt content. Highest levels of Cd, Cr, Ni, Zn and clay were found for group 2 and lowest levels for group 1, while Pb was highest for group 4 and lowest for group 3 (Table 1.3). Cu was highest for both group 2 and group 4 and lowest for group 1. Especially for Cr, the grouped data were highly variable. The low metal concentrations for group 1 can at least partly be related to a lower clay and OC content of the dredged sediment (Table 1.3). Both sediment properties are important for metal binding. It thus can be concluded that important pollution of the sediment with metals was already going on before 1965. For one site in the most downstream section in the vicinity of Ghent, records exist that the landfill was constructed between 1930 and 1935. Maximum metal concentrations measured on that site were 16 mg Cd kg^{-1} dry soil, 290 mg Cr kg^{-1} dry soil, 960 mg Pb kg^{-1} dry soil and 2600 mg Zn kg^{-1} dry soil.

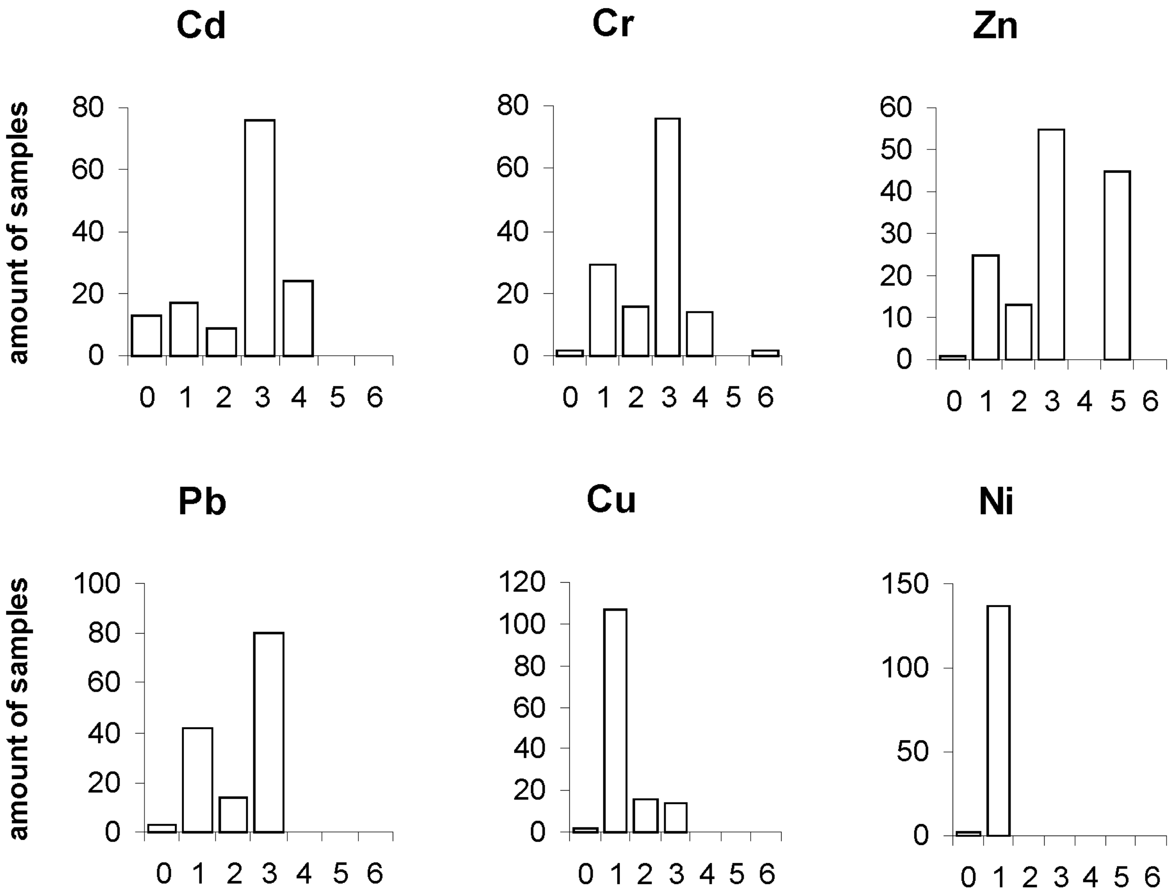


Fig. 1.5. Classification of the metal concentrations of the samples of the dredged sediment landfills over the sanitation standard values of the Flemish Decree on Soil Recovery (0: < background concentration levels, 1: between background concentration level and pollution criterion, 2: > pollution criterion, 3: > sanitation standard value for nature and agriculture, 4: > sanitation standard value for habitation, 5: > sanitation standard value for recreation and 6: > sanitation standard value for industry). The number of samples in each class is shown on the Y-axis.

4. Discussion

The results of this survey served two purposes. On the one hand, they allowed to reconstruct historical sediment pollution of the River Leie. On the other, this study gives information about the influence of sediment disposal on soil quality in the alluvial plains. Along the Leie river, sediments have mostly been disposed close to the location where they were dredged, this in contrast to the Upper (Vandecasteele et al., 2002b) and Sea Scheldt (Vandecasteele et al., 2003a). This greatly facilitates detection of spatial and temporal trends in sediment disposal and sediment quality for the river Leie.

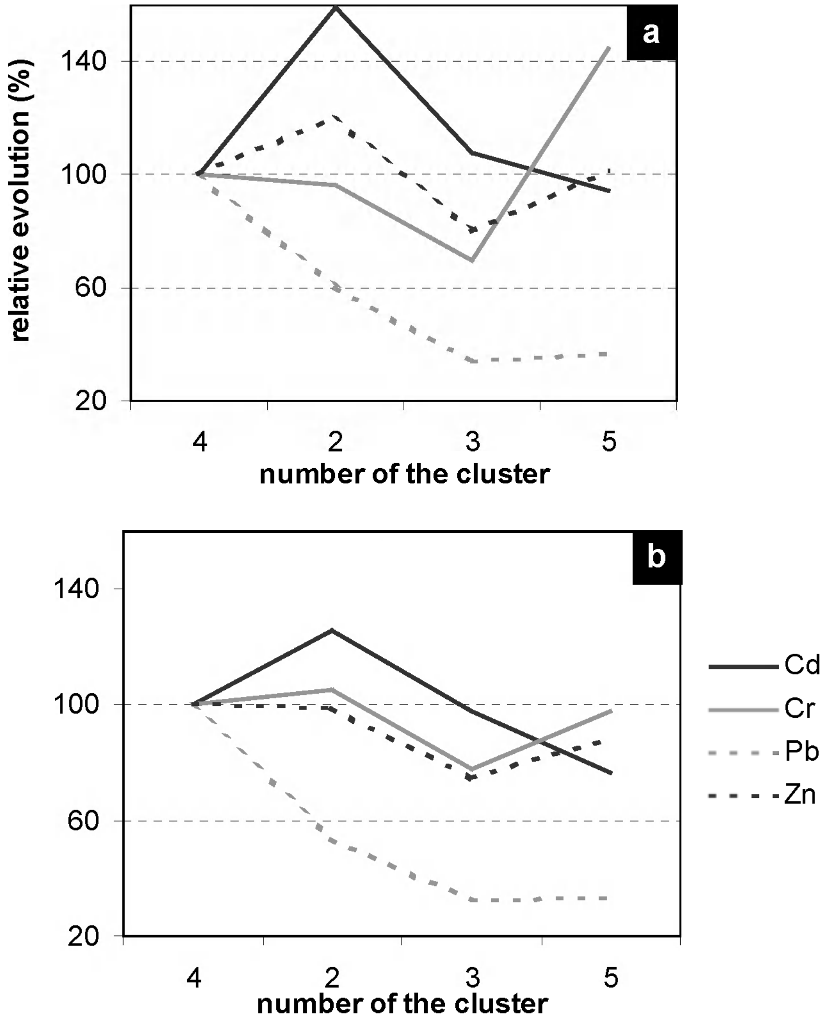


Fig. 1.6. Temporal trends in dredged sediment quality for metals expressed relative to the average value in group 4 (DSDS constructed downstream from Deinze before 1970), with (b) or without (a) normalisation towards the clay content.

4.1. Temporal trends in sediment quality

The collected 'off-line' sediment data (Winter et al., 2001) can be used as a reconstruction of past sediment pollution of the River Leie, especially because long-term sediment monitoring programs are not available. The pre-requisite of metal stabilization in the sediment for temporal comparison of sediment data (Christiansen et al., 2002) is largely met for these calcareous landfilled sediments because of the very low mobility of metals (Vandecasteele et al., 2002b). It is evident from the data that sediment pollution was occurring since long before 1970, especially for the area downstream from Deinze. With respect to the contamination levels encountered, it has to be taken into consideration that sediments were mostly removed from river sections with high accumulation due to a lower flow velocity. Contamination will therefore be concentrated because fine particles with high metal loads preferentially settle in these areas.

The Rabot landfill used for the 1994 dredging operation in the Deinze area was constructed outside the Leie alluvial plains. Comparison of group 4 (before 1970), 2 (1970-1983), 3 (1984-1990) and 5 (1994) reveals a temporal sediment quality trend. Fig. 1.6a depicts the trend in the raw data, while Fig. 1.6b shows the data normalized for sediment clay content. No significant trends were found, except for Pb. Pb exhibits a marked decrease since the seventies. This is also observed in sediment studies of other river basins (e.g. (Smit et al., 1997) for the Rhine-Meuse Delta). The lower metal concentrations for group 3 relative to group 2 represents an important sediment quality improvement in a short period.

Differences in pollution levels may also be related to differences in sediment properties between groups. The strong positive correlation between clay content, organic carbon and metal concentrations is clearly illustrated in the biplot (Fig. 1.4). To correct for the effect of clay content on metal concentration levels, data were normalized with respect to clay (Fig. 1.6b). This resulted in a decrease in variation in Cr, Cd and Zn, but hardly affected the trend observed for Pb. Clearly, the decrease in sediment Pb can be attributed to a significant decrease of Pb inputs into the river system. Cd concentrations show some decrease since the seventies, while no trend is observed for Cr and Zn.

4.2. Structural changes in river systems and sediment quality

The River Leie is mostly canalized and embanked, especially in the upstream part, and water levels are strongly regulated. Overbank sedimentation therefore is very unlikely to happen. In a natural river system, overbank sedimentation is a natural process by which sediments are removed from the water during high discharge events. For two UK rivers, overbank sedimentation rates were determined to be up to 40% of the total sediment load (Walling et al., 1999). For the River Leie, this process was replaced by dredging operations. Riverine sedimentation leads to a thin sediment layer extended over larger areas in the alluvial plains (Martin, 2000), while dredged sediment disposal in the alluvial plains results in discrete landfills scattered over the alluvial plains.

Making rivers navigable locally resulted in levelling up and covering of the original soil and the specific habitat (geographical impact). At the same time, significant contamination with metals and high nutrient levels were introduced. This survey allowed to map the pollution of the terrestrial compartment of the Leie alluvial plain which is connected to pollution of the water body through dredging activities. The data from our survey can be used for ecological risk assessment comparing several rehabilitation and management scenarios for the alluvial plains (Kooistra et al., 2001).

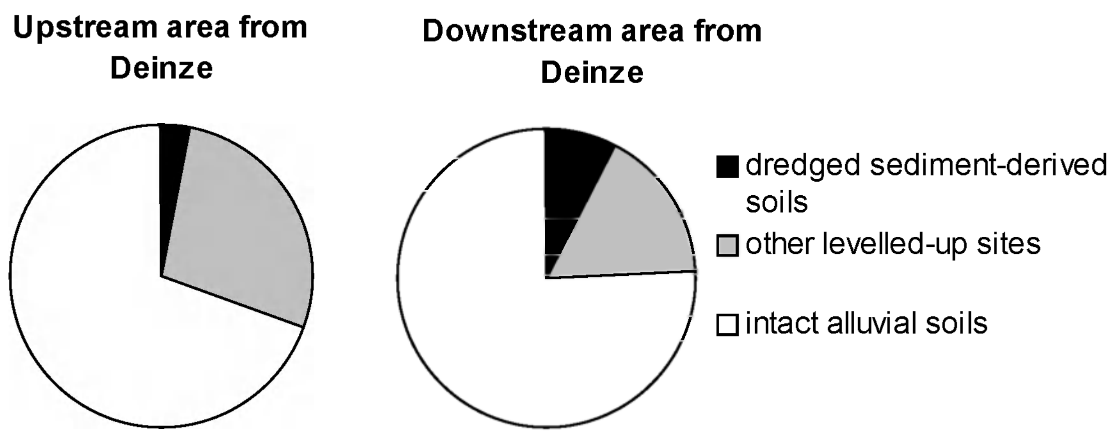


Fig. 1.7. Relative importance of dredged sediment-derived soils and other levelled-up sites for the upstream en downstream area of Deinze.

4.3. Potential adverse effects

The high $CaCO_3$ contents and the small difference between actual and potential pH of all DSDS suggest a strong buffering capacity against acidification of the substrate. Potential leaching of metals is not expected to be a concern (Singh et al., 2000a). However, despite high OC and/or clay contents, bio-availability of metals towards plants and soil dwelling organisms may be enhanced (Singh et al., 1998; Tack et al., 1999; Stephens et al., 2001). Earthworms occupy a key position in the transfer of pollutants towards other trophic levels and have a high potential for Cd accumulation (Hendriks et al., 1995). Bioconcentration of Cd by earthworms was calculated to be a risk for higher trophic levels in the foodweb when concentrations in DSDS exceed 10 mg Cd kg^{-1} dry soil (Beyer and Stafford, 1993). This concentration is exceeded in the superficial layer of the dredged sediment landfills along the Leie in more than half of the sampling points (75 samples from 52 points). On a dredged sediment landfill along the river Leie, elevated Cd and Zn concentrations in leaves of *Salix fragilis* and high Cd concentrations in small mammals were found, indicating potential adverse effects of dredged materials for ecosystems (Mertens et al., 2001). Volunteer willows rooting in polluted dredged sediment landfills showed elevated foliar Cd and Zn concentrations (Vandecasteele et al., 2002c): the Cd concentrations were clearly higher than the 1.14 mg Cd kg^{-1} dry weight threshold value for feedstuffs defined in the EU directive 1999L0029, and in most cases higher than the 6 mg Cd kg^{-1} dry weight permissible level in raw material used as fertilizer (VLAREA, 1998).

4.4. Management of dredged sediment-derived soils

The geographical impact of DSDS can be locally high. Dredged sediments are classified as waste products by European (EU waste catalogue) and regional legislation (VLAREA, 1998). We define the volume-to-area ratio (V:A) as an important measure representing the surface use efficiency for risk and management evaluation purposes. On a site where sediments are filled up to 2 meter the available space is used more efficiently, which is shown by a higher V:A. In general 2 groups of DSDS can be distinguished: (1) recent landfills, with special protective measures, sometimes constructed in former sandpits or above waste dumps and with larger surfaces and higher V:A and (2) the older landfills, which are scattered over the alluvial plain and have a very low V:A. For the recent landfills, clearly stated legislative limitations for reuse of the site are delineated and safe land-uses, usually after capping the site, are planned. For the older landfills, no land-use limitations related to the presence of contamination currently exist, as these sites officially are not identified as being a DSDS. Especially for the last group of DSDS, the low V:A must be evaluated adequately. For all sites without a functional capping, save land-use must be guaranteed and future monitoring of soil processes such as acidification rates and turnover of organic matter is necessary.

4.5. Nature rehabilitation

Rehabilitation of the alluvial plains in the frame of integrated water management will be confronted with several obstructions, e.g. the destroyed meandering pattern, the levelling-up of the wetter parts during the river broadening works (Defoort and Roggeman, 1996), the intensive urbanization pressure on the open areas (upstream from the canal Roeselare-Leie), and both pollution status and geographical distribution of DSDS especially in the vicinity and downstream from Deinze. Landfilling of dredged sediments caused a pollution of the alluvial plains with a more permanent character than the *in situ* sediment pollution itself (Vandecasteele et al., 2002b). River sediment quality is expected to improve gradually once input of pollutants into the river system has been reduced. For example, ongoing sedimentation results in burial of older sediments. Sediment quality improvement in the river can lead to surface sediments becoming non-toxic (Borgmann and Norwood, 2002).

In the upstream part from Deinze, impact of polluted DSDS is lowest. However, in this area the effect of the meander cutting-off and the river broadening works were highest. The proportion of DSDS and other levelled-up sites in the Leie alluvial plain is shown in Fig. 1.7. From an ecological viewpoint, the area downstream from Deinze is most problematic because more strongly polluted dredged sediments were landfilled in this area. At the same time, this is the area that has the highest potential for river wetland restoration because meandering is still relatively intact. The sites all originated before 1980, mainly (80%) before 1965. No protective measures were applied in these periods and these DSDS have since been used as a normal soil, mostly as pasture.

In case of nature restoration projects, more detailed soil survey and research is needed to allow for a correct assessment of the ecological potential of the area. Nature rehabilitation resulting in overbank sedimentation and spreading of polluted sediments must be avoided as long as sediment quality is inappropriate (Smit et al., 1997). When a management objective for nature development on polluted DSDS is proposed, both risks of pollution and feasibility of the selected target must be considered. Three aspects can limit the feasibility: (1) DSDS are levelled-up and thus have a changed hydrology and sometimes another soil texture, (2) DSDS are rich in nutrients and thus can limit the development of certain species and stimulate another vegetation type, and (3) pollution can result in toxicity for target species. First the substrate characteristics and the relative suitability as rooting medium or habitat for plants and soil organisms must be considered. When nature management goals can not be met due to pollution, remediation measures must be considered. Polluted dredged sediment-derived soils in alluvial plains used for nature conservation or agriculture must be properly managed aiming at ecological risk reduction. The presented results warn us that reliable data on soil quality in alluvial areas are necessary, rather than *a priori* assuming pristine soil conditions.

Chapter 1.2. Soil quality in the alluvial plains of Scheldt and Leie river

In Part 1, a field survey was performed for assessment of geographical distribution of polluted dredged sediment-derived soils (DSDS) in the study area. Based on observations about known landfill sites, criteria were developed to identify sites that have likely been affected by dredged sediment disposal (Vandecasteele et al., 2002a). Using these criteria and archive data, dredged sediment affected areas were localised. The data allow for assessment of spatial and temporal trends in metal concentrations. More than 425 ha of the affected areas were found to be polluted with one or several metals (Vandecasteele et al., 2002b, Vandecasteele et al., 2003a, Vandecasteele et al., 2004a). Dredged sediments were found to be highly polluted with metals before 1970. It may point out that sediments were historically polluted. Based on the criteria outlined by the Flemish Decree on Soil Sanitation (VLAREBO, 1996) and Waste Management (VLAREA, 1998), concentrations of organic pollutants (hydrocarbons, PCBs, PAHs) in DSDS in the study area were assessed to be a minor problem when compared with metal concentrations (Vandecasteele et al., 2000). In the period between 1970 and 1990 large quantities of polluted sediments were removed during dredging operations and landfilled in the alluvial plains of the Leie and Scheldt.

Floodplains adjacent to the river (Cottenie and Verloo, 1985; Swennen and Van der Sluys, 2002) and tidal marshes are also affected by sedimentation of contaminated sediments. A new development in integrated water management is the creation of stormwater wetlands, in which overbank sedimentation of polluted sediments may be expected to occur. When discussing sediment-derived soils, “naturally” formed overbank sedimentation zones (OSZ), tidal marshes, and stormwater wetlands on one hand and “human-made” dredged sediment-derived soils (DSDS) on the other must be considered. Soil properties of sediment-derived soils clearly deviate from normal unaffected alluvial soils due to the pollution status and the high nutrient concentrations. An important feature of the sediment substrates is their initially high carbonate content and the resulting high buffering capacity against acidification and the subsequent low leaching potential for metals.

Comparison of sediment quality data for several compartments (river, tidal branches, tidal mud flats and marshes, and DSDS) in the Scheldt basin indicates that the finer fraction of the sediment is stored on the tidal marshes and in tidal branches, or is removed during dredging operations at locations with high sedimentation rates (Vandecasteele et al., 2003c). Metal pollution was found to be similar for these three endpoints. These similar substrates are however very different in legal approach. Dredged sediments are classified as waste products by European (EU waste catalogue) and regional legislation but overbank sedimentation and sedimentation on tidal marshes are considered to be natural processes (Vandecasteele et al., 2003c). Since most old DSDS are used as soil and not as a waste material, the term dredged-sediment-derived soil is appropriate.

Part 2.
Safe management of polluted sediment-derived soils

In Part 1, the alluvial plains of the Leie and the Derivation Canal, the Canal Ghent-Bruges and the Upper and Sea Scheldt were surveyed for the presence of dredged sediment-derived soils (DSDS), and the metal contamination at these sites was assessed. In Part 2, land use and spatial planning objectives for dredged sediment-derived soils in the study area are summarised, and actual and potential risks for polluted DSDS are assessed. When polluted DSDS are assessed to be a serious threat for the environment, sanitation is mandatory. One alternative remediation technique is the safe management of these polluted sites. The feasibility of this option is further explored in Part 3 (research on metal bioavailability) and Part 4 (research on ecosystem effects).

1. Land use and spatial planning

The major river alluvial plains are increasingly being protected for nature conservation goals. However, significant areas were found to be affected by disposal of contaminated dredged sediments or overbank sedimentation. Unlike spatial planning, several DSDS are currently under agriculture. The DSDS generally split up in two groups, with on one hand the recent DSDS being confined disposal sites with a high sediment layer thickness and thus a high surface use efficiency, and a range of protective measures. After landfilling has ended, several recent landfills are protected as nature rehabilitation area. On the other hand, we found several older DSDS, characterised by low sediment layer thickness and mostly situated in lowland areas protected as nature areas. Especially the problems of the latter group are rather complex, as these sites are not generally known as DSDS, no land use limitations exist and areas are planned tot be used as normal alluvial soils for nature protection and integrated water management with inclusion of wetlands in river processes.

2. Risk assessment for historically polluted sites

DSDS are assessed as historically polluted sites according to the Flemish Decree on Soil Sanitation (VLAREBO 1996). Sanitation is only mandatory when pollution is assessed to be a serious threat for the environment. However, risk assessment is a complex issue. Soils in landfilled dredged sediments are high in organic matter, clay and calcium carbonate. Leaching of metals and subsequent ground water pollution is therefore of less environmental concern. However, metal bioavailability for plants and soil biota is a possible threat, in particular in the long-term. Both pollution levels and affected areas determine the extent of the ecological risk involved.

Actual risks are mainly linked with species diversity and abundance and risks of secondary poisoning and dispersal of pollutants. Potential risks are dominated by soil acidification, resulting in a tremendous increase of metal bioavailability (Ma & van der Voet, 1993) and leaching risks (Singh et al., 2000a). Potential risks are a function of the SDS configuration (sediment layer thickness, flooding frequency, hydrology and topography) and land use.

Actual risks are encountered on different levels: (1) plant and soil organisms populations can suffer from chronic toxicity or altered stress levels due to pollution or high nutrient levels and this results in a shift of species and abundance (exposure assessment). Especially the rare or endangered species being most vulnerable are involved here; (2) the presence of SDS result in habitat fragmentation, (3) soil processes and functions such as forest floor decomposition can be hampered (effects assessment) and (4) there is a risk of secondary poisoning for higher levels of the trophic chains.

Soils polluted with metals can result in a reduced food abundance for higher trophic levels (reduced food quantity) and in higher body concentrations for soil invertebrates serving as feed for higher organisms (reduced food quality) (Klok et al., 2000). Higher metal concentrations in calcareous SDS resulted in higher body concentrations for earthworms and isopods (Beyer & Stafford, 1993, Hendriks et al., 1995). An indirect effect of soil pollution is a possible food shortage for higher levels in the food chain (Hörnfeldt & Nyholm, 1996) or a changed, less optimal diet (Groen et al., 2000). Secondary poisoning is highly dependent on configuration of the polluted area and ecology of the target species (Menzie et al., 1992).

Potential risks are mainly dominated by the rate of soil acidification. In calcareous soils, both acid rain and root activity can result in soil acidification. In upland soils, the decalcification rates are 0.015-0.04% $CaCO_3$/year (Van Breemen & Protz, 1988), but in periodically waterlogged soils decalcification rates increase with one order of magnitude to 0.1-0.3%/year (van der Sluis, 1970; van den Berg & Loch, 2000). In permanent waterlogged situations, decalcification rates might be comparable with upland situations. Input of $CaCO_3$ in the topsoil is possible as a result of flooding and circulation of elements due to litter fall. Both fertiliser application (Lorenz et al., 1994) and tillage of polluted SDS result in a decreased pH and a higher metal bioavailability. Agricultural use of polluted SDS must thus be avoided, all the more soil pollution status was found to result in exceeding the permissible levels of metals in public health standards for fodder (Smilde et al., 1982). In contrast, ameliorating the soil nutrient status can reduce toxic effects of pollutants in oligotrophic situations.

3. Soil sanitation and risk reduction

The feasibility for sanitation *sensu stricto* of polluted DSDS with high clay contents is restricted, as metals are strongly bound to the sediment matrix. Alternative remediation techniques are (1) functional capping and isolation of the landfill, (2) application of uncontaminated but permeable covering topsoil, (3) excavation and relocation of the sediment substrate, (4) liming and phosphogypsum amendments, (5) safe management through afforestation or an appropriate hydrological regime and (6) phytoextraction combined with biomass production. The latter option is not further discussed here, but was explored by Meers et al. (2003), Vervaeke et al. (2003) and Vervaeke (2004).

The application of (1) a functional capping system is expensive and is not further discussed here. (2) A clean cover topsoil (Fig. 2.1.) on top of the DSDS provide a clean habitat for plants and soil organisms. For effective risk reduction for soil biota, a capping layer of 40-50 cm must be sufficient (Bosveld et al., 2000). For all plants without higher metal uptake on sediment-derived soils or with a shallow root system, a cover topsoil of this thickness is sufficient to prevent interaction with the deeper sediment layer. For willows and poplars, a thicker covering layer is necessary, and only a liner can probably prevent root penetration into the sediment layer and the resulting higher foliar concentrations (Vandecasteele et al., 2002c; Vandecasteele et al., 2003b). When cover topsoil is used, it is better to avoid willows ands poplars to grow (Fig. 2.1.). We propose to use cover topsoil with similar grain size distributions as the sediment layer. The application of this covering layer results in a levelling up of the site and must be evaluated against landscape and habitat criteria. Therefore we think that it is unreasonable to use clean cover topsoil of 30-50 cm for sites with a sediment layer < 50-60 cm.

(3) Excavation and relocation is a drastic operation resulting in a uncontaminated habitat for plants and soil organisms. Excavation and relocation of older DSDS was applied successfully along the Ghent-Bruges Canal because of nature rehabilitation purposes (Decleer, 1994). In the Netherlands, floodplains were partly lowered by excavation for water management and river rehabilitation objectives (Faber et al., 2001). Feasibility is very dependent on the sediment layer thickness of the sediment-derived soils. We propose that maximum layer thickness acceptable for excavation is 50-70 cm (Fig. 2.1.). For thicker layers, application of uncontaminated cover topsoil is a more feasible management objective.

(4) Amendments of lime and gypsum can reduce soil decalcification and metal bioavailability. Carbonell et al. (1999) concluded that phosphogypsum amendment to Missisipi River alluvial sediments reduced aqueous concentrations of toxic metals to trace levels under anoxic conditions by precipitating these toxic elements as insoluble sulfides. Pierzzynski & Schwab (1993) compared the effect of various soil amendments or combinations of amendments on Zn, Cd and Pb bioavailability in a metal-contaminated alluvial soil and conclude that the limestone amendment was the most effective treatment.

(5) Safe management through afforestation or an appropriate hydrological regime (Fig. 2.1.) aims at reducing the ecological risk and is the issue of Part 3 and 4. Metal bioavailability for willows (Part 3) as the first trophic level in food webs on DSDS and effects of soil pollution on earthworm populations and forest floor decay (Part 4) were focussed on.

4. Safe management of polluted dredged sediment-derived soils

We consider 2 reasons for risk-based management of polluted DSDS: (1) to avoid an increase in ecotoxicological risks when an alternative management for polluted sites is proposed e.g. for nature rehabilitation goals, and (2) to select an appropriate risk-reducing management for polluted sites as an alternative for expensive sanitation operations.

When a management objective for nature development on polluted DSDS is proposed, both risks of pollution and feasibility of the selected target must be considered. Three aspects can limit the feasibility: (1) DSDS are levelled-up and thus have a changed hydrology and sometimes another soil texture, (2) DSDS are rich in nutrients and thus can limit the development of certain species and stimulate another vegetation type, and (3) pollution can result in toxicity for target species. The first aspect is a rather hard condition which is also visually observable in contrast with the remote eutrophic and polluted nature of the DSDS which might be more limiting for the species. First we must consider the substrate characteristics and the relative suitability as rooting medium or habitat for plants and soil organisms. In the Netherlands, an evaluation of toxicity for target species typical for the selected habitat target is used. When nature management goals can not be met due to pollution, remediation measures must be considered (Worm et al., 1998).

Afforestation of upland DSDS and volunteer vegetation on wetland DSDS were selected (Fig. 2.1) as two endpoints for further research about safe management of DSDS. Both endpoints are part of the alluvial landscape in the Scheldt basin. It is thus particularly opportune to monitor metal bioavailability and ecosystem effects in field circumstances.

Submersion and reduction of sediments may result in immobilisation of Cu, Cd and Zn due to binding as sulphides, a lower leaching potential and a reduced bioavailability (Gambrell, 1994). Bioavailability of metals for volunteer willows on wetland DSDS was compared with other contaminated sediment-derived soils and with uncontaminated soils in Chapter 3.1. The effect of submersion on the metal availability for volunteer *Salix cinerea* is further explored in Chapter 3.2. and 3.3.

Revalorisation of polluted soils (Ernst, 1996; Dickinson, 2000) and especially polluted dredged sediment landfills (De Vos, 1989) through forestation has been proposed as a useful tool for landscape planning and forest expansion in urbanised areas. Forest floor decay was monitored during a year, and net changes in soil properties after 16 years of landfilling and 12 years after afforestation (Chapter 4.1.). Earthworm populations were compared for DSDS, overbank sedimentation zones and uncontaminated alluvial soils to assess the effects of metal pollution and colonisation rates (Chapter 4.2.).

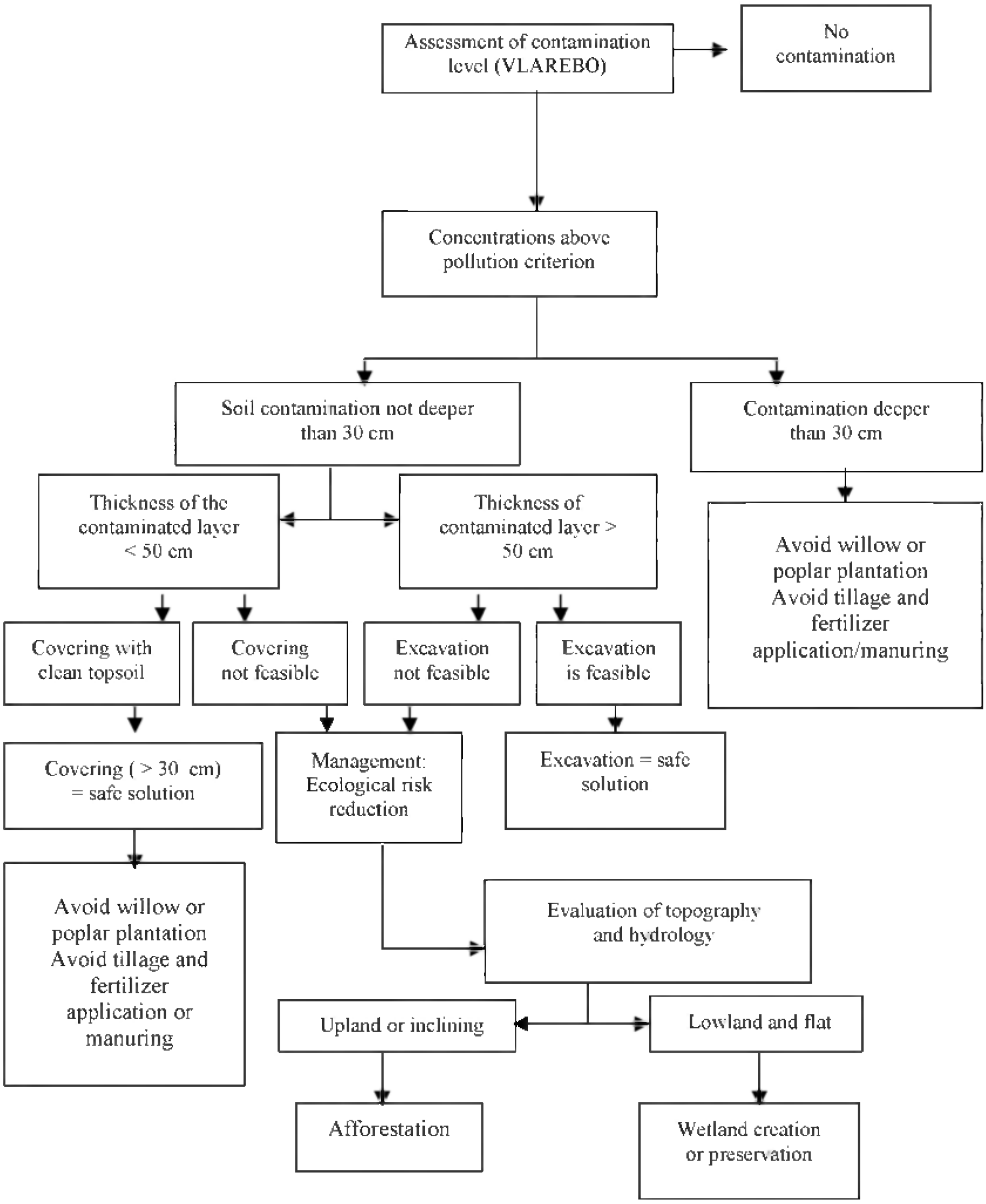


Fig. 2.1. Flowchart for sanitation and management of dredged sediment-derived soils in the Scheldt and Leie catchment

Part 3. Bioavailability

Chapter 3.1. Foliar concentrations of volunteer willows growing on polluted sediment-derived sites versus sites with baseline contamination levels*

Many alluvial soils along navigable waterways are affected by disposal of dredged sediments or overbank sedimentation and contain metal concentrations that are elevated compared to baseline levels. Uptake patterns for metals and other elements by several volunteer *Salix* species growing on these sites were determined during a growing season in field plots and compared with the same species growing on soils with baseline contamination levels. For Cd and Zn, foliar concentrations were clearly higher on dredged sediment landfills. Uptake patterns differed significantly between species. A high uptake of Mn and low uptake of Cu, K and S in *S. cinerea* was attributed to wetland soil chemistry.

Site effects on metal uptake were evaluated in more detail for *Salix cinerea* and *S. alba* growing on different sediment-derived sites under field conditions. Foliar Cd concentrations were higher in *S. cinerea* than in *S. alba.* This appeared a genetic feature not influenced by soil chemical properties, as it was observed both on clean sites and polluted sediment-derived sites. For *S. cinerea*, soil chemistry was reflected in foliar concentrations, while foliar Cd concentrations and bioavailability were found to be independent of the thickness of the polluted horizon. Dredged sediment landfills and freshwater tidal marshes with comparable Cd soil pollution had significantly different foliar Cd concentrations.

1. Introduction

Salix species naturally invade dredged sediment landfills and are the climax vegetation on freshwater tidal marshes (Bal et al., 2001) and other sediment-derived substrates contaminated with metals (Vandecasteele et al., 2002c). Uptake of metals in this vegetation is of environmental concern. Based on DTPA extraction, Zn, Cd and Cu in dredged sediment disposal sites were estimated to be highly plant-available (Singh et al., 1998). Elevated metal concentrations in the pore water also point towards the possibility of enhanced metal availability for plant uptake on such sites (Tack et al., 1998). Overbank deposition of polluted

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sediments on alluvial soils can result in higher plant metal concentrations (Pierzynski and Schwab, 2003).

Willow leaves were observed to be good bioindicators for Cd, Mn and Zn (Piczak et al., 2003; Vandecasteele et al., 2002c). Two evaluation criteria can be used for foliar quality: toxicity towards the plant on the one hand, and bioavailability of metals in the leaves towards higher trophic levels on the other. The adverse effects of metals on willows can be diverse. Pollution causes environmental stress, which, for instance, may be reflected in reduced regrowth after animal browsing (Zvereva and Kozlov, 2001). High metal concentrations in the soil solution can cause the inhibition of root elongation and development (Punshon and Dickinson, 1997a) or reduced transpiration and photosynthesis (Trapp et al., 2000). Elevated foliar concentrations can directly result in higher body concentrations in herbivorous insects (Merrington et al., 2001; Vandecasteele et al., 2003b), birds (Pedersen and Saether, 1999) and mammals (Mertens et al., 2001; Lodenius, 2002), and indirectly influence litter-dwelling organisms (Drobne and Hopkin, 1995) due to higher metal concentrations in the forest floor.

The aim of this paper was to determine site effects on metal bioavailability for volunteer willows. In the first part of the study metal uptake patterns over the growing season were compared between two soil substrates, soils on dredged sediment landfills and soils containing baseline metal concentrations, and between four willow species (Table 3.1). All sampled trees were volunteer species. The primary goal was to examine for a range of elements whether site or species effect was dominant in determining leaf metal concentrations. The second part further focussed on the uptake patterns of *Salix cinerea*, a typical wetland willow species, and *S. alba*, a riparian willow species. Foliar data collected once during the growing season for several sediment-derived sites and control sites with baseline contamination levels were compared to assess the site influence on bioavailability of metals (Table 3.1). We tested the hypothesis that contamination level as well as soil profile genesis and soil physical properties (grain-size distribution and hydrological regime), generally referred to as site effects, influenced foliar concentrations.

2. Materials and methods

2.1. Study area and sampled plots

A plot is defined in this text as a location with relatively homogeneous soil properties where four trees of the same *Salix* species and approximately the same age and diameter were sampled. A site is a larger unit (mostly a landfill) where several plots were sampled.

In the first part of this study, uptake patterns for nutrients and metals over the growing season were assessed (Table 3.1). Selected sites were dredged sediment landfills (DSL), and infrastructure spoil landfills (ISL). Both the DSL and ISL were established by hydraulic filling, but the latter consists of pure mineral and thus uncontaminated soil material from river profile broadening. Eight plots on DSL and five plots on uncontaminated ISL (Table 3.2) with volunteer willows were sampled in 2002 with a 3-week interval between week 18 (first week of May) and week 45 (first week of November). The sampled species were: *S. alba* L., *S. caprea* L., *S. cinerea* L. and *S. viminalis* L. For 1 site on an ISL, four species were found relatively close to each other. On the D3 plot, only three *S. alba* trees could be sampled.

In the second part of this study, uptake of metals and nutrients by volunteer *S. cinerea* and *S. alba* were compared for several sites (Table 3.1) based on foliar samples collected in the second half of August. Selected sites were DSL, overbank sedimentation zones (OSZ), and freshwater tidal marshes (FTM) representing polluted sites, and infrastructure spoil landfills (ISL) and alluvial soils (ALLUV) representing sites with baseline contamination levels. Soils on all sites were thus relatively recent in age. All sampled trees were volunteer with exception of *S. alba* specimens sampled at FTM, for which the origin is not known with certainty. The advantage of studying naturally established plants is that they are assumed to be adapted to the site conditions studied.

The sampled sites for *S. cinerea* (Table 3.3) can be classified in four groups: (a) dredged sediment landfills (3 sites: DSL1 (4 plots, containing plot D1), DSL2 (5 plots, containing plot D2) and DSL7 (6 plots)), (b) sites with polluted floodplain soils referred to as overbank sedimentation zones (OSZ) (5 plots along the Upper Scheldt), (c) sites with slightly contaminated alluvial soils along several rivers and brooks in the province of East-Flanders (ALLUV, 4 plots) and (d) sites with baseline contamination levels (BAS) including infrastructure spoil landfills (5 plots). Baseline contamination levels are defined here as normal baseline concentrations in Flanders (Tack et al., 1997) and are not influenced by contamination due to overbank sedimentation. The DSL7 site (26.5 ha) was used between 1990 and 2000 for sediment disposal from maintenance dredging works in the catchment of the river Yzer and in the polder canals. The DSL1 site (12.5 ha) was landfilled between 1976 and 1983 with sediments dredged in the Leie near Deinze. The DSL2 site (13.3 ha) was landfilled between 1992 and 1995 with sediments dredged in the Upper Scheldt. Both the DSL2 and DSL1 site were used as one basin, while the DSL7 site was subdivided in 12 smaller entities which were landfilled separately. The DSL2 and DSL1 sites can be characterised as wetlands with stagnant water during late autumn, winter and spring. The subsites at the DSL7 landfill had clear texture gradients with the clayey part being wetland, and the sandy part being well-drained.

S. cinerea allowed for comparison of different sediment-derived sites, but was not observed on FTM. *S. alba* specimens were therefore sampled on (a) freshwater tidal marshes along the Sea Scheldt between Wetteren and Temse (11 plots), (b) the relatively uncontaminated DSL7 site (3 plots), (c) several polluted recent DSL in the Scheldt catchment (11 plots, referred as DSL), and (d) infrastructure spoil landfills (6 plots).

Sampled sites are indicated in Fig. 3.1. Site characteristics are summarised in Table 3.2 and 3.3. Metal concentrations in most sediment-derived sites were elevated compared to normal baseline concentration levels in Flanders (90 % percentile values between 0.6 – 2; 37 – 77 and 56 – 100 mg kg^{-1} for Cd, Cr and Zn, respectively (Tack et al., 1997)). Sediment-derived sites are also expected to be polluted with PAHs, PCBs and pesticide residues, but systematic survey data are currently not available.

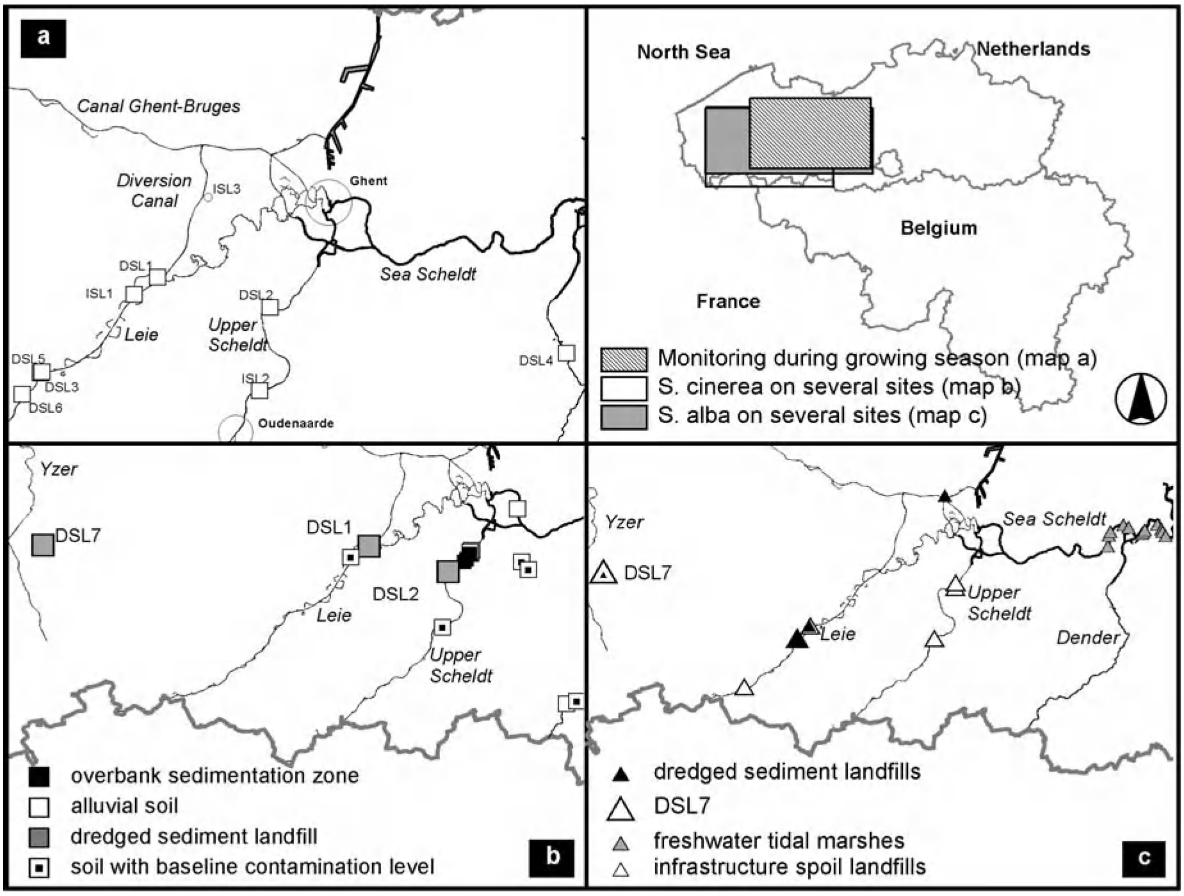

Fig. 3.1. Study area with the sampled sites for (a) several *Salix* species during the growing season, (b) *Salix cinerea* and (c) *S. alba*. DSL: dredged sediment landfills, ISL: infrastructure spoil landfills (= no or low contamination).

Table 3.1. Schematic summary of the sampled sites for the 2 objectives in this study. DSL: dredged sediment landfills, ISL: infrastructure spoil landfills (= no or low contamination), ALLUV: slightly contaminated alluvial soils, OSZ: overbank sedimentation zones (= polluted floodplains), FTM: freshwater tidal marshes, BAS: sites with baseline metal concentrations.

Part 1: uptake pattern of nutrients and metals over the growing season sampling: 3-week interval from May (week 18) to November (week 45)			
	No or low contamination	Slightly contaminated	Contamination high
Salix cinerea			
ISL (□)	ISL1, ISL2		
DSL (■)			DSL1, DSL2
Salix alba			
ISL (△)	ISL2		
DSL (▲)			DSL3, DSL5, DSL6
Salix viminalis			
ISL (○)	ISL2		
DSL (●)			DSL3, DSL6
Salix caprea			
ISL (◇)	ISL2		
DSL (◆)			DSL4
Part 2: Differences in uptake for different sites sampling: second half of August			
	No or low contamination	Slightly contaminated	High contamination
Salix cinerea			
DSL (■)	DSL7		DSL1, DSL2
Other soils	BAS	ALLUV	OSZ
Salix alba			
DSL (▲)	DSL7		DSL
Other soils	ISL		FTM

2.2. Tree and soil sampling

S. cinerea was identified based on leaf morphology (Weeda et al., 1999), and the presence and the length of the *striae* on one-year old branches (Meikle, 1984). Striae are longitudinal lines underneath the bark. *S. cinerea* can be distinguished from *S. aurita* and *S. x multinervis* based on the presence of solely long *striae* on one year-old branches (Meikle, 1984). *S. alba* was mainly determined based on leaf morphology and pubescence (Weeda et al., 1999; Meikle, 1984). In contrast to other *Salix* species, *S. cinerea* is uncommonly used for willow cultures and selection programs.

The standard sampling strategy focused on individual trees or shrubs. To account for the variability associated with sampling, four different trees or shrubs of the same plant species and approximately the same age and dimensions were sampled within a circle with a diameter of 15 m. At least four branches from different heights and positions in the crown were sampled. The use of replicates results in a location specific concentration but with an indication of the variability between individual trees within the population. Samples for the evaluation of the temporal influence on foliar concentrations were taken from trees in a fixed sequence with a three week interval between May and November. Only half of the trees had sufficient leaves to be sampled in week 45. Samples for the second part of the study were taken in the second half of August (week 33). Samples were collected by means of a large catapult (Mathias, 2001) for higher trees (> 10 m) or an extension crosscut saw for trees smaller than 10 m (Blair, 1995). Approximately 1000 cm^3 of leaf samples were collected per tree on each sampling location. Leaves were not washed as washing procedures have to be avoided for food web research purposes (Ernst, 1990) and seem to produce misleading results due to incomplete removal of metals on the leaf surface and partial leaching of metals from the leaf tissues (Kozlov et al., 2000). Luyssaert (2001) showed that foliar Cd concentrations of trees grown at a dredged sediment landfill in the vicinity of the sites studied here are chiefly determined by soil pollution. Absolute contribution of aerial deposition on oleander leaves in a mediterranean area with dry hot summers was in the range of 0.25 mg Cd kg^{-1} DW and 7 mg Zn kg^{-1} DW for the most affected sites (Aksoy and Öztürk, 1997). Baseline concentrations for washed willow leaves are in the range 0.2-3.4 mg Cd kg^{-1} DW and 110 and 560 mg Zn kg^{-1} DW (Severson et al., 1992), thus importance of aerial contamination of willow leaf surfaces is relatively low. Leaves then were dried for 7 days at 40 °C, mechanically ground (Pulverisette 14, Fritsch, Idar-Oberstein, Germany) and stored in dark glass vials before analysis.

On each plot where *Salix* was sampled, the A horizon was sampled in quadruplicate for physico-chemical characterisation. The minimum observed thickness for the A horizon was 15 cm. For the DSL and FTM, the lower depth of the A horizon was hard to define and therefore samples were taken to a depth of 30 cm. Soil profiles on DSL were previously sampled to a depth of at least 1 m for exploratory goals.

2.3. Chemical analysis

DA:DW was determined after ashing oven-dried material in a muffle oven. Ashing was performed at 550 °C with gradual heating and cooling down during 72 hours. Total foliar N was measured by the Kjeldahl method. Total foliar element concentrations were extracted with HNO_3 (p.a. 65%) and H_2O_2 (ultrapur) in a 3:1 ratio using microwave digestion and measured with ICP-AES (Varian Liberty Series II, Varian, Palo Alto, CA). Digestion was performed using microwave (Milestone 1200 MS Mega) with the following program: 250 W (5 min.), 0 W (5 min.), 400 W (5 min.), 500 W (5 min.), 600 W (5 min.), ventilation (10 min.). The accuracy of the foliar element analysis was checked using BCR 60 (Aquatic plant) for Cd, Cu, Mn and Zn, and CRM 100 (Beech leaves) for Ca, Mg, Na, K, S and P. Values obtained in mg kg^{-1} DW were for Cd 2.21 (certified value: 2.2) , for Cu 54.1 (certified value: 51.2), for Mn 1747 (certified value: 1759), for Zn 325 (certified value: 313), for Ca 5443 (certified value: 5300), for Mg 878 (certified value: 878), for Na 234 (certified value: 255), for K 9814 (certified value: 9600), for S 2683 (certified value: 2690) and for P 1599 (certified value: 1550).

The methods used for soil analysis are described in Chapter 1.1. Soil total concentrations of Cd, Cr, Cu, Ni, Pb, S, P and Zn are actually pseudo-total *aqua regia* extractable concentrations measured with ICP-AES after microwave digestion.

2.4. Statistics

ANOVA was used for comparison of both soil and foliar characteristics for the *S. cinerea* and *S. alba* data set after variables were tested for normality and homoscedasticity. Soil data for Cd, S and electrical conductivity (EC) were log10-transformed for both *S. cinerea* and *S. alba*, and Mn was log10-transformed for *S. cinerea*. Foliar Cd and Mn, and BCF for Cd and Zn were log10-transformed for both *S. cinerea* and *S. alba*. Multiple comparison between the sampled soil types and the soil and foliar reference was performed with the Dunnett test (0.95 confidence level). For *S. cinerea*, foliar data for BAS, ALLUV and OSZ were subsequently applied as reference for DSL2, DSL1 and DSL7. Soil data for *S. cinerea* and soil and foliar data for *S. alba* were compared with BAS and ISL data, respectively. Bioconcentration factors (BCF) for Cd and Zn were defined as the ratio foliar concentration/total soil concentration (*aqua regia*).

3. Results

3.1. Foliar concentrations over the growing season

3.1.1. Trends in uptake patterns

Fig. 3.2 reveals a generally decreasing trend in foliar concentrations during the growing season for N, P and Cu, and an increasing trend for Ca, Cd, Zn, Mn and dry ash to dry weight ratio (DA:DW). For Na, concentrations strongly declined during the first weeks of the growing season but increased sharply during autumn. Trends were not clearly expressed for S, Mg and K.

Fig. 3.2 also revealed some trends as a function of the site (DSL or ISL) and willow species. For Zn and Cd, foliar concentrations were mainly determined by soil type, with high foliar concentrations for the dredged sediment landfills (DSL, black symbols). For K, Mn and DA:DW, clear distinctions occurred between *S. cinerea* (square symbols on Fig. 3.2) and the other willow species, irrespective of the soil type. Differences were less expressed for Cu and S. Mn concentrations were higher for *S. cinerea*, while K, Cu and S concentration and DA:DW were lower.

3.1.2. Site effects for Cd and Zn

Site effect was important for Zn and Cd only. Changes in bioconcentration factor (BCF) of these elements during the growing season are shown in Fig. 3.3. In contrast with the absolute foliar concentrations, BCF values were clearly lower for the willows on DSL, indicating that foliar concentrations did not increase linearly with concentration in the soil. The relative uptake pattern for Zn on dredged sediment landfills was proportional to the uptake of Cd and Mn for all species except *S. cinerea*, where Zn uptake was markedly slower early in the growing season (Fig. 3.4).

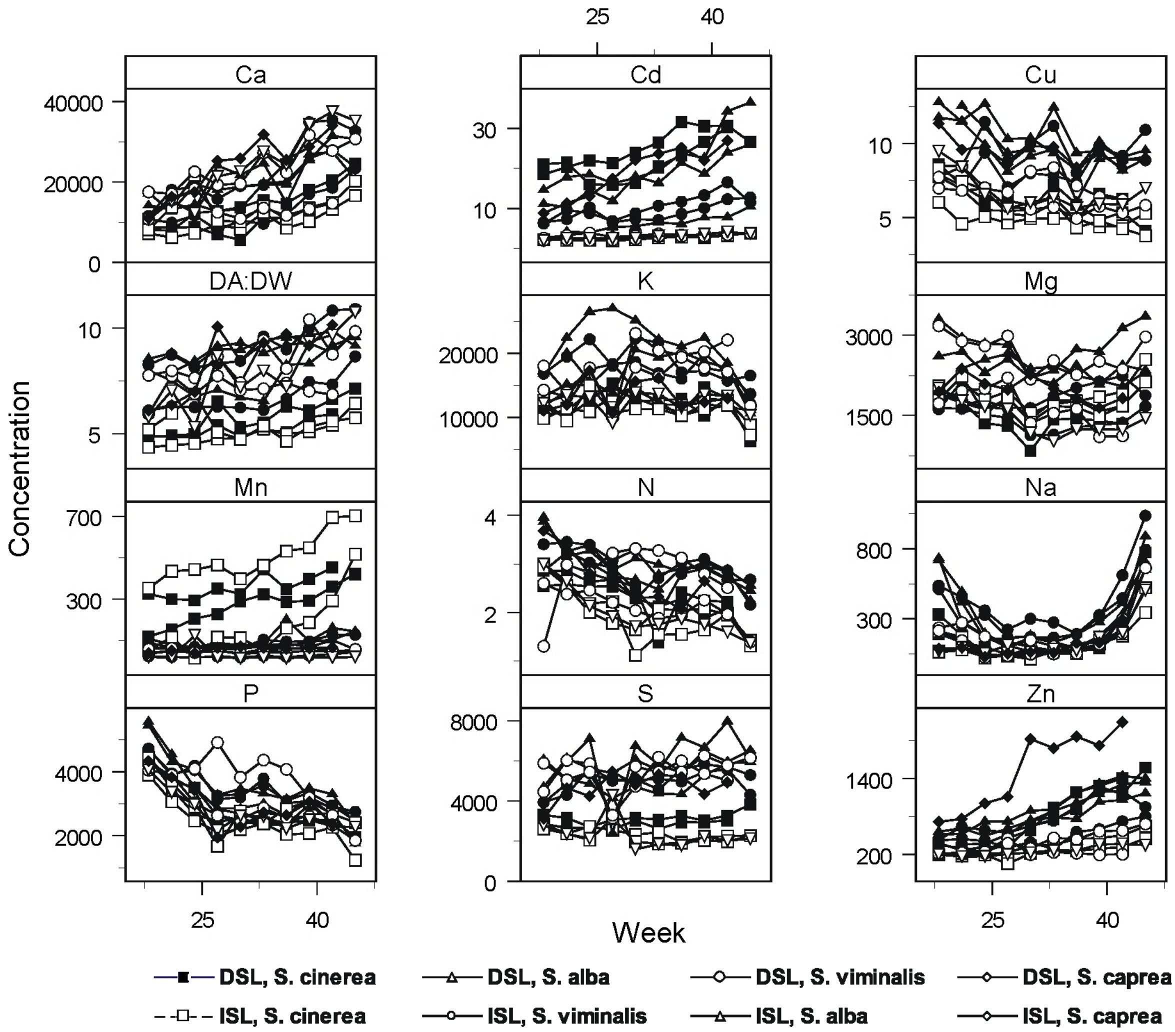

Fig. 3.2. Foliar uptake patterns for Cd, Zn, Cu, Mn, K, Ca, DA:DW, N, S, P, Mg and N during a growing season for willows on dredged sediment landfills (black symbols) and infrastructure spoil landfills (open symbols) for *S. cinerea* (squares), *S. alba* (triangles), *S. viminalis* (circles) and *S. caprea* (diamonds). N and DA:DW is expressed as %, other elements are expressed as mg kg^{-1} DW.

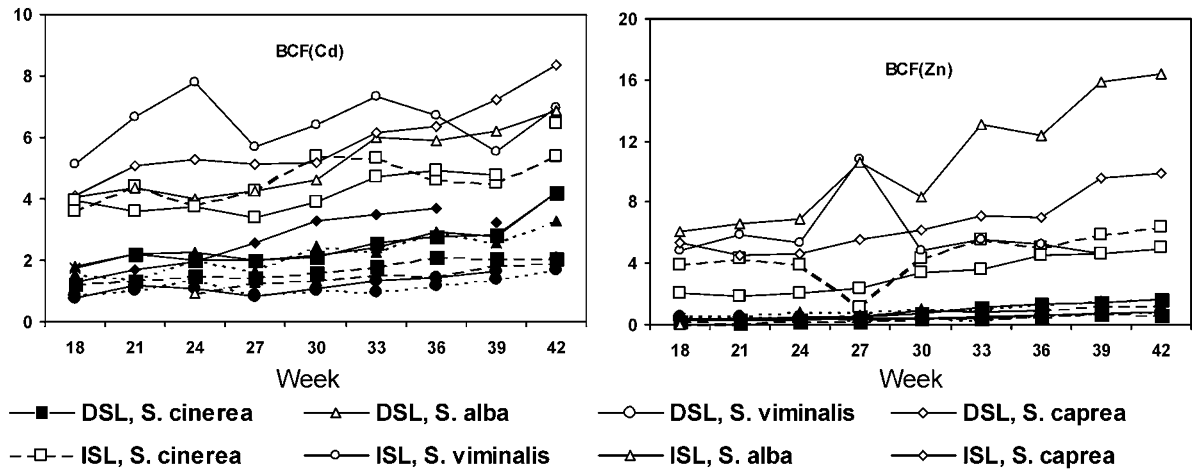

Fig. 3.3. Evolution of the bioconcentration factor (BCF) during a growing season for willows on dredged sediment landfills (black symbols) and infrastructure spoil landfills (open symbols) for *S. cinerea* (squares), *S. alba* (triangles), *S. viminalis* (circles) and *S. caprea* (diamonds).

3.2. Comparison of foliar data for *S. cinerea* and *S. alba* grown on different soil types: site effects

Soil-plant relations for Cd and Zn for *S. cinerea* and *S. alba* (Fig. 3.5) were strongly influenced by other factors than soil concentrations, while for Cu (Fig. 3.5), S (Fig. 3.5), Mn and K (Table 3.4), the species effect on foliar concentrations was more dominant than the effect of soil concentrations. Prior to statistical analysis of foliar data for *S. alba* and *S. cinerea*, soil properties were compared between sites with ANOVA (Table 3.3).

3.2.1. S. cinerea

In general, all sites have a more clayey soil texture and higher soil organic matter (SOM) concentrations than the BAS sites (Table 3.3). DSL2, DSL1 and OSZ are clearly polluted with metals, and are calcareous soils with high P concentrations (Table 3.3).

For foliar Mg, K, Na and N, no differences were detected between the sites. For Cd, Ca, Cu, P, S, DA:DW, Zn and Mn, differences from the BAS sites were significant ($p < 0.01$).

For foliar Cd and BCF(Cd), differences between sites were highly significant ($p < 0.001$). Multiple comparison of the means confirmed that foliar Cd concentrations were significantly higher for DSL2, DSL1 and OSZ than for BAS. In contrast, ALLUV and DSL7 were not significantly different relative to BAS. BCF(Cd) was significantly lower for all sites except for DSL7. BCF for Zn was significantly lower ($p < 0.001$) for all sediment-derived sites (DSL and OSZ) and ALLUV than for the BAS sites.

In addition, foliar data for ALLUV and OSZ were used as reference for multiple comparison with data for DSL2, DSL1 and DSL7. The choice of the reference largely influenced the evaluation of Cd and Zn foliar concentrations on DSL (Table 3.4).

3.2.2 S. alba

Distinct differences in soil properties between sampled sites were observed (Table 3.3). SOM, clay content and N_{soil} was comparable on all sites with exception for the low contents on the infrastructure spoil landfills. The pH_{H20} on all sites was higher than 7. $CaCO_3$ content was lower for the DSL7 site, while EC was particularly high for all DSL. Soil P concentration was high for DSL and FTM, S was high at DSL7, intermediate at DSL and FTM and lowest for ISL. Metal concentrations were higher for DSL and FTM, and lowest for DSL7 and ISL. Only for Zn, soil concentrations were higher for DSL than for FTM.

In contrast to the soil Cd and Zn concentrations for both FTM and DSL, which were significantly higher than the reference, foliar Cd and Zn concentrations were significantly higher only for DSL (Table 3.4). Both BCF(Cd) and BCF(Zn) were lower for FTM and DSL in comparison with ISL (Table 3.4). There was a clear difference in tree age between the sites, demonstrated by the average tree perimeters at breast height (Dbh). These were 20, 25, 44 and 104 cm for DSL7, DSL, ISL and FTM, respectively.

Table 3.2. Soil properties of the dredged sediment landfills (DSL) and infrastructure spoil landfills (ISL) used as reference where willows were sampled during the growing season. Soil metal concentrations are aqua regia extractable concentrations (mg kg^{-1} dry soil). Values in parentheses denote standard deviations (3 replicates)

Site	Plot	Sampled species	Clay (%)	$CaCO_3$ (%)	SOM (%)	pH_{H2O}	Mn	Cu	Cr	Zn	Cd
ISL1	I1	*S. cinerea*	22 (6)	2.3 (1.2)	3.5 (1.4)	7.4 (0.5)	1036 (590)	17 (9)	57 (6)	99 (18)	0.6 (0.1)
ISL2	I2	*S. cinerea*	12 (1)	6.5 (0.8)	2.1 (0)	8.3 (0.2)	208 (14)	6 (1)	41 (3)	50 (16)	0.6 (0.2)
	I3	*S. viminalis*	14 (5)	7.6 (2.8)	2.1 (0.5)	8.3 (0.1)	228 (92)	6 (4)	34 (14)	42 (16)	< 0.5
	I4	*S. alba*	11 (1)	5.7 (0.4)	2 (0)	8.5 (0.1)	172 (7)	4 (1)	31 (4)	36 (6)	< 0.5
	I5	*S. caprea*	9 (0)	4 (0.5)	2.1 (0.4)	8.2 (0.2)	156 (6)	3 (1)	29 (2)	37 (11)	< 0.5
DSL1	D1	*S. cinerea*	39 (2)	8.5 (1.6)	8 (0.9)	7.4 (0)	407 (33)	171 (17)	292 (26)	1766 (175)	15 (1.4)
DSL2	D2	*S. cinerea*	33 (2)	9.9 (0.3)	8.2 (0.5)	7.2 (0.1)	744 (47)	104 (4)	374 (11)	858 (33)	10.7 (0.6)
DSL3	D3, D4	*S. alba, S. viminalis*	30 (10)	7.7 (0.9)	5.3 (2.1)	7.6 (0.3)	336 (68)	120 (57)	166 (73)	864 (448)	7.3 (4.2)
DSL4	D5	*S. caprea*	31 (1)	2 (1)	5.8 (0.1)	7 (0.1)	524 (7)	44 (0)	69 (4)	2143 (37)	6.8 (0.1)
DSL5	D6	*S. alba*	31 (4)	8 (0.6)	6.7 (0.8)	7.3 (0.2)	317 (28)	107 (26)	132 (29)	830 (236)	4.2 (1.4)
DSL6	D7, D8	*S. alba, S. viminalis*	40 (1)	7.2 (1)	11.6 (0.3)	7.1 (0.1)	464 (38)	208 (5)	199 (13)	1602 (77)	8.1 (2.6)

Table 3.3. Average soil characteristics for the sampled sites for *S. cinerea* and *S. alba*. Values indicated in bold are significantly different (Dunnett test with a 0.95 confidence level, ***: $p < 0.001$, **: $p < 0.01$, *: $p < 0.05$) from the BAS (*S. cinerea*) and ISL (*S. alba*) sites used as reference. Soil metal concentrations are *aqua-regia* extractable concentrations. Number of replicates (n) is indicated on the first row (ALLUV: alluvial soils, OSZ: contaminated overbank sedimentation zones, DSL: dredged sediment landfills, FTM: freshwater tidal marshes, NA: not assessed)

	S. cinerea							*S. alba*				
	BAS	ALLUV	OSZ	DSL1	DSL2	DSL7	p	ISL	FTM	DSL7	DSL	p
soil samples (n)	18	14	19	12	12	24		15	48	16	33	
plots	5	4	5	5	4	6		6	3	11	11	
clay (%)	19.7	**31.8**	**33.8**	**36.1**	**38.3**	**40.0**	***	19.1	**39.6**	**39.8**	**38.1**	***
Cd (mg kg^{-1} dry soil)	0.6	**2.3**	**21.0**	**12.8**	**11.9**	0.8	***	1.21	**8.68**	0.72	**7.67**	***
Zn (mg kg^{-1} dry soil)	89	337	**1215**	**1425**	**1137**	315	***	130	**900**	290	**1427**	***
Cu (mg kg^{-1} dry soil)	14.7	35.8	**73.2**	**132.4**	**168.8**	**45.5**	***	17	**137**	41	**182**	***
Mn (mg kg^{-1} dry soil)	540	473	435	417	**724**	NA	*	301	NA	NA	372	
Ca (mg kg^{-1} dry soil)	9177	13773	12173	**38907**	**56283**	NA	***	29502	NA	NA	35537	
Mg (mg kg^{-1} dry soil)	3065	**4677**	3303	**6201**	**4788**	NA	***	3900	NA	NA	**5240**	*
P (mg kg^{-1} dry soil)	670	1115	**1585**	**3093**	**4036**	**1738**	***	658	**4711**	**1685**	**4017**	***
S (mg kg^{-1} dry soil)	379	**1199**	**2022**	**1707**	**2902**	**7287**	***	342	**2619**	**7562**	**3439**	***
SOM (%)	3.7	**10.6**	**13.4**	**8.4**	**8.6**	**15.2**	***	2.7	**8.8**	**11.2**	**8.1**	***
$CaCO_3$ (%)	1.9	2.4	**4.1**	**7.9**	**11.6**	**3.7**	***	6.5	**8.5**	**4.9**	**8.5**	***
pHH_20	6.9	7.1	7.2	7.3	7.4	**6.3**	***	8.1	**7.5**	**6.8**	**7.2**	***
$pHCaCl_2$	6.1	6.5	6.7	**6.9**	**7.2**	6.0	***	7.5	**7.0**	**6.7**	**6.9**	***
EC (µS cm^{-1})	115	**263**	**473**	**504**	**757**	**1291**	***	162	**318**	**1729**	**1174**	***

Table 3.4. Average foliar characteristics for the sampled *S. cinerea* and *S. alba*. Values indicated in bold are significantly different (Dunnett test with a 0.95 confidence level, ***: $p < 0.001$, **: $p < 0.01$, *: $p < 0.05$) from the foliar reference (BAS resp. ISL). Additionally, *S. cinerea* data for DSL2, DSL1 and DSL7 were compared with ALLUV and OSZ, and significantly different results are in *italics* resp. underlined (p-values not shown). Number of replicates (n) is indicated on the first row (ALLUV: alluvial soils, OSZ: contaminated overbank sedimentation zones, DSL: dredged sediment landfills, FTM: freshwater tidal marshes)

	S. cinerea							*S. alba*				
	BAS	ALLUV	OSZ	DSL1	DSL2	DSL7	p	ISL	FTM	DSL7	DSL	p
foliar samples (n)	20	16	20	16	20	22		24	44	10	43	
plots	5	4	5	4	5	6		6	11	3	11	
Cd (mg kg^{-1} DW)	5.1	9.0	***21.3***	***15.5***	***15.2***	*4.7*	***	3.3	3.3	2.2	**7.8**	***
Zn (mg kg^{-1} DW)	508	765	**909**	578	700	638	**	300	361	**626**	**732**	***
Cu (mg kg^{-1} DW)	6.2	5.5	5.4	**4.7**	5.1	*6.7*	***	10	9.4	9.3	10.6	
Mn (mg kg^{-1} DW)	536	429	262	267	389	815	**	33	64	**82**	**122**	***
Ca (mg kg^{-1} DW)	16123	**12635**	**12459**	13116	**11700**	**12483**	**	25438	**20589**	22795	**19143**	***
P (mg kg^{-1} DW)	3774	4222	3968	*3135*	*3081*	*3220*	**	2852	2973	**3570**	3064	*
S (mg kg^{-1} DW)	2596	2676	2896	**3114**	2966	***3235***	**	5692	6188	**7289**	6073	*
N (%)	2.16	2.39	2.40	2.25	2.10	2.33		2.39	**2.67**	2.3	**3.07**	***
DA:DW (%)	6.1	**5.3**	5.6	**5.1**	**4.9**	**5.2**	***	9.4	8.9	8.6	**8.3**	**
Na (mg kg^{-1} DW)	79	77	93	66	81	106		107	179	195	80	**
K (mg kg^{-1} DW)	11659	11005	12881	12005	10370	12396		21514	**17456**	19543	19490	**
Mg (mg kg^{-1} DW)	2146	1989	**1675**	**1704**	1851	1777	*	2370	2624	**3016**	2138	***
BCFCd	9.18	**4.53**	***1.73***	***1.29***	***1.40***	5.41	***	3.92	**0.41**	3.35	**1.09**	***
BCFZn	6.48	**2.66**	***1.11***	***0.44***	***0.68***	**2.28**	***	4.85	**0.42**	2.46	**0.56**	***

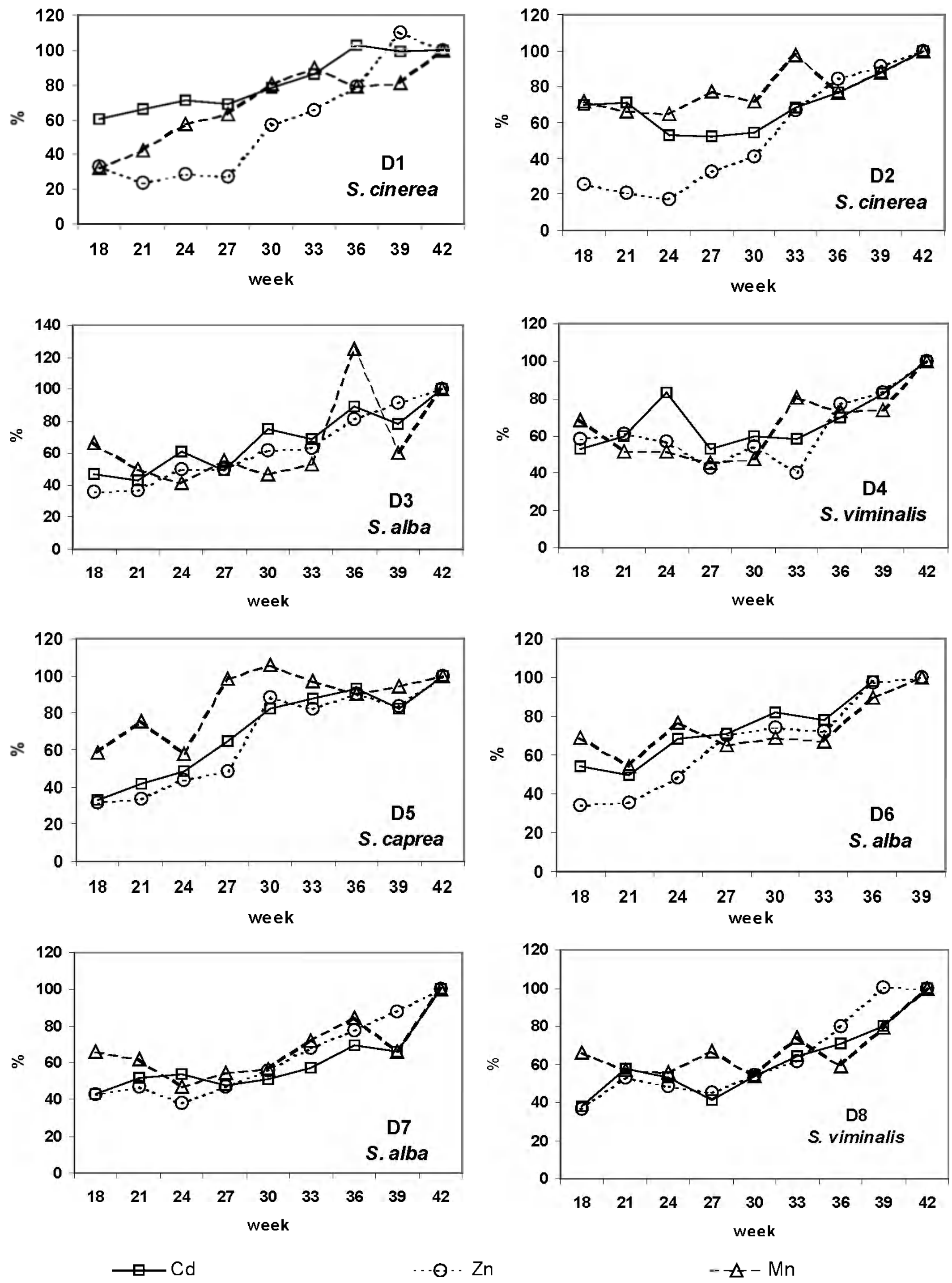

Fig. 3.4. Relative uptake of Cd (squares), Mn (triangles) and Zn (circles) during the growing season for willows on 8 plots (plots numbered D1-D8, see Fig. 3.1 and Table 3.2) on polluted dredged sediment landfills. Values are averages for 4 individually sampled trees and are expressed relative to the concentration measured in week 42.

4. Discussion

4.1. Evolution of foliar concentration over a growing season

4.1.1. Trends in uptake patterns

The observed trends over the growing season for Cd, Mn, Na, Mg, N, P, Ca, and S (Fig. 3.2) are comparable with data from Ernst (1990) for several deciduous tree species, from Piczak et al. (2003) for trees in urban areas and data from Vervaeke and Lust (2001) for young *S. triandra* and *S. fragilis* on a dredged sediment landfill. Increasing Zn concentrations over the growing season were also reported by Vervaeke and Lust (2001) and by Piczak et al. (2003).

Foliar S, K, Mn and Cu concentrations in *S. cinerea* were clearly different to other sampled *Salix* species (Fig. 3.2, Table 3.4). *S. cinerea* is a characteristic species in willow brushwood on submerged sites, while the other sampled species are typically found in riparian willow forest that is periodically flooded (Bal et al., 2001). Even in summer *S. cinerea* can survive in stagnant water (Weeda et al., 1999), and is thus characterised as a wetland plant species. The observed differences in uptake patterns may be related to a different biogeochemical behaviour of the elements between the soils. In reduced soils, Cu and S may present as insoluble sulphides, while Mn may be more available when oxidation-reduction potential decreases (Gambrell, 1994). Zn seems to be less available than Cd and Mn in the first half of the growing season for *S. cinerea* (Fig. 3.4). The differences in foliar concentrations between *S. cinerea* and *S. alba* for Cu, Mn, S and K are also clearly illustrated by the data in Table 3.4 and Fig. 3.5. In contrast to observations on cereals on a polluted dredged sediment landfill (Smilde et al., 1982), no Mn deficiency was observed or expected based on the foliar concentrations.

4.1.2. Site effects for Cd and Zn

Both on DSL and sites with baseline contamination levels, foliar Cd concentrations are higher for the wetland plant species *S. cinerea* (Table 3.4), while one would expect a lower availability based on the lower oxidation-reduction potential in wetlands (Gambrell, 1994). Most sampled plots for *S. cinerea* on sediment-derived sites were characterised by stagnant water until mid May. Soil drying and oxidation in the beginning of June may have caused an increased bioavailability of Cd on these particular plots. Subsequent soil drying and oxidation is expected to result in a higher bioavailability of metals in the soil substrate (Tack et al., 1998). Van den berg et al. (1998) concluded that fluctuating hydrological conditions in a polluted wetland soil resulted in higher metal concentrations in pore water during summer as a result of both organic matter decomposition and sulphide dissolution.

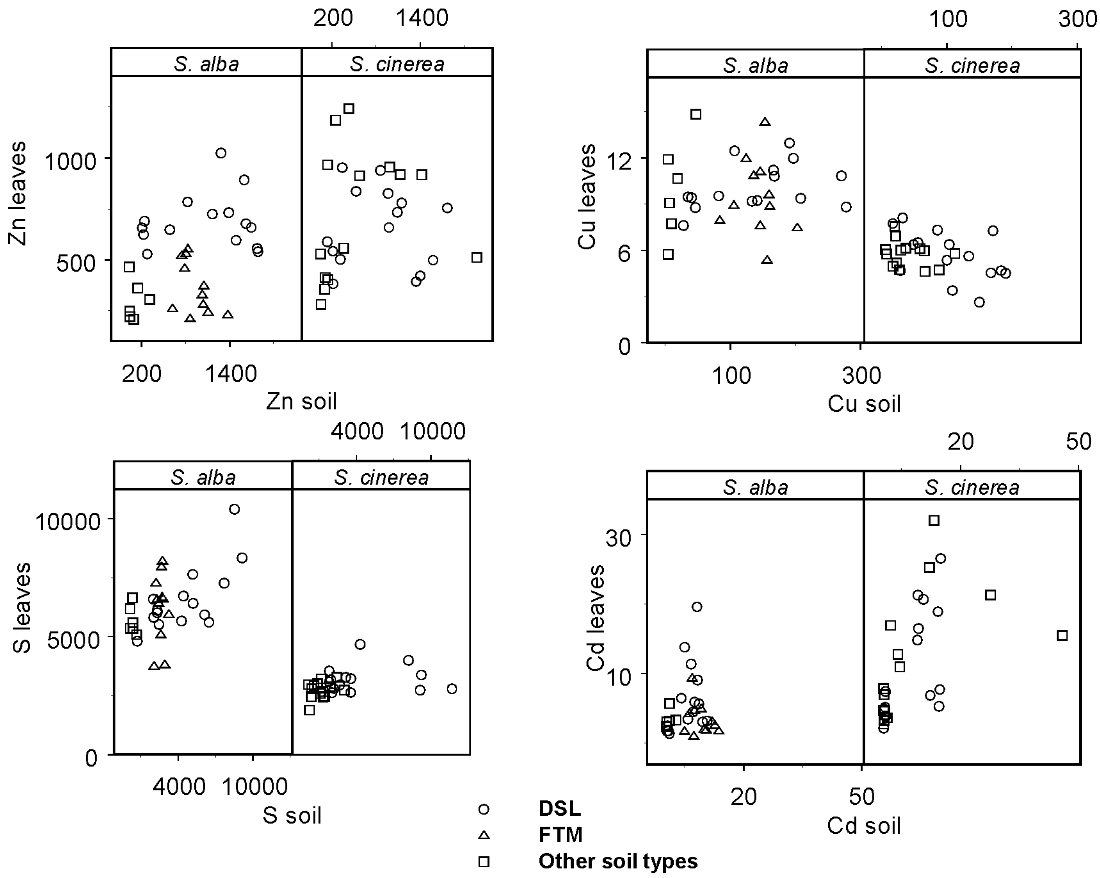

Fig. 3.5. Foliar Cd, Cu, Zn and S concentrations (mg kg^{-1} DW) in relation to soil concentrations (mg kg^{-1} dry soil) for dredged sediment landfills (circles), freshwater tidal marshes (triangles) and other soils (squares). Values are averaged per plot.

4.2. Site effects on foliar uptake for *S. cinerea* and *S. alba*

For both *S. cinerea* and *S. alba*, foliar Cu concentrations (Table 3.4) were hardly influenced by Cu concentrations in the soil (Fig. 3.5, Table 3.3). However, a large difference in foliar concentration between both species was observed as a consequence of site characteristics. Nevertheless, values were substantially lower than foliar concentrations between 183 and 681 mg Cu kg^{-1} DW measured for *Salix* species near a Cu refinery site (Dickinson, 2000).

Otte and Wijte (1993) concluded that besides soil composition and trace element speciation, other habitat or site characteristics determine plant Cu and Zn concentration for *Urtica dioica*. In our study, we also observed some site effects for Cd and Zn bioavailability. Foliar Cd concentrations were significantly higher for the polluted DSL and the OSZ relative to the reference situations (Table 3.4). However, BCF for Cd was highest for the reference situation (Fig. 3.3). Foliar baseline values for Cd and Zn in willow leaves are characterised by high concentration factors (Vandecasteele et al., 2002c; Eriksson and Ledin, 1999; Nissen and Lepp, 1997). No difference in BCF for Cd was observed between the polluted DSL and the OSZ (Table 3.4). The soil pollution status of the upper horizon was comparable for OSZ, DSL1 and DSL2 (Table 3.3). However, both substrate types have a different morphology, history and genesis. Due to sedimentation, the pathway of pollution for OSZ results in pollution concentrated in the upper cm of the soil profile. The developed soil profile is gradually polluted from the top layer. In the DSL studied here, the new soil profile is established at once over a larger thickness (> 100 cm in the studied sites), with the soil-forming processes starting from a reduced sediment layer. DSL are thus considered to be new soils at the time of disposal. The undetectable influence of the thickness of the polluted sediment layer on foliar concentrations, observed through comparison of OSZ and DSL, was remarkable. For *S. cinerea*, foliar Cd concentrations and bioavailability were found to be independent from the thickness of the polluted soil horizon, which is an important conclusion for ecological risk assessment. Foliar Zn concentrations were only significantly higher for OSZ compared to BAS. BCF(Zn) for OSZ was higher than for DSL2 and DSL1, while soil Zn concentrations for the three sites were comparable (Table 3.4).

A second effect, previously defined as species effect, is also a site effect, as volunteer *Salix* species occur when typical soil conditions prevail. These soil conditions (e.g. oxidation-reduction potential) are reflected in the foliar composition. A third site effect was found for *S. alba* with a distinct difference in Cd uptake for DSL and FTM (Table 3.4) while both soil types had a comparable soil pollution status for Cd. The differences observed in the tree diameter and hydrological regime, and the Zn concentrations in the soil and subsequently the difference in soil Cd:Zn ratio, might be the reason for the difference in foliar Cd concentration between DSL and FTM.

Otte (2001) argued that wetland plants are strongly adapted to flooded and anaerobic substrate situations, and that they are only stressed when conditions outside the usual range prevail e.g. conditions of environmental pollution. We found no evidence of acute toxicity for willows on the sampled landfills.

Future research should focus on the relation between hydrology (and oxidation-reduction potential) of polluted sediment-derived soils and metal uptake by willows, as one might expect a lower bioavailability in reduced soils.

4.3. Ecological risk assessment

Selecting adequate indicators for ecological risk assessment of metal contamination is not easy since potential responses or indicators are acute toxicity, growth reduction, reduced reproduction or elevated concentrations in willows, leaf-feeding insects or litter-dwelling organisms.

Punshon and Dickinson (1997a) for willow cuttings grown in solution cultures with high Cd, Zn and/or Cu concentrations and Vervaeke and Lust (2001) for *S. fragilis* on a dredged sediment landfill found foliar concentrations over 80 mg Cd kg^{-1} DW. These concentrations did not cause any visible signs of phytotoxicity. The highest foliar Cd concentration in this study was 72 mg kg^{-1} DW measured for a *S. alba* on the D7 site. Lunácková et al. (2003) reported a negative effect of Cd on leaf expansion of several *Salix* and *Populus* spp. Reduced leaf area for *S. cinerea* was not observed as an effect of pollution (Vandecasteele et al., 2004c). Morphological toxicity symptoms (reduced shoot length and leaf yield) for *Zea mays* were observed at foliar concentrations of 123 and 73 mg Cd kg^{-1} DW in the third and fourth leaf (Lagriffoul et al., 1998). No reference data about toxic foliar concentrations for willow species were found, but most authors refer to general toxic plant concentration ranges given by Kabata-Pendias (2002), who states that critical ranges in plants are between 100 and 400 mg kg^{-1} DW for Zn and between 5 and 30 mg kg^{-1} DW for Cd. Eriksson and Ledin (1999) reported baseline Cd concentrations in leaf samples between 0.31 and 1.96 mg kg^{-1} DW for *S. viminalis* on different non-polluted soils in Sweden. Nissen and Lepp (1997) found Zn concentrations in several willow species in the UK ranging between 82 and 296 mg Zn kg^{-1} DW, while a baseline range of 0.5-2.9 mg Cd kg^{-1} DW and 128-338 mg Zn kg^{-1} DW was reported by Vandecasteele et al. (2002c). Severson et al. (1992) reported baseline concentrations for *Salix repens* on the remote Frisian islands in Germany of <0.9-3.8 mg Cd kg^{-1} DW and 130-480 mg Zn kg^{-1} DW. Foliar concentrations are important indicators for Cd, Zn and Cu in site-specific ecological risk assessment, since these data are also indicative for food web transfer of metals.

The large differences in baseline concentrations and uptake patterns between plant species complicate a straightforward definition of general normal and toxic plant concentrations. Data in Table 3.4 illustrate the subjectivity of choosing reference values for evaluation of foliar concentrations. We propose that foliar data for uncontaminated dredged sediment landfills (DSL7 in this study) can be used for site-specific ecological risk assessment, as these sites are showing the background contamination of a highly urbanised area and have similar site characteristics as the polluted DSL. By choosing these sites as a reference, we avoid the application of unrealistically strict baseline data.

5. Conclusions

Foliar concentrations for several volunteer willow species were compared for sediment-derived soils and uncontaminated soils and a distinction was made between species effects and site effects on element concentrations. The site effect was important for Zn and Cd only, while other elements were mainly species-specific. Over a range of soil types, distinct differences in foliar Mn, Cu, S and K concentrations were observed between *S. alba* and *S. cinerea*.

Relative uptake patterns throughout the growing season were similar for Cd, Zn and Mn for all species except *S. cinerea*, for which Zn uptake was markedly slower early in the growing season. On polluted dredged sediment landfills, Cd and Zn foliar concentrations increased during the growing season until autumn.

Results illustrated that the selection of appropriate reference values for evaluation of foliar concentrations on polluted sites is species-specific. Foliar concentrations on polluted dredged sediment landfills were elevated compared with an uncontaminated dredged sediment landfill. On the other hand, foliar Cd and Zn concentrations measured on “man-made” polluted dredged sediment landfills were as high as values measured for “naturally” polluted overbank sedimentation zones. Soil genesis and thickness of the polluted horizon were not found to affect foliar concentrations for Cd and Zn.

Chapter 3.2. The effect of hydrological regime on the bioavailability of Cd, Mn and Zn for the wetland plant species *Salix cinerea*

The hydrological conditions on a site constitute one of the many factors that may affect the availability of potentially toxic trace metals for uptake by plants. Bioavailability of Cd, Mn and Zn in a contaminated dredged sediment-derived soil under different hydrological regimes was determined by measuring metal uptake by the wetland plant species *Salix cinerea*, both in field circumstances and in a greenhouse experiment. Longer submersion periods in the field caused lower Cd concentrations in the leaves and the bark. The wetland hydrological regime in the greenhouse experiment resulted in normal Cd and Zn concentrations in the leaves, and normal Cd and high Mn concentrations in the cuttings, while the upland hydrological regime caused elevated Cd and Zn concentrations in the leaves and elevated Cd concentrations in the cuttings. Foliar concentrations were strongly influenced by the initial concentrations in the cuttings because of translocation. In general, the results from the greenhouse experiment clearly demonstrate the importance of analysing cuttings prior to the initiation of a lab test. Field observations and the greenhouse experiment suggest that a hydrological regime aiming at wetland creation is a potential management option that favours reducing metal bioavailability for plants. This would constitute a safe management option of metal-polluted, willow-dominated wetlands provided wetland conditions can be maintained throughout the full growing season.

1. Introduction

Metal availability for plants on polluted sites is of concern: elevated concentrations within the first trophic level in food webs, e.g. plants, may cause higher body concentrations in herbivorous insects (Crawford et al., 1996; Vandecasteele et al., 2003b; Merrington et al., 2001) birds (Pedersen and Saether, 1999; Świergosz and Kowalska, 2000) and mammals (Mertens et al., 2001; Lodenius, 2002), and may indirectly influence litter dwelling organisms (Scheifler et al., 2002) due to higher metal concentrations in the litter layer.

Willows occur as volunteer vegetation on sediment-derived soils such as dredged sediment landfills, overbank sedimentation zones and freshwater tidal marshes (Bal et al., 2001). They have a potential to be used for biomonitoring on contaminated sediment-derived soils, since foliar samples provide an indication of metal bioavailability (Vandecasteele et al., 2002c). However, metal concentrations in willow are strongly dependent on species (Lunáčková et al., 2003; Vandecasteele et al., 2004c), clone (Landberg and Greger, 2002), individuality due to phenotypic plasticity (Landberg and Greger, 1996; Punshon and Dickinson, 1997a), gender (Ernst, 1990), part of the willow sampled (Punshon and Dickinson, 1997a), growth performance (Klang-Westin and Perttu, 2002), root density and distribution over the soil profile (Keller et al., 2003) and sampling period (Vandecasteele et al., 2004c).

Plant bioavailability is a function of physicochemical soil properties, hydrology and oxidation-reduction (redox) potential. Flooding of wetland soils affects metal solubility and immobilisation, and leads to a lower environmental availability of Cd, Cu, Zn and Ni (Gambrell, 1994; Kashem and Singh, 2001). Soil flooding results in oxidation-reduction potential (Eh) lowering and consequently an increasing oxygen demand for both soil and plant roots (Pezeshki, 2001) leading to stressed roots. A detrimental effect of flooding on plants may be the production or accumulation of phytotoxic compounds (e.g. organic acids, methane, sulfides, reduced Mn and Fe) contributing to root injury, growth reduction and mortality (Kozlowski, 1997; Pezeshki, 2001). Eh also affects plant availability of essential elements and contaminants. Reduced soils were found to have lower Cd and Zn uptake in several plant species (Gambrell and Patrick, 1989; Gambrell, 1994). Stoltz and Greger (2002a) proposed to grow wetland plant species on submerged mine tailings to reduce oxygen concentrations in the topsoil and immobilise metals.

The aim of this study was to determine whether the hydrological circumstances, in particular the duration and intensity of submersion, of a polluted dredged sediment-derived soil (DSDS) affect the foliar and bark metal concentration for *S. cinerea*, a typical wetland willow species. The advantage of willows for research on metal bioavailability is that they reflect time-integrated accumulated concentrations for the studied soil profiles. Results of field-collected foliar and bark samples were confronted with the results of a greenhouse experiment with willow cuttings and soil samples collected on the same sites. In that experiment, the effect of submersion and metal translocation from the cutting on foliar metal uptake was screened.

2. Materials and methods

2.1. Field study

2.1.1. Soil sampling and measurements

Metal concentrations in volunteer willow vegetation growing on three plots (Table 3.5) were compared. Plot1 and plot2 were located on a dredged sediment landfill near Deinze (Belgium), referred here as dredged sediment-derived soil (DSDS). The DSDS (12.5 ha) was landfilled between 1976 and 1983 with sediments dredged in the river Leie near Deinze, and can be characterised as a freshwater wetland with stagnant water during late fall, winter and spring. The two plots were selected representing two hydrological regimes: plot1 was waterlogged during winter but water level decreased strongly in spring, while plot2 was regularly waterlogged in summer as well, depending on the weather conditions. The volunteer willow vegetation on this polluted DSDS site was compared with volunteer willows on a nearby infrastructure spoil landfill with baseline metal concentration levels (Table 3.5, plot3). On each plot, the 0-30 soil horizon was sampled in triplicate. Plot3 was selected due to the occurrence of *S. cinerea*, and the comparable soil genesis and characteristics. Both the DSDS and the infrastructure spoil landfill were established by hydraulic filling. For the 3 plots studied here, the new soil profile was established at once over a larger thickness (> 100 cm) , and are thus considered to be new soils at the time of filling. All plots are calcareous clayey-textured soils (Table 3.5).

Time is expressed as the week number during the year. Redox potential was weekly measured between week 15 (last week of March) and week 24 (second week of June) with 4 replicates on all plots with a combined redox electrode (platina and Ag/AgCl reference electrode; HANNA instruments, HI 3131B) and a WTW multiline P3 meter, and values were reported after conversion towards the standard hydrogen electrode. The combined electrode was pushed into the soil to a depth of 5 cm and a stable reading was awaited. After measurement, the resulting hole was gently refilled with the surrounding soil. Redox measurements for plot3 was not possible from week 22 (last week of May) onwards as the electrode could not penetrate the soil as the soil was too dry. Simultaneously with the redox measurements, water level above the soil surface for the waterlogged soils was measured.

2.1.2. Foliar and bark sampling

On each plot, 4 *Salix cinerea* L. trees of approximately the same age and diameter were sampled. Our sampling strategy for leaves and bark focused on individual trees. For foliar sampling, at least 4 branches from different heights and positions in the crown were sampled. To account for the variability associated with sampling, 4 different trees of approximately the same age and dimensions were sampled individually within a circle with a diameter of 10 m. Foliar samples were taken with a three week interval between week 18 (last week of April) and week 24 (second week of June). Samples were collected by means of an extension crosscut saw (Blair, 1995). Approximately 1000 cm^3 of leaves were collected at each sampling location. Leaves were not washed as washing procedures seem to produce misleading results due to incomplete removal of metals on the leaf surface and partial leaching of metals from the leaf tissues (Kozlov et al., 2000). The plots are not under direct influence of point sources of aerial metal emission, but background metal concentrations in the air are relatively high in this industrialised an urbanised region. The effect of the plot on the foliar concentration was tested with one-way ANOVA after aggregation of the foliar data per tree. Multiple comparison was executed with the Sidak method.

Due to new landfilling activities, vegetation at the DSDS had to be removed. Therefore, trees previously used for the foliar samplings were sawn down in week 25 (third week of June) and the bark, bast (inner bark) and cambium, further termed 'bark', was sampled. *S. cinerea* is a tall shrubby tree with branching close to the soil surface. For each tree, 3 large branches were sampled at a height of 1 and 2 m, where a 30 cm long piece of wood was removed. The diameter of each piece was measured. The bark of each branch was sampled at 4 directions (north, east, south and west direction). Combination of 2 plots, 4 trees per plot, 3 branches per tree, two heights per branch and 4 directions resulted in 192 bark samples.

The aim of the bark samplings was to determine if differences in bark concentrations between plots with varying soil conditions are similar to the foliar differences found, accounting for several potential sources of variability. The factors 'plot', 'height' and 'direction' were recognised as fixed factors, while the factors 'tree' and 'branch' were random factors, being nested within the factor 'plot'. The ANOVA model used was [bark concentration ~ plot/tree/branch + plot * height * direction]. The plot-effect on the bark concentration was tested with one-way ANOVA after aggregation of the data per tree.

Table 3.5. Soil properties of the dredged sediment-derived soils (plot1 and plot2) and the plot on an uncontaminated infrastructure spoil landfill (plot3) where willow was sampled during the first weeks of the growing season (EC: electrical conductivity, OM: organic matter). Soil metal concentrations are *aqua regia* extractable concentrations (mg kg^{-1} dry soil). Values in parentheses denote standard deviations (3 replicates)

		plot1	plot2	plot3
clay	%	38.6 (1.7)	34.3 (3.8)	21.9 (6.4)
CaCO3	%	8.5 (1.6)	7.8 (0.6)	2.3 (1.2)
EC	µS cm^{-1}	628 (187)	618 (157)	140 (30)
pH-H2O		7.4 (0)	7.5 (0.1)	7.4 (0.5)
OM	%	8 (0.9)	7 (2)	3.5 (1.4)
Cu	mg kg^{-1} dry soil	171 (17)	150 (24)	17 (9)
Cr	mg kg^{-1} dry soil	292 (26)	259 (38)	57 (6)
Pb	mg kg^{-1} dry soil	258 (28)	221 (29)	26 (9)
Ni	mg kg^{-1} dry soil	64 (5)	53 (7)	24 (6)
Mn	mg kg^{-1} dry soil	407 (33)	384 (42)	1036 (590)
Zn	mg kg^{-1} dry soil	1766 (175)	1400 (253)	99 (18)
Cd	mg kg^{-1} dry soil	15 (1.4)	12.3 (2.2)	0.6 (0.1)

2.2. Greenhouse experiment

2.2.1. Experimental design

The greenhouse experiment was aimed to determine the effect on the foliar concentrations of two group of factors, soil hydrological conditions and pollution status (factor 'soil type') on the one hand and the origin of the tree cuttings (factor 'origin of the cutting') on the other. We also compared metal concentrations in the cuttings before and after the greenhouse trial to get insight into the metal translocation within the plant. Cuttings of 4 trees and soil material from plot1 and plot3 were used for a greenhouse experiment. Four trees were selected based on the level of foliar Cd concentrations measured in the field during the growing season 2002 (Table 3.6) with the methods described above. Two trees were situated on plot1 (cin1 and cin2), and two trees were situated on plot3 (cin3 and cin4), and on each plot, the tree with the highest and lowest Cd concentrations was selected. For each tree, willow cuttings from one-year-old dormant stems were collected in week 49 (first week of December 2002) and stored in the refrigerator.

Table 3.6. Cd, Mn and Zn concentrations (mg kg^{-1} DW) in the leaves during the growing season 2002 (between week 33 and week 42), and in the cuttings collected on the field in week 49 (December 2002). Means that are not significantly different are denoted with the same letter (Sidak multiple comparison of means at the 95% level of significance, n: number of replicates).

	cin1	cin2	cin3	cin4
Cd foliar conc. Week 33-42 2002 (n = 4)	57.1 (a)	21.0 (b)	4.0 (c)	2.0 (c)
Mn foliar conc. Week 33-42 2002 (n = 4)	335.3 (ab)	221.2 (b)	575.6 (a)	430.1 (ab)
Zn foliar conc. Week 33-42 2002 (n = 4)	1365.4 (a)	1018.0 (a)	382.7 (b)	291.6 (b)
Cd concentration in field-collected cuttings (n=5)	25.9 (a)	13.6 (b)	2.8 (c)	2.5 (c)
Mn concentration in field-collected cuttings (n=5)	45.9 (b)	37.0 (b)	90.8 (a)	69.6 (a)
Zn concentration in field-collected cuttings (n=5)	284.7 (a)	249.5 (a)	122 (b)	110.3 (b)

The 0-30 cm soil horizon was sampled on plot1 (contaminated DSDS) and plot3 (uncontaminated soil). Soil material was intensively mixed and homogenised and stones, plastics and branches were removed. Cuttings were 20 cm long and were planted in 13x13x13 cm containers with 1 kg of soil at field capacity. Two cuttings were planted in each container. All containers were placed in a greenhouse with regular irrigation. Each 'upland' replicate was provided with a dish underneath to avoid export of soil material or dissolved elements. For each type of cutting, the 8 'wetland' replicates were placed in a 60x40 cm container with a water level 2 cm above the soil level. Three soil types were thus established: 'uncontaminated upland soil', 'contaminated upland DSDS, and a 'contaminated wetland DSDS'. For each tree, 8 replicates per hydrological regime were used. In total, 96 containers were used in the trial. The greenhouse experiment started in week 13 (last week of March).

Redox potential in the 'contaminated wetland DSDS' was measured on a depth of 5 cm between week 16 (Third week of April) and week 26 (last week of June) with a 10-day interval with a combined platina and Ag/AgCl electrode (HANNA instruments, HI 3131B) and a WTW multiline P3 meter. Measured average redox potential for measurements with a

10-day interval (after conversion towards the standard hydrogen electrode) was 60±65, 12±58, 50±66, 8±44, 8±41, 4±53 and 60±64 mV. After measurement, the resulting hole was gently refilled with the surrounding soil. Redox potential measurements in the upland treatments was not possible as the electrode could not penetrate the soil.

For each tree, 5 additional cuttings were analysed at the start of the experiment. Concentrations in the cuttings were compared with one-way ANOVA for the fixed factor 'origin of the cutting', and multiple comparison was executed with the Sidak method.

2.2.2. Sampling

Leaves were sampled the first time in week 26 (91 days after planting the cuttings). The youngest 5 leaves were systematically not sampled as they were generally not yet full-grown. For each container, the amount of sampled leaves and the total shoot length per cutting were determined as growth parameters.

Regrowth after the first harvest was only sufficient for the 'contaminated wetland DSDS' replicates to allow for a second foliar harvest in week 36 (first week of August). New stems for the 'wetland' replicates were sampled separately. At the end of the experiment, cuttings were removed from all replicates, and stems and roots were removed and soil particles were washed from the cuttings.

Two-way ANOVA was used for comparing growth parameters and both foliar (first harvest) and cutting concentrations with 'soil type' and 'origin of the cutting' as fixed factors. Cd concentrations in cuttings and leaves and Mn and Fe concentrations in cuttings were square root-transformed prior to analysis. Multiple comparison was performed with the Sidak method (0.95 confidence level).

2.3. Chemical analyses

Bark, foliar and cutting samples were dried for 7 days at 40 °C, mechanically ground (Pulverisette 14, Fritsch, Idar-Oberstein, Germany), and stored in dark vials before analysis. Total foliar and bark N was measured by the Kjeldahl method (Van Ranst et al., 1999). Total foliar, bark and cutting element concentrations are extracted with HNO_3 (p.a. 65%) and H_2O_2 (ultrapur) in a 3:1 ratio using microwave digestion and measured with ICP-AES (Varian Liberty Series II, Varian, Palo Alto, CA). The accuracy of the foliar and bark element analysis was checked using BCR 60 (Aquatic plant) for Cd, Cu, Mn and Zn, and CRM 100 (Beech leaves) for Ca, Mg, Na, K, S and P. Soil total concentrations of Cd, Cr, Cu, Ni, Pb, S, Mn, P and Zn are actually pseudo-total *aqua regia* extractable concentrations measured with ICP-AES after microwave digestion. Methods are described in Chapter 1.1.

3. Results

3.1. Field study

Plot3 was not found to be submerged in week 15, while plot2 was submerged until the end of the field experiment. Plot1 was submerged until week 21 (Fig. 3.6). Redox potential values in the topsoil were lower than 60 mV for both plot1 and plot2, while the soil became aerobic (Eh exceeding 350-400 mV) in week 20 for plot3. For plot2, a distinct sulphide odour was perceived in the first weeks of observation.

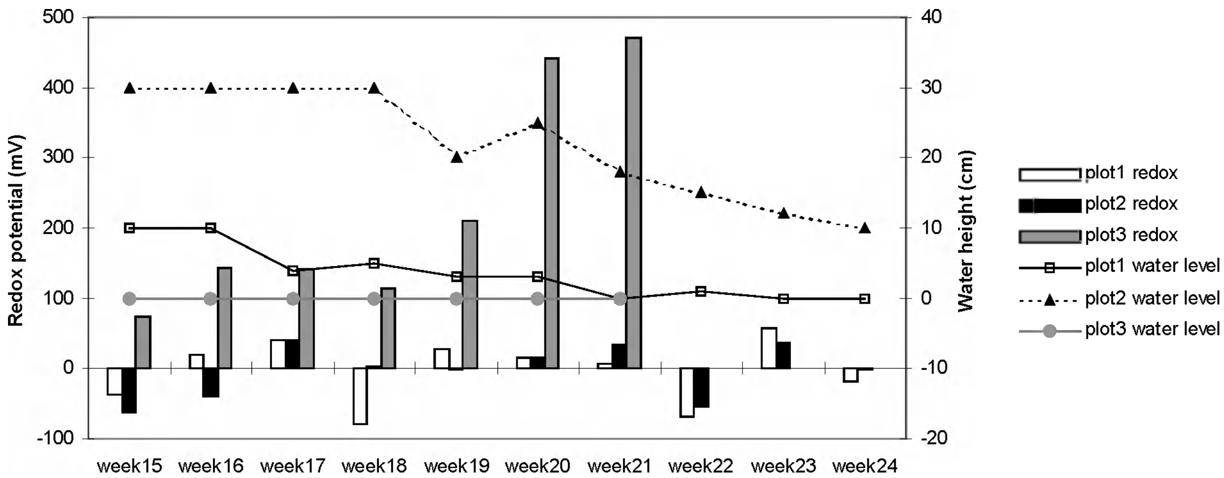

Fig. 3.6. Evolution of the weekly measured soil redox potential on a depth of 5 cm and the water height on the soil surface for the 3 sampled plots

Foliar concentrations are shown in Fig. 3.7. Differences were highly significant for Cd and Mn ($p < 0.01$) and significant for Cu, S and Fe ($0.01 < p < 0.05$). Multiple comparison demonstrated that Cd, Cu, S and Fe concentrations were significantly higher for plot1 than for the plot3 and/or plot2. Foliar Mn concentrations are higher for plot3 than for the plot1 and plot2.

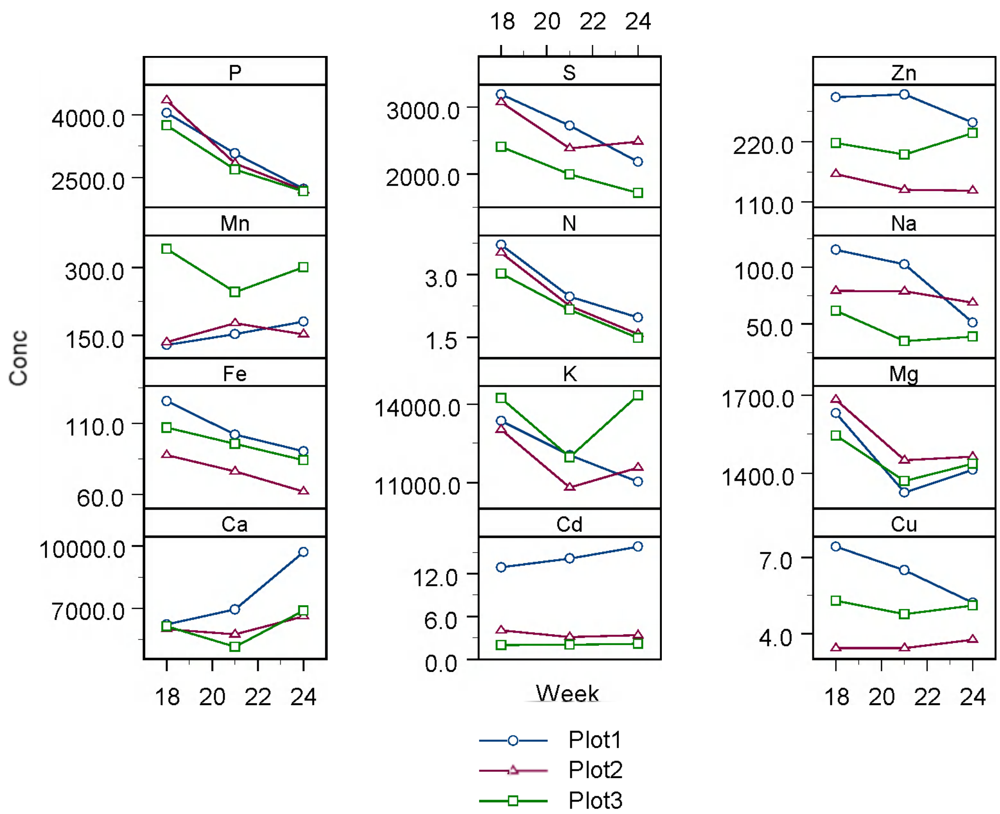

Fig. 3.7. Average foliar accumulation patterns for Cd, Zn, Cu, Mn, K, Ca, Fe, N, S, P, Mg and N during the start of the growing season (week 18 to week 24) for *S. cinerea* (4 trees per plot) on a dredged sediment-derived soil (plot1: circles, plot2: triangles) and an uncontaminated infrastructure spoil landfill (plot3: squares). N is expressed as %, other elements are expressed as mg kg^{-1} DW.

The ANOVA results indicate no or limited interaction between the fixed factors ‘plot’, ‘height’ and’ direction’ for the bark concentrations. Direction has low influence on the bark concentration, while sampling height resulted in statistically significant differences but the effects are small and hence without practical significance. Measures of scatter for the random factors ‘branch’ and ‘tree’ within a single branch, within branches of a tree, and within the trees of a plot are summarised in Table 3.7. The plot-effect tested on the bark data aggregated per tree only resulted in a significant difference for Cd ($p = 0.014$). Bark Cd concentrations were higher for plot1 than for plot2. For the other elements, the differences in bark concentrations between the plots were small or were not significant due to the large scatter of the data. Bark Cd concentrations aggregated per height are displayed in Fig. 3.8. Irrespective of the large scatter within and between trees per plot, there is a substantial difference in Cd concentration between the trees of plot1 and plot2. When Cd concentrations in the bark averaged per tree were compared with foliar concentrations in week 24, higher bark concentrations were associated with higher foliar concentrations (correlation coefficient r = 0.87). The positive correlation between stem or bark concentrations and foliar concentrations were reported before by Punshon and Dickinson (1997a) and Klang-Westin and Eriksson (2003).

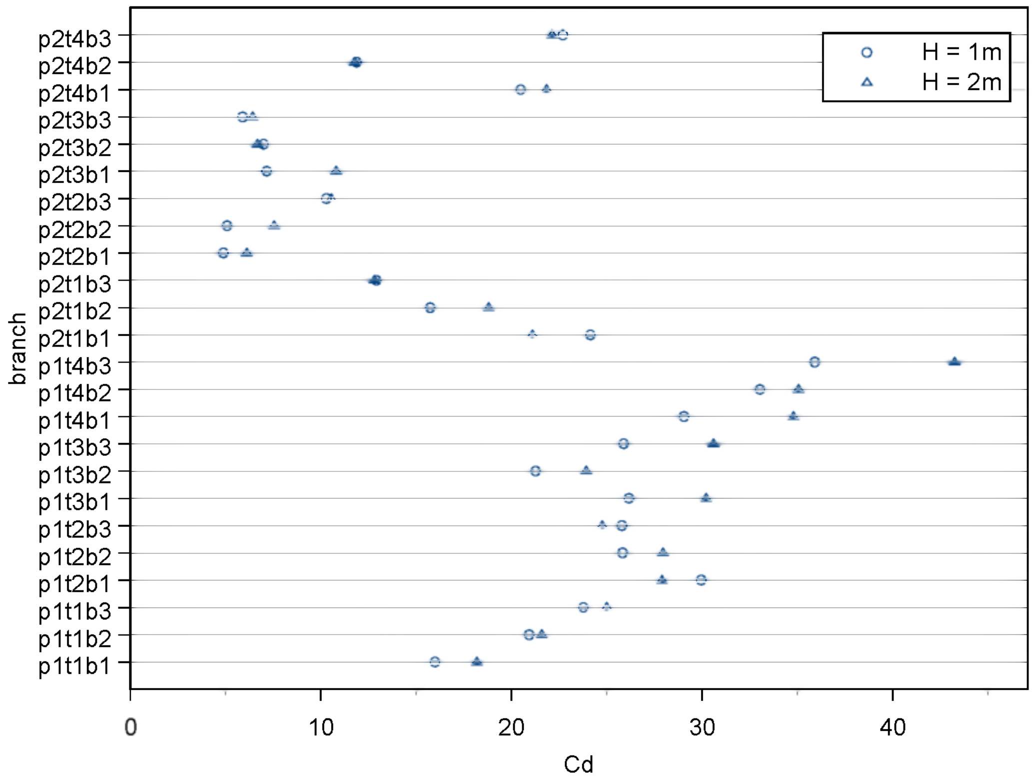

Fig. 3.8. Bark Cd concentrations (mg kg^{-1} DW) for 8 trees on 2 plots. Three branches per tree (b = branch, t = tree, p = plot) were sampled on two heights (circles = 1m, triangles = 2m, P1 = plot1, P2 = plot2).

Table 3.7. The average bark concentration (mg kg^{-1} DW) for 2 plots on a DSDS, and a measure of scatter (square root of variance component) in the bark concentrations for noise (r_{noise} = scatter within a branch), branch (s_{branch} = scatter within the branches of one tree) tree (s_{tree} = scatter within the trees of one plot) and plot (s_{plot} = scatter between the plots) (nc = not calculated due to a negative value for the estimated variance).

	Plot1	Plot2	s_{noise}	s_{branch}	s_{tree}	s_{plot}
Cd	27.4	12.7	2.0	3.4	5.6	9.9
Zn	426.0	338.3	24.5	27.5	95.9	38.2
Cu	3.0	2.9	0.7	nc	0.5	nc
P	700.2	685.7	82.2	nc	71.5	nc
S	818.3	845.8	86.6	60.2	133.5	nc
Ca	22812	21904	1941	1125	3373	nc
K	3415	2966	789	nc	400	223
Mg	570.0	589.2	86.5	32.6	71.0	nc
Na	122.4	127.9	55.3	nc	59.9	nc
Fe	71.5	68.6	16.4	12.7	15.7	nc
Mn	60.5	67.4	10.1	nc	11.5	nc
N	4744	6291	1024	248	780	998

Table 3.8. Average concentration (mg kg^{-1} DW) in leaves (first harvest, week 26) and cuttings (second harvest, week 36) of the greenhouse experiment as a function of 'origin of cutting' and 'soil type'. No interaction was observed between both factors for the presented results. Means that are not significantly different are denoted with the same letter (Sidak multiple comparison of means at the 95% level of significance).

	Origin of the cutting				**Soil type**		
	cin1	**cin2**	**cin3**	**cin 4**	**Upland DSDS**	**Wetland DSDS**	**Upland reference soil**
Foliar Cd	22.3 (a)	11.6 (b)	3.6 (c)	3.7 (c)	15.0 (a)	9.0 (b)	7.8 (b)
Foliar Zn	363.9 (a)	322.2 (ab)	197.3 (b)	256.2 (c)	390.6 (a)	210.9 (b)	264.9 (c)
Cd in cuttings	14.8 (a)	10.8 (b)	4.6 (c)	3.5 (d)	12.7 (a)	6.5 (b)	5.9 (b)
P in cuttings	804 (a)	614 (c)	669 (b)	558 (c)	625 (b)	769 (a)	584 (b)
Mn in cuttings	23.9 (c)	29.9 (c)	49.6 (a)	40.1 (b)	27.6 (b)	47.2 (a)	33.1 (b)
Ca in cuttings	8272 (a)	5748 (c)	8147 (a)	6773 (b)	6703 (b)	7761 (a)	7319 (ab)
Mg in cuttings	376.9 (a)	394.4 (a)	408.1 (a)	381.3 (a)	353.1 (b)	404.5 (a)	413.8 (a)
K in cuttings	2559 (a)	2140 (b)	2278 (ab)	2405 (ab)	2506 (a)	1954 (b)	2605 (a)
S in cuttings	349.6 (a)	373.0 (a)	362.0 (a)	305.2 (b)	325.1 (b)	365.9 (a)	350.1 (ab)
Fe in cuttings	271.9 (a)	298.9 (a)	213.8 (ab)	174.8 (b)	78.5 (b)	549.5 (a)	66.3 (b)

3.2. Greenhouse experiment

No significant concentration differences in cuttings collected in the field were found for Ca, Cu, Fe, Mg, Na and S. Highly significant ($p < 0.01$) differences were found for Cd, Mn and Zn (results summarised in Table 3.6). Significant differences ($p < 0.05$) were found for K (cin1 > cin3) and for P (cin1 > cin3). Average concentrations for Ca, Cu, Fe, Mg, Na, K, P and S are 6630, 5.3, 189, 504, 46.3, 2638, 1040 and 568 mg kg^{-1} DW. Concentrations in the cuttings were slightly lower than foliar concentrations at the end of the growing season for Cd, and clearly lower for Mn and Zn (Table 3.6). The concentration sequence for these elements was identical for cuttings and leaves (Table 3.6).

For the growth parameters no interaction between the factors 'soil type' and 'origin of the cutting' was detected with ANOVA. Results for the growth parameter 'amount of sampled leaves' were only significantly influenced by the origin of the cutting, with slightly lower amounts (30% lower) for cin1 compared with cin3 and cin4. The growth parameter 'total shoot length per cutting' was only significantly influenced by the soil type, with clearly higher shoot length for the 'contaminated wetland DSDS' treatment, being on the average 40% higher than for the other treatments. Similar observations were reported by Hansen and Phipps (1983) for a growth room study with *Populus* cuttings. In contrast to results of Robinson et al. (2000) for a greenhouse experiment with poplar and willow clones and Klang-Westin and Perttu (2002) for willow stem Cd concentrations in a lysimeter experiment, no distinct growth differences and thus differences in dilution by growth were observed during the period of the experiment.

No interaction between the factors 'soil type' and 'origin of the cutting' was observed for foliar Cd and Zn concentrations (first harvest), while interaction was significant for Fe, Mg, Na ($p < 0.05$) and highly significant for Cu, Ca, K, Mn, P, S ($p < 0.01$). These results indicate that, with exception for Cd and Zn, foliar accumulation patterns for different soil types are depending on the cutting. Interaction plots confirmed the differences in accumulation patterns between cuttings (interaction plots not shown). For these elements, no straightforward conclusions can be drawn.

Highest foliar Cd and Zn concentrations at first harvest were measured in the upland contaminated DSDS treatment, while for Cd no difference was observed between the 'contaminated wetland DSDS' and the 'uncontaminated upland soil' (Table 3.8). The sequence in foliar Cd concentrations for the cuttings used in the greenhouse experiment is identical to the concentration sequence found in the field-collected cuttings and in the field-collected leaves. Moreover, the measured concentrations in the leaves (first harvest) of the greenhouse experiment and the field collected cuttings are similar (Table 3.6, Table 3.9).

Table 3.9. Cd, Mn and Zn concentrations (mg kg^{-1} DW) in the leaves in week 26 (first harvest) and week 36 (second harvest), and in the stems (week 36, second harvest) of the greenhouse experiment. Means that are not significantly different are denoted with the same letter (Sidak multiple comparison of means at the 95% level of significance)

	cin1	cin2	cin3	cin4
Cd foliar conc. first harvest, wetland DSDS	20.5 (a)	8.9 (b)	3.3 (c)	3.0 (c)
Mn foliar conc. first harvest, wetland DSDS	124.4 (b)	120.9 (b)	267.0 (a)	166.2 (b)
Zn foliar conc. first harvest, wetland DSDS	265.1 (a)	223.4 (ac)	157.4 (b)	206.6 (bc)
Cd foliar conc. second harvest, wetland DSDS	13.6 (a)	5.5 (b)	4.1 (b)	5.8 (b)
Mn foliar conc. second harvest, wetland DSDS	177.7 (b)	156.9 (b)	294.9 (a)	317.5 (a)
Zn foliar conc. second harvest, wetland DSDS	259.8 (ab)	191.1 (b)	206.4 (b)	340.9 (a)
Cd conc. in stems, wetland DSDS	6.4 (a)	4.0 (b)	4.1 (b)	4.4 (b)
Mn conc. in stems, wetland DSDS	38.5 (b)	33.6 (b)	74.3 (a)	85.0 (a)
Zn conc. in stems, wetland DSDS	83.4 (ab)	53.2 (b)	66.8 (b)	105.0 (a)

Foliar Cd and Zn concentrations for cin1 and cin2 in the second harvest for the 'contaminated wetland DSDS' were lower than concentrations in the first harvest, and higher for cin3 and cin4, while Mn concentrations were higher in the second harvest for all cuttings (Table 3.9). Stem concentrations for Cd, Zn and Mn were lower than foliar concentrations (Table 3.9).

No interaction between the factors 'soil type' and 'origin of the cutting' was observed for Cd, Mn, Fe, Ca, K and P concentrations in the cuttings at the end of the greenhouse experiment, while both factors were significantly ($p < 0.001$) influencing the cutting concentrations for these elements. For Mg, only 'soil type' had a highly significant ($p < 0.001$) effect on concentrations in the cuttings, while for S only the 'origin of the cutting' had a highly significant effect. Results are shown in Table 3.8. For Na, Zn and Cu interaction between both factors was significant. For Cu, no significant differences were found between replicates. For Zn, the replicates of cin1 in the 'uncontaminated upland soil' showed clearly higher concentrations than the other replicates (488 vs. 118-281 mg Zn kg^{-1} DW). The overall average Cu concentration in the cuttings was 4.0 mg kg^{-1} DW. Foliar concentrations for K, P, Mn and Mg are substantially higher than in the cuttings.

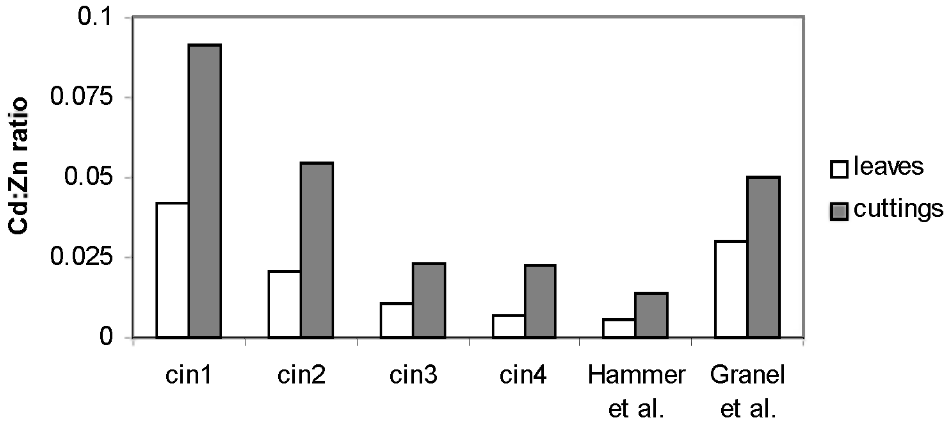

Fig. 3.9. Cd:Zn ratio in leaves and cuttings for the trees used in the greenhouse experiment, and Cd:Zn ratios in leaves and stems calculated from data of Granel et al. (2002) and Hammer et al. (2003).

4. Discussion

4.1. Field study

4.1.1. Effect of submersion

For plot1 with a shorter submersion period (Fig. 3.6.) foliar concentrations for Cd, Cu, S and Fe were highest (Fig. 3.7.). The mobility and availability of Cd, Cu and S is higher in oxidising conditions (Gambrell, 1994). Higher Fe availability for plot1 might be caused by free Fe^{2+} in the soil solution while Fe at plot2 might be immobilised as FeS in more reduced soil conditions. In the first weeks of the growing season, foliar Cd concentrations on plot1 were markedly higher than measured concentrations on sites with baseline contamination levels (Vandecasteele et al., 2004c), while values for plot2 and plot3 were in the normal range. Only for Cd a distinct difference in bark concentrations was observed (Fig. 3.8., Table 3.7), with the highest concentrations for plot1, the plot with the shorter submersion period.

4.1.2. Bark sampling

Cu concentrations in the bark (Table 3.7) are comparable with data from Nissen and Lepp (1997) for several willow species, while Zn concentrations are twice as high or more. The scatter of metal concentrations in the bark between branches of a tree and between trees of a plot is rather large (Table 3.7). Guidelines for sampling willow bark are that (a) several branches within a tree must be sampled, (b) wind direction is not an important factor an thus, bark rings can be collected as an alternative, and (c) at least 2 heights must be sampled, and the standardisation for height can be expressed both absolutely (e.g. 1 and 2m) or relatively (1/3 and 2/3 of the branch length).

4.1.3. Bark samples for biomonitoring

Results demonstrate that the use of willow bark for biomonitoring soil pollution is facing some limitations: the tree individuality is that large (Table 3.7) that this factor must be kept constant when constructing time series of bark concentrations. The destructive sampling is a disadvantage of *in situ* biomonitoring using willow bark.

4.2. Greenhouse experiment

4.2.1. Effect of origin of the cutting

Both the soil type and the origin of the cutting had a significant effect on the foliar concentrations for Cd and Zn (Table 3.8). We must however realise that the effect of the cutting is a kind of delayed soil effect. Results of Punshon and Dickinson (1997a) in a 128-day greenhouse experiment with 4 willow clones clearly demonstrated higher Cd and Zn concentrations in roots, cuttings, new stems and leaves after a cumulative exposure to increasing metal concentrations relative to control treatments. The original cutting concentration in our study (Table 3.6) was determined by the soil pollution status of the plot where the cuttings were collected, since higher soil concentrations result in higher foliar and bark concentrations.

Foliar metal concentrations in the field are influenced by the previous growing seasons due to translocation of elements from stems to leaves. Landberg and Greger (1994) found a significantly positive correlation between initial Cd concentrations in 103 field-collected cuttings of diverse *Salix* species and Cd concentrations in the shoots after hydroponic screening for 20 days in a 1 μM Cd solution. Foliar concentrations in the greenhouse experiment were thus partly determined by translocation of Cd and Zn from cuttings to leaves. This is illustrated by the results of the first and second harvest of the 'contaminated wetland DSDS' treatment, with lower Cd concentrations for cin1 and cin2 in the second harvest than in the first harvest (Table 3.9), indicating lower bioavailability in wetland soils and reduced translocation from the cutting to the leaves. In general, the results from the greenhouse experiment clearly demonstrate the importance of analysing cuttings prior to the initiation of a lab test.

4.2.2. Effect of submersion on plant uptake

Apart from the distinct effect of the origin of the cutting, the soil type largely affects foliar and cutting concentrations. Analysis of the cuttings after our greenhouse experiment revealed clearly higher Mn, P and Fe concentrations for the 'contaminated wetland DSDS' treatment, and clearly higher Cd concentrations for the 'contaminated upland DSDS' treatment (Table 3.8). The latter observation is similar to the pattern observed for Cd in the leaves (Table 3.8). Reduced soil conditions thus result in a lower Cd availability for cuttings and leaves. Higher P, Mn and Fe concentrations in the cuttings for the 'contaminated wetland DSDS' treatment (Table 3.8) point at the higher availability of these elements at low Eh (Gambrell, 1994; Satawathananont et al., 1991), without formation of FeS. Flooding of well-aerated upland soils resulted in a fast reduction of Mn^{4+} and Fe^{3+} into soluble Mn^{2+} and Fe^{2+} as soon as the soils were incubated under anoxic conditions (Peters and Conrad, 1996). In an experiment with a range of overfertilised agricultural soils under reducing conditions, solution concentrations of Fe and Mn and associated P strongly increased during soil reduction phases with complete oxygen depletion and Mn reduction (Scalenghe et al., 2002). Rewetting of a wetland soil after a dry period resulted in an increased extractable P pool (Olde Venterink et al., 2002). Cuttings from the 'contaminated wetland DSDS' are characterised by significantly lower K concentrations than the other treatments. In field-collected foliar samples, *S. cinerea* growing on the submerged dredged sediment-derived sites had clearly lower foliar K concentrations than other *Salix* species (Vandecasteele et al., 2004c). An important conclusion is that analysis of cuttings might be an alternative for foliar analysis in greenhouse experiments.

4.2.3. Duration of the submersion

The presented results from field samplings and the greenhouse experiment confirm the hypothesis that bioavailability of metals and especially Cd is reduced for willows when wetland conditions are stimulated. However, in previous field studies on contaminated DSDS, it was observed that wetland hydrological regimes were only present in the winter and the early growing season, and drier soil conditions at the end of the growing season resulted in elevated foliar Cd and Zn concentrations (Vandecasteele et al., 2004c). Results of the field samplings (Fig. 3.7.) and the greenhouse experiment (Table 3.9) demonstrate that especially Cd is strongly translocated from cuttings to leaves in the early growing season, while the results of the cuttings collected in the field on contaminated DSDS at the end of the growing season (Table 3.6) contained high Cd levels and point at high bioavailability of Cd at this point in time. Cd:Zn ratio in cuttings and stems is twice as high as Cd:Zn ratio in the leaves (Fig. 3.9), illustrating the differences between Cd and Zn in translocation behaviour from stems to leaves. Distinct differences in Cd:Zn ratios between stem and leaves were also illustrated by data from Granel et al. (2002) and Hammer et al. (2003). Hydrological regime aiming at freshwater wetland creation is a potential management option for reducing Cd bioavailability for plants and thus for establishing a safe management of willow-dominated wetlands polluted with metals as long as wetland conditions can be maintained throughout the full growing season. However, for phytoremediation objectives aiming at maximal metal uptake in aboveground biomass, soil oxidation must be guaranteed. Aerobic conditions in the topsoil are also prerequisite for degradation of organic pollutants in dredged sediment (Vervaeke et al., 2003).

Chapter 3.3. Effect of duration of seasonal flooding on Cd and Zn availability for the flood-tolerant *Salix cinerea* rooting in contaminated sediments

Several authors suggest that a hydrological regime aiming at wetland creation is a potential management option that favours reducing bioavailability for metal-contaminated sites. The hydrological conditions on a site constitute one of the many factors that may affect the availability of potentially toxic trace metals for uptake by plants. Bioavailability of Cd, Mn and Zn on a contaminated dredged sediment-derived soil with variable duration of submersion was evaluated by measuring metal concentrations in the wetland plant species *Salix cinerea* in field conditions. Longer submersion periods in the field caused lower Cd and Zn concentrations in the leaves. Foliar Cd and Zn concentrations were highest on the plot flooded in the early growing season but emerging earlier in the growing season. Foliar Zn concentrations were also high at a sandy textured, oxic plot with low soil metal concentrations. Zn uptake in the leaves was markedly slower than Cd uptake for trees growing on soils with prolonged waterlogging during the growing season, pointing at a different availability. Zn availability was lowest when soil was submerged, but translocation from branches to leaves masks the lower availability of Cd in submerged soils. Especially for Cd, a transfer effect from one growing season to the next season was observed: submersion conditions in the previous growing season seem to determine at least partly the foliar concentrations for *Salix cinerea* through this translocation mechanism. Duration of the submersion period is a key factor since initially submerged soils emerging only in the second half of the growing season resulted in elevated Cd and Zn foliar concentrations.

1. Introduction

Soil pH and oxidation-reduction potential (Eh) are dominant factors determining metal forms and behaviour, and hence mobility and availability. Flooding results in the creation of reducing conditions which may be accompagnied by changes in pH, typically towards the more alkaline region. Reduction processes are initiated shortly after flooding. Submersion of well-aerated upland soils resulted in a fast reduction of Mn^{4+} and Fe^{3+} into soluble Mn^{2+} and Fe^{2+} (Peters and Conrad, 1996; Scalenghe et al., 2002). Flooding of wetlands typically results in a decreased environmental availability of Cd, Zn, Cu and Ni (Gambrell, 1994; Kashem and

Singh, 2001). Tack et al. (1998) observed a short period of mobilisation of Cd and Zn, and subsequently lower concentrations in the saturation extract of a continuously wet treatment ('flooded') of landfilled sediments, compared with the concentrations for an alternating dry and wet regime. Similar observations were reported by Charlatchka and Cambier (2000) for an agricultural polluted soil.

Oxidation-reduction potential measurements allow for a fast although mostly qualitative assessment (Tanji et al., 2003) of reducing conditions developing upon flooding. Submersion resulted in a fast decrease of Eh (Kashem and Singh, 2001; Seybold et al., 2002; Haraguchi, 1991; Tanji et al., 2003), unless nitrate was artificially added (Charlatchka and Cambier, 2000). Water depth and Eh status were the main variables in determining the Zn speciation in a seasonally flooded wetland (Bostick et al., 2001). Hydrology (water level) was observed to be the primary factor that controlled Eh fluctuations (Haraguchi, 1991; Seybold et al., 2002; de Mars and Wassen, 1999), but absolute water table depth had little effect on Eh when water level was beneath the soil depth where Eh was measured (Haraguchi, 1991). On a local scale, spatial variation of Eh may determine vegetation composition (McKee, 1993).

Wetlands with alternating submerged and emerged periods have a specific soil chemistry. Zn speciation reversibly fluctuated in a seasonally flooded wetland: increasing water levels promoted the formation of ZnS and $ZnCO_3$, while in dry periods with oxidized soils fractionation was dominated by ZnO and Zn adsorbed on hydrated oxides (Bostick et al., 2001). Van den Berg et al. (1998) reported increased Cd and Zn pore water concentrations during the summer in a seasonally inundated wetland polluted with metals. However, pore water concentrations first increased for Zn, and afterwards for Cd. Increased Zn mobility was thought to be partly caused by oxidation of labile sulphides, while Cd might be released during biodegradation of organic matter (Van den Berg et al., 1998). Alternating oxidation and reduction of hydric soils in periodically waterlogged wetlands may cause intense decalcification of the upper soil layers (Van den Berg and Loch, 2000), and thus may affect metal availability.

Hydromorphic conditions result in an increasing oxygen demand for both soil and plant roots (Pezeshki, 2001) and may cause the production or accumulation of phytotoxic compounds (e.g. organic acids, methane, sulphides, reduced Mn and Fe) contributing to root injury, growth reduction and mortality (Kozlowski, 1997; Pezeshki, 2001). Pezeshki et al. (1997) and Pezeshki and DeLaune (1998) reported large differences in plant tolerance and effects on plant physiology and growth to reduced soil Eh conditions. Eh also affects the availability of essential elements and contaminants to plants. Several plant species showed lower Cd and Zn uptake in reduced soils (Gambrell and Patrick, 1989; Gambrell, 1994). Stoltz and Greger (2002a, 2002b) proposed to grow wetland plant species on submerged mine tailings as a phytostabilisation measure to maintain high pH values, reduce oxygen concentrations in the topsoil and immobilise metals. As this evidence suggests that flooding and subsequently reduction of contaminated soils may result in lower environmental bioavailability, it may constitute a valid management option for polluted soils.

The aim of this study was to determine whether the duration (period of anoxic soil conditions) and intensity (degree of soil reduction) of submersion of a polluted dredged sediment-derived soil (DSDS) affect the foliar metal concentration for *S. cinerea*, a typical wetland willow species. Foliar samples were collected on a DSDS during a growing season and soil oxidation-reduction condition was determined simultaneously. The advantage of willows for research on metal bioavailability is that they reflect time-integrated accumulated concentrations for the studied soil profiles.

2. Materials and methods

2.1. Soil sampling and measurements

A dredged sediment landfill in Semmerzake near Ghent (Belgium), referred to here as dredged sediment-derived soil (DSDS), was selected for this study. Willows occur as volunteer vegetation on sediment-derived soils such as dredged sediment landfills, overbank sedimentation zones and freshwater tidal marshes (Bal et al., 2001). The volunteer willow vegetation on this polluted DSDS site was compared with volunteer willows on a nearby infrastructure spoil landfill with baseline metal concentration levels. The DSDS (13.3 ha) was landfilled between 1992 and 1995 with sediments dredged in the Upper Scheldt, and can generally be characterised as a wetland with stagnant water from late autumn to mid-spring. On this site, 6 plots (Table 3.10) were selected representing variable hydrological regimes: plot 1 was located on the higher, sandy textured and least contaminated part of the DSDS, and plot 2 was not submerged anymore in week 15. Plot 3, plot 4 and plot 5 were waterlogged during winter but water level decreased strongly in spring and early summer, while plot 6 in the reed-dominated part was regularly waterlogged in summer as well, depending on the weather conditions. All plots were on calcareous soils. On the infrastructure spoil landfill used as reference site, 1 plot (Table 3.10, plot 0) was selected. On each plot, the 0-30 soil horizon was sampled in triplicate.

Time is expressed as the week number during the year. Oxidation-reduction potential was weekly measured between week 15 and week 31 with 4 replicates on all plots with a combined redox electrode (platina and Ag/AgCl reference electrode; HANNA instruments, HI 3131B) and a WTW multiline P3 meter, and values were reported after conversion towards the standard hydrogen electrode. The combined electrode was pushed into the soil to a depth of 5 cm and a stable reading was awaited. Oxidation-reduction measurements for plot 3 and plot 0 was not possible from week 22 onwards as the electrode could not penetrate the soil since the soil was too dry. Oxidation-reduction measurements were repeated in week 42. Simultaneously with the oxidation-reduction measurements, water level above the soil surface for the waterlogged soils was measured.

Table 3.10. Soil properties of the dredged sediment-derived soils (plot 1-plot 6) and the plot on the infrastructure spoil landfill (plot 0) where willow was sampled during the growing season (EC: electrical conductivity, OM: organic matter). Soil metal concentrations are *aqua regia* extractable concentrations (mg kg^{-1} dry soil). Soil oxidation-reduction potential (Eh) on a depth of 5 cm and water height on the soil surface are measured in week 15.

		plot1	plot2	plot3	plot4	plot5	plot6	plot0
clay	%	11	44	43	37	36	39	22
$CaCO_3$	%	6	11.4	9.9	11.6	11.8	11.6	2.3
EC	µS cm^{-1}	101	226	360	603	1281	824	140
pH-H_2O		7.91	7.60	7.60	7.50	7.29	7.40	7.40
OM	%	1.9	8.2	8.0	8.6	8.2	9.3	3.5
Cu	mg kg^{-1} dry soil	39	195	147	184	190	168	17
Cr	mg kg^{-1} dry soil	149	479	488	410	398	522	57
Pb	mg kg^{-1} dry soil	64	150	139	140	141	165	26
Ni	mg kg^{-1} dry soil	18	43	42	38	37	43	24
Mn	mg kg^{-1} dry soil	300	772	744	644	615	815	1036
Zn	mg kg^{-1} dry soil	329	1197	1165	971	1089	1342	99
Cd	mg kg^{-1} dry soil	1.9	12.7	14.2	9.1	9.4	14.7	0.6
Water level	cm	0	0	30	10	15	25	0
Eh	mV	466	464	103	19	113	132	87

2.2. Foliar and stem sampling

S. cinerea was identified based on leaf morphology (Weeda et al., 1999), and the presence and the length of the *striae* on one-year old branches (Meikle, 1984). *Striae* are longitudinal lines underneath the bark. *S. cinerea* can be distinguished from *S. aurita* and *S. x multinervis* based on the presence of solely long *striae* on one year-old branches (Meikle, 1984). To account for the variability associated with sampling, 4 *Salix cinerea* L. trees of approximately the same age and diameter were sampled within a circle with a diameter of 10 m on each plot. Foliar samples were taken with a three week interval between week 18 and week 42 during the growing season 2003. For plot 3, foliar samples were previously collected with the same methodology between week 24 and week 42 in 2001 and 2002, allowing for analysis of temporal and spatial variability. Our sampling strategy for leaves focused on individual trees. For foliar sampling, at least 4 branches from different heights and positions in the crown were sampled by means of an extension crosscut saw (Blair, 1995). Approximately 1000 cm^3 of leaf samples were collected at each sampling location. Leaves were not washed as washing procedures seem to produce misleading results due to incomplete removal of metals on the leaf surface and partial leaching of metals from the leaf tissues (Kozlov et al., 2000). Luyssaert (2001) showed that foliar Cd concentrations of trees grown at a dredged sediment landfill in the vicinity of the sites studied here are chiefly determined by soil pollution. For each tree, 2 willow cuttings from one-year-old dormant stems were collected in week 49 (December 2003). Trees on plot 1 were severely affected by herbivory between week 18 and week 24, but regrowth of leaves allowed for continuous sampling.

In week 33 *S. cinerea* was sampled with the sampling strategy described above on reference sites grouped according to soil contamination level (Table 3.11). Trees were sampled at 8 reference sites with baseline contamination levels (REF1), at 6 plots on a dredged sediment-derived soil with low metal concentrations (REF2) and at 6 sites with slightly contaminated soils (REF3).

2.3. Chemical analyses

Foliar and stem samples were dried for 7 days at 40 °C, mechanically ground (Pulverisette 14, Fritsch, Idar-Oberstein, Germany), and stored in dark vials before analysis. Total foliar N was measured by the Kjeldahl method (Van Ranst et al., 1999). Total foliar and stem element concentrations are extracted with HNO_3 (p.a. 65%) and H_2O_2 (ultrapur) in a 3:1

ratio using microwave digestion and measured with ICP-AES (Varian Liberty Series II, Varian, Palo Alto, CA). The accuracy of the foliar and stem element analysis was checked using BCR 60 (Aquatic plant) for Cd, Cu, Mn and Zn, and CRM 100 (Beech leaves) for S. Soil total concentrations of Cd, Cr, Cu, Ni, Pb, Mn and Zn are actually pseudo-total *aqua regia* extractable concentrations measured with ICP-AES after microwave digestion. Soil pH_{H2O} and electrical conductivity (EC) were measured in a 1:5 soil to water suspension. Organic matter (OM) was determined by the method of Walkley-Black, assuming that this method measures 75% of the total organic matter. $CaCO_3$ content was determined by back-titration with 0.5 M NaOH of an excess of H_2SO_4 added to 1 g air-dried sediment. The grain size distribution of the soil samples was determined using laser diffractometry (Coulter LS200, Miami, FL) with the clay fraction defined as the 0-6 µm fraction.

Table 3.11. 10th-90th Percentile range for soil properties of the reference plots where *Salix cinerea* was sampled in week 33 (EC: electrical conductivity, OM: organic matter, NA: not assessed). REF1: sites with baseline contamination levels, REF2: dredged-sediment derived soil with low metal concentrations, REF3: slightly contaminated soils. Soil metal concentrations are *aqua regia* extractable concentrations (mg kg^{-1} dry soil).

		REF1	REF2	REF3
clay	%	11 - 33	34 - 48	17 - 44
$CaCO_3$	%	0.4 - 9.2	2.6 - 5.5	1.7 - 4.3
EC	µS cm^{-1}	75 - 140	695 - 2040	100 - 312
pH-H_2O		6.05 - 8.59	4.87 - 7.45	6.88 - 8.44
OM	%	1.1 - 6.3	8.8 - 24.4	1.8 - 11.8
Cu	mg kg^{-1} dry soil	4 - 21	28 - 85	8 - 59
Cr	mg kg^{-1} dry soil	21 - 62	53 - 82	77 - 108
Pb	mg kg^{-1} dry soil	7 - 66	26 - 55	13 - 173
Ni	mg kg^{-1} dry soil	7 - 34	36 - 41	14 - 40
Mn	mg kg^{-1} dry soil	166 - 1130	NA	173 - 665
Zn	mg kg^{-1} dry soil	41 - 135	211 - 528	118 - 576
Cd	mg kg^{-1} dry soil	<0.3 - 0.7	0.7 - 1.2	0.8 - 4.6

2.4. Statistics

Effect of tree, year and week and second order interaction terms (tree:week, week:year and tree:year) on foliar Cd and Zn concentrations for plot 3 were tested with ANOVA. The mean square was applied as a measure of variance. Foliar data for week 33 for the 6 plots on the DSDS were compared with the results for the 3 reference data sets (REF1, REF2, REF3) with ANOVA and the Sidak method was applied for multiple comparison. Bioconcentration factors (BCF) for Cd and Zn on a dry weight (DW) base were defined as the ratio [foliar concentration/total soil concentration (*aqua regia*)].

3. Results and discussion

3.1. Hydrological regime

Redox measurements throughout the observation period reflect the changes in submersion state. Plot 2 and plot 0 were not any more submerged in week 15 (three weeks before the start of the foliar sampling campaign), while plot 4, plot 5 and plot 6 remained submerged until week 24. Plot 3 initially showed the highest water level, but water disappeared faster than on the other plots. It was submerged until week 19 (Fig. 3.10a). Oxidation-reduction potential values in the topsoil (Fig. 3.10b) were lower than 200 mV for plot 4, plot 5 and plot 6. The initial low Eh for plot 3 and plot 0 increased strongly in week 20 to levels typical for oxic (aerated) soils (Pezeshki, 2001). Eh values for the submerged plots were decreasing until week 22 (end of the submersion), sharply decreased and then increased towards 200-250 mV, values still pointing at prevailing anaerobic conditions. Eh values of 350-400 mV are the boundary between presence and absence of oxygen (Pezeshki, 2001). Plot 4, 5 and 6 were subjected to more prolonged periods of waterlogging. Eh values at plot 4, 5 and 6 until week 29 point at reductive dissolution of Mn and Fe and formation of sulphides (Peters and Conrad, 1996; Bostick et al., 2001). A decrease in Eh just after emergence was also reported by Haraguchi (1991) for a submerged floating peat mat both in field and in pot experiments. Haraguchi (1991) attributed this decrease to prolonged water saturation and restricted oxygen diffusion, combined with strong organic matter decomposition (Haraguchi, 1991). The observed Eh patterns over the growing season may vary with soil depth (Seybold et al., 2002), and may be modified by the presence of adult root systems (McKee, 1993). After emergence of plot 4, plot 5 and plot 6, soil was still waterlogged, while plot 3 emerged earlier and was fast transformed to an aerated soil with a gradually developed crumbly topsoil structure. The summer of 2003 was exceptionally hot and dry. This may explain why plot 6, found still to being submerged in August 2002, was already emerged in June 2003.

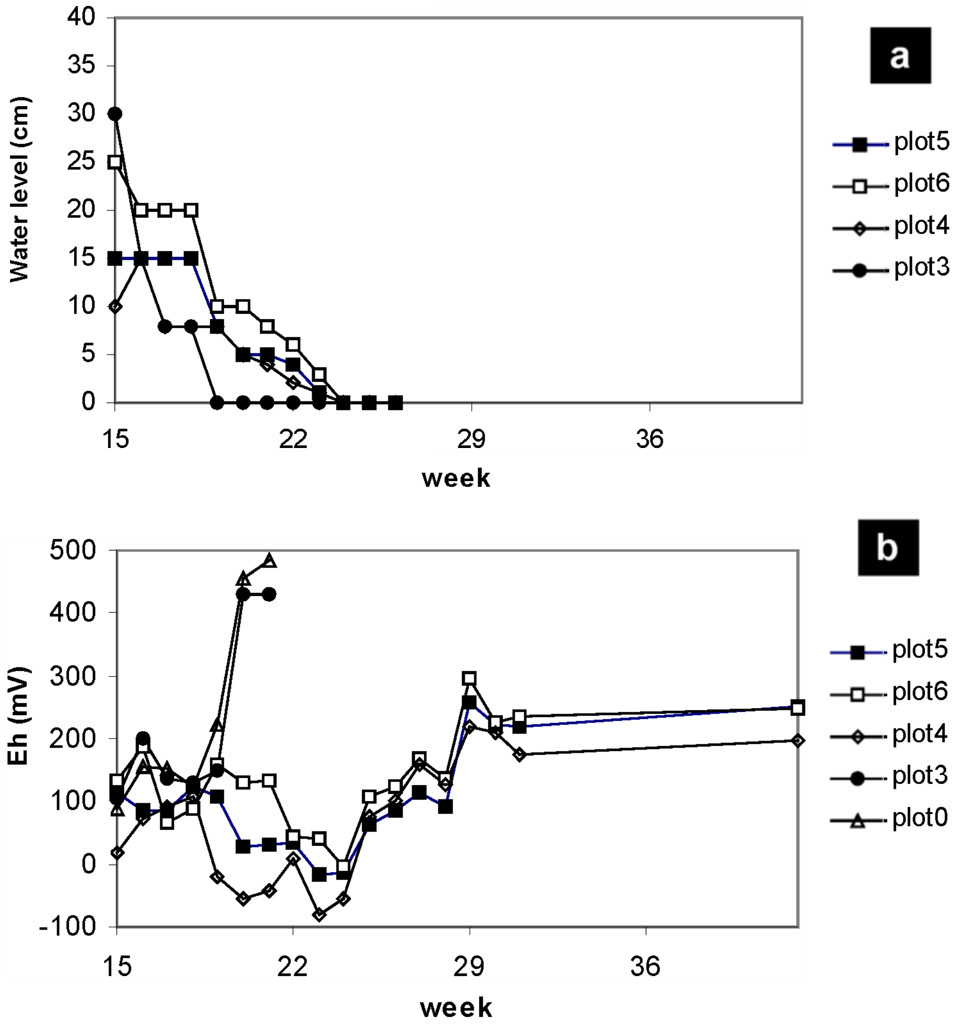

Fig. 3.10. Evolution of (a) the water height on the soil surface and (b) the weekly measured soil oxidation-reduction potential on a depth of 5 cm for the 6 sampled plots

3.2. Temporal and spatial variability

Four trees on plot 3 were sampled between week 24 and week 42 in 2001, 2002 and 2003. Factors ‘week’, ‘tree’ and ‘year’ significantly affected foliar Cd and Zn concentrations (Fig. 3.11, Table 3.12). Although all factors were highly significant in explaining variability, variance associated with factor ‘week’ and ‘tree’ was higher than variance for the sampling year (Table 3.12). Cd concentrations were highest for tree 1, especially in 2003. Concentration patterns (Fig. 3.11) of Cd and Zn differ in that Cd initially tends to remain constant or decrease slightly, while Zn concentrations increased right from the beginning of the observation period.

Non-destructive foliar samplings allow for *in situ* biomonitoring using willows. Current data illustrate the large variability between individual trees on the same stand. It is thus necessary that a same tree is sampled to assess changes in foliar concentrations with time.

Table 3.12. ANOVA descriptives for foliar Cd and Zn concentrations (mg kg^{-1} DW) on plot 3, where 4 trees were sampled between week 24 and week 42 in three consecutive years.

	Factor	Mean square	Pr (F)
Cd	Tree	1197	0.000
	Week	507	0.000
	Year	147	0.001
	Tree:Week	53	0.004
	Week:Year	29	0.151
	Tree:Year	96	0.001
	Residuals	18	
Zn	Tree	1130226	0.000
	Week	2804138	0.000
	Year	396656	0.000
	Tree:Week	80288	0.000
	Week:Year	23144	0.102
	Tree:Year	180260	0.000
	Residuals	13400	

3.3. Baseline concentrations in week 33

Foliar concentrations measured in week 33 were compared with concentrations measured in the same period on reference plots to evaluate the effect of the submersion period (Fig. 3.12). Multiple comparison revealed significantly higher foliar Cd concentrations in week 33 for plot 1, plot 2 and plot 3 than for the REF1 plots with baseline soil concentrations, while results for plot 4, plot 5 and plot 6 where not significantly different. Similar results were obtained for the comparison with the REF2 plots on an uncontaminated DSDS. Only plot 3 had significantly higher Cd foliar concentrations than the concentrations measured on slightly contaminated soils (REF3). Differences were less outspoken for Zn: only for plot 3 foliar concentrations significantly differed from the three reference data sets, and concentrations for plot 1 were significantly higher than the plots with baseline soil concentrations (REF1). No significant differences were found for Fe and Mn, and Cu concentrations at plot 1 were significantly higher than concentrations at REF1 and REF3. Foliar Cd concentrations in week 33 were thus in the normal range for plot 4, plot 5 and plot 6 (the plots with the longest duration of submersion), while slightly elevated concentrations were measured for the oxic plot 1 and plot 2 and deviant concentrations were measured for plot 3. High foliar Cd (Punshon and Dickinson, 1997b) and Zn (Álvarez et al., 2003) accumulation in *S. cinerea* was also observed for other oxic contaminated soils.

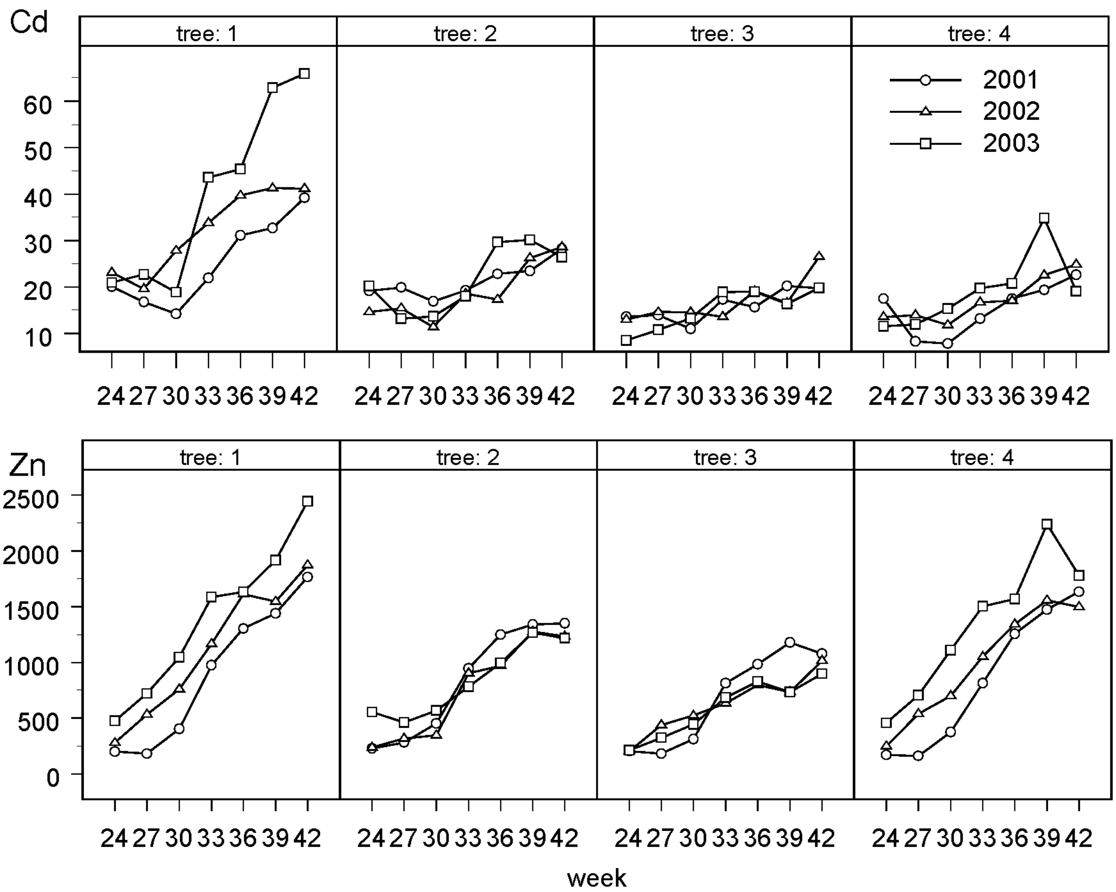

Fig. 3.11. Foliar uptake patterns for Cd and Zn (mg kg^{-1}DW) during the growing season (week 24 to week 42) for 4 *S. cinerea* trees (tree 1, 2, 3, 4) on a dredged sediment-derived soil (plot 3) in three consecutive years.

3.4. Uptake patterns

Uptake patterns over the growing season (Fig. 3.13) demonstrate increasing trends for Cd, Zn, Fe and Mn, while S concentrations decrease in the first weeks of the growing season and increase afterwards. Cu concentrations decrease in the first weeks of the growing season and increase marginally towards harvest. S concentration increases strongly from week 30 on and is particularly high for plot 5 and to a lesser extent for plot 4 at the end of the growing season. Oxidation of the sediment topsoil resulting in a higher S availability might be the reason.

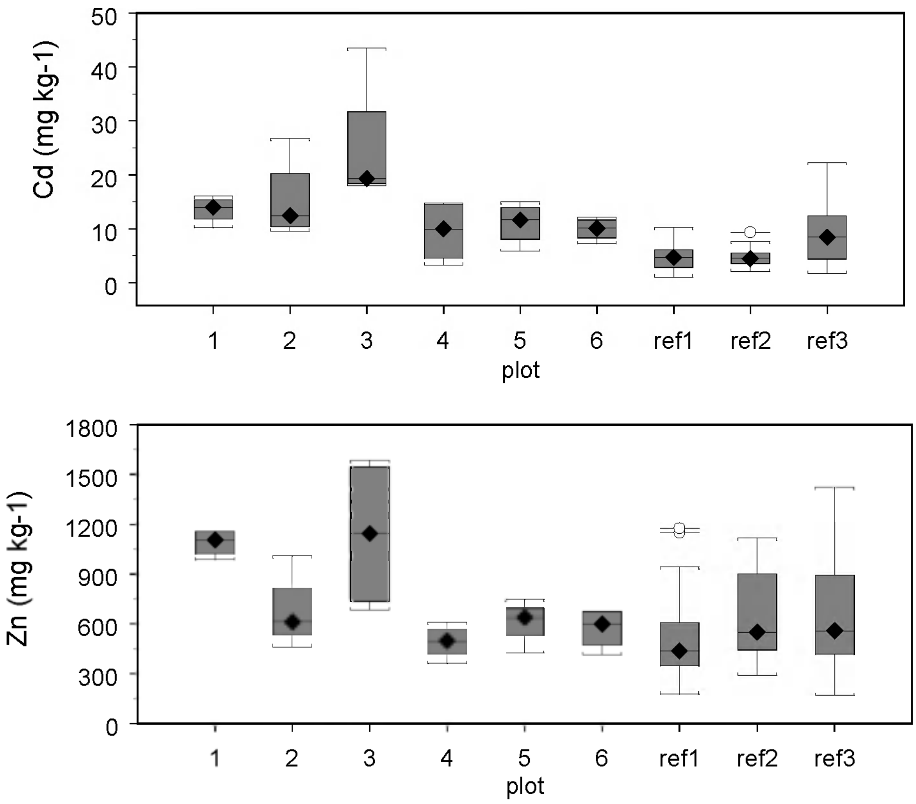

Fig. 3.12. Comparison of the foliar concentrations for Cd and Zn (mg kg^{-1} DW) on the 6 plots on a dredged sediment-derived soil with three reference data sets (ref1: plots with baseline contamination levels, ref2: plots on a dredged sediment-derived soil with low metal concentrations and ref3: plots on slightly contaminated soils). Open circles indicate outliers.

For plot 1, plot 2 and plot 3 Cd concentrations slightly decrease between week 21 and week 30. The initially high foliar concentrations might indicate translocation from stems to foliage in the early growing season for the plots with the highest Cd availibility, and the subsequent decrease might be a result of dilution by growth. Cd concentration is relatively high for plot 3, especially from week 33 on. Mn concentrations are highest for plot 6, the plot in the wettest part of the landfill, and lowest for plot 1. This may reflect higher Mn availability in reduced soil conditions (Peters and Conrad, 1996; Scalenghe et al., 2002). Cu is highest for the sandy-textured plot 1. This is the plot with the lowest soil Cu concentration, but also with the lowest OM content (Table 3.10). OM is known to largely reduce Cu availability. Zn concentrations are high for plot 1 and plot 3 in the second part of the growing season. BCF for Cd and Zn was highest for plot 1, some lower for plot 0 and lowest for plot 6 (Fig. 3.14). Willows on uncontaminated and slightly contaminated soils typically have high BCF for Cd and Zn (Granel et al., 2002; Meers et al., 2003), while lower BCF for Cd and Zn on polluted DSDS were found for other willow species as well (Vandecasteele et al., 2004c).

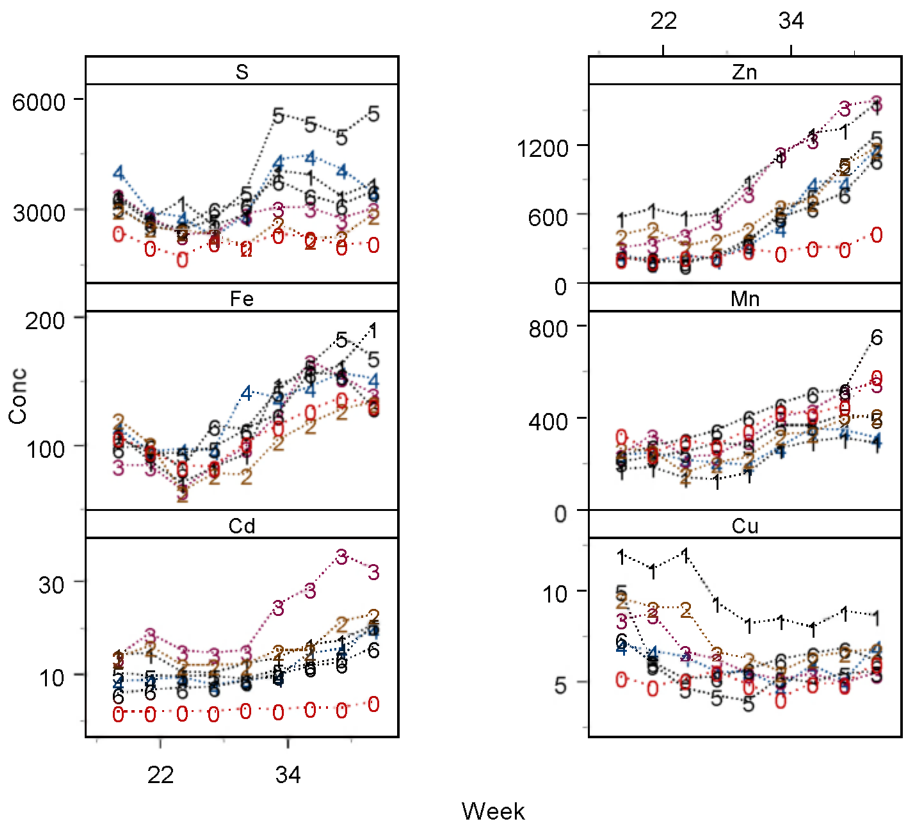

Fig. 3.13. Average foliar uptake patterns for Cd, Zn, Cu, Mn, Fe and S (mg kg^{-1} DW) during the growing season (week 18 to week 42) for *S. cinerea* (4 trees per plot) on a dredged sediment-derived soil (plot 1-plot 6) and an uncontaminated infrastructure spoil landfill (plot 0).

Cd:Zn ratios in leaves (Fig. 3.15) clearly decreased during the growing season. All plots, except plot 1 and plot 0, have a pronounced increase of the Cd:Zn ratio in the first weeks of the growing season, and a decrease afterwards. This graph illustrates that Cd is more available for leaves than Zn in the early growing season. Fig. 3.15 also demonstrates that the Cd:Zn ratio in the stems (sampled in week 49) matches the foliar ratio better in the early growing season than in the late growing season.

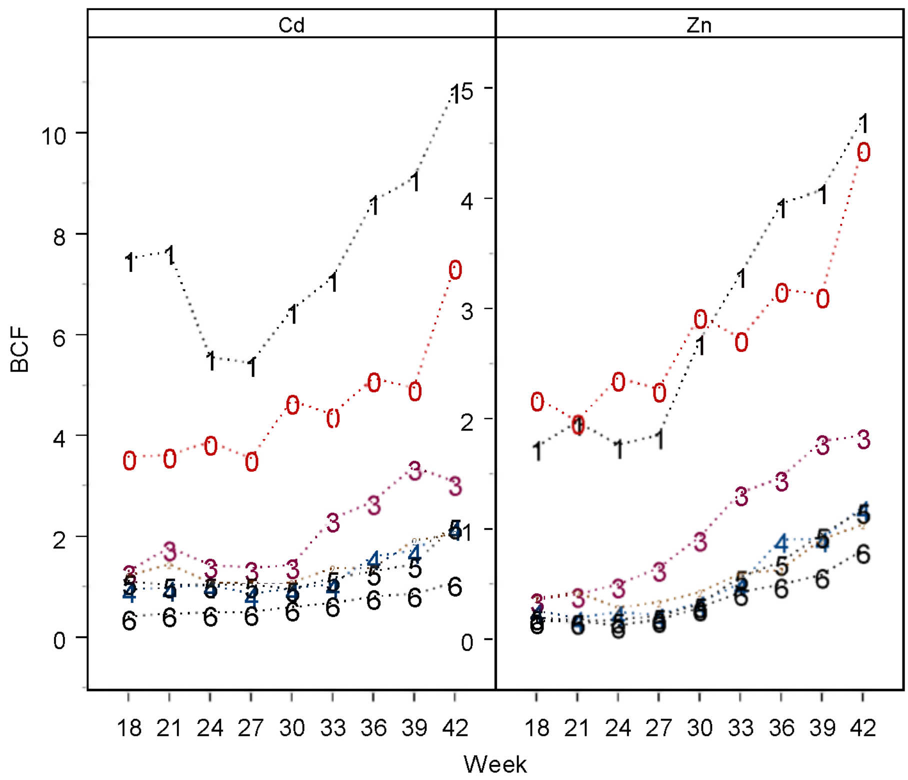

Fig. 3.14. Evolution of the bioconcentration factor (BCF) for Cd and Zn in the leaves of *S. cinerea* during a growing season for 6 plots on a dredged sediment-derived soil and 1 plot on an infrastructure spoil landfill with baseline contamination levels.

Cd, Zn and Mn concentrations expressed relatively towards the concentration in week 42 (Fig. 3.16) indicate large differences in accumulation dynamics for Cd and Zn. Relative concentrations increase proportionally for Cd and Zn for plot 0, but the Zn concentration increase is much slower for plot 2, plot 4, plot 5 and plot 6 (proportional to Cd from week 36 or later on) than for plot 1 and plot 3 (proportionality already met in week 30). The initial Cd and Zn concentration as percentage of the concentration in week 42 is relatively low for plot

3, plot 4, plot 5 and plot 6, while foliar concentrations for plot 1, plot 2 and plot 0 are relatively high in the early growing season (Fig. 3.16).

3.5. Effect of submersion on Cd and Zn availability

As opposed to Zn, Cd concentrations were relatively high in the early growing season, even for submerged soils. This might indicate a higher Cd availability in reduced soils, but contradicts other observations. In general Cd and Zn exhibit a similar environmental behaviour in DSDS which is reflected in similar fractionation and extractability (Singh et al., 1998). Van den Berg et al. (1998) reported that Zn pore water concentrations increased earlier than Cd in an emerged wetland. We, in contrast, observed higher Cd foliar concentrations early in the growing season.

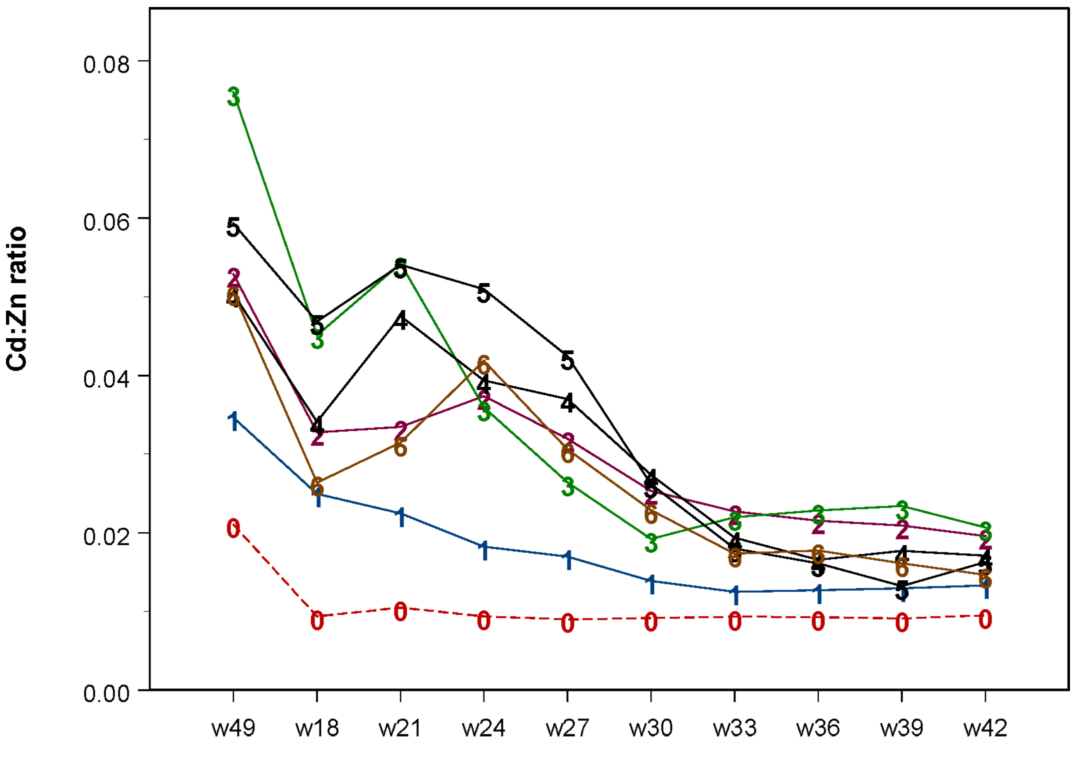

Fig. 3.15. Cd:Zn ratio in stem cuttings (taken in week 49) and leaves (sampled between week 18 and week 42) for 6 plots on a dredged sediment-derived soil and 1 plot on an infrastructure spoil landfill with baseline contamination levels (Table 3.10).

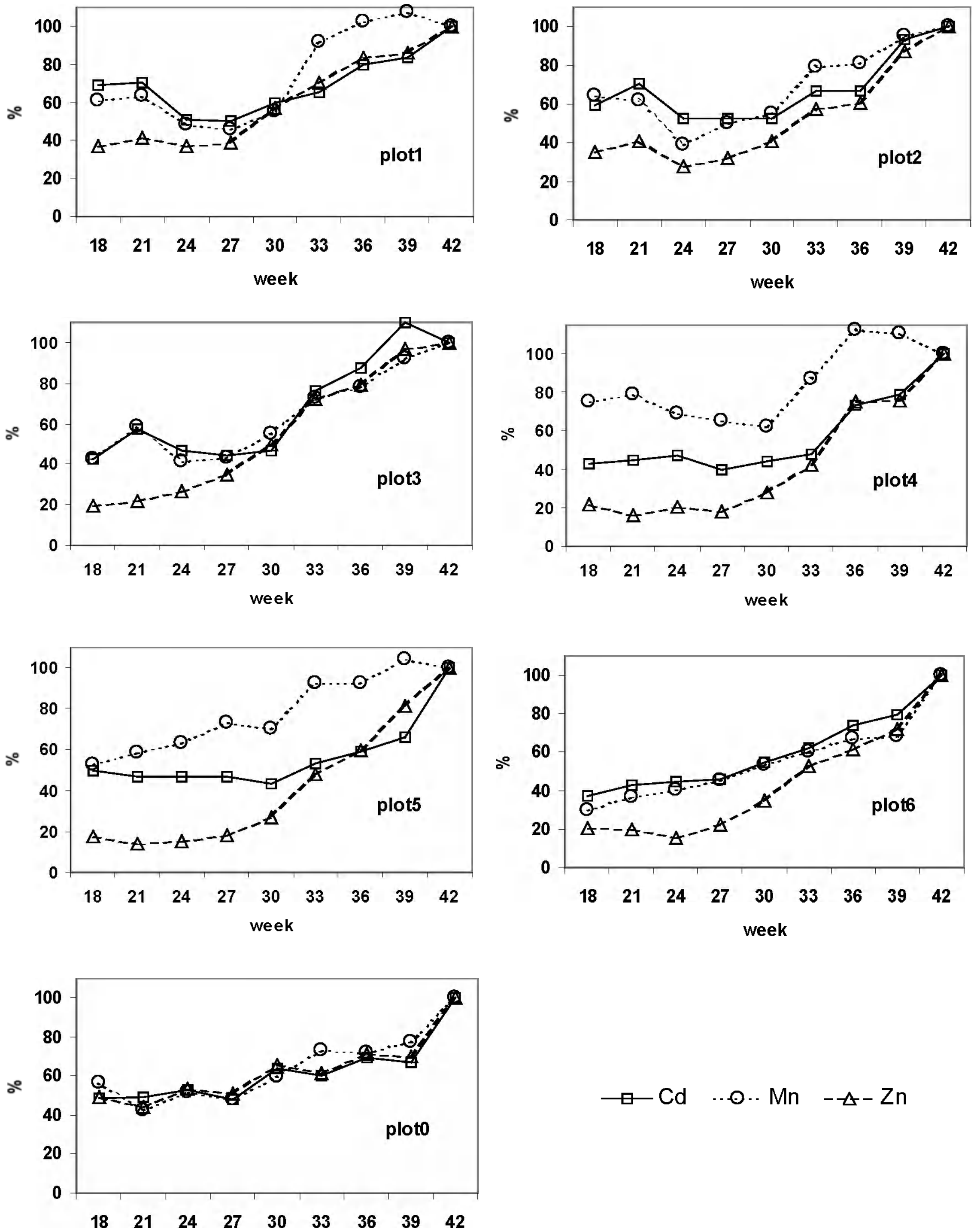

Fig. 3.16. Relative uptake of Cd (squares), Zn (triangles) and Mn (circles) during the growing season for *S. cinerea* on 6 plots (1-6, see Table 3.10) on a dredged sediment-derived soil and 1 plot on an infrastructure spoil landfill with baseline contamination levels. Values are averages for 4 individually sampled trees and are expressed relative to the concentration measured in week 42.

Table 3.13. Cd, Zn and Mn concentrations in stem cuttings collected in week 49 on the 6 plots on a dredged sediment-derived soil and 1 plot on an infrastructure spoil landfill with baseline contamination levels.

		Plot0	Plot1	Plot2	Plot3	Plot4	Plot5	Plot6
Cd	mg kg^{-1} DW	2.9	16.2	15.2	18.9	12.5	13.0	7.9
Zn	mg kg^{-1} DW	158	440	275	242	244	234	156
Mn	mg kg^{-1} DW	94.6	55.1	78.0	130.1	63.4	53.5	67.9

Foliar metal concentrations in the field might be influenced by the previous growing season due to translocation of elements from the stems to the leaves. Landberg and Greger (1994) found a significantly positive correlation between initial Cd concentrations in field-collected cuttings and Cd concentrations in the shoots of diverse *Salix* species. Foliar concentrations in willows are thus partly determined by Cd translocation from stems to leaves. Strong positive correlation was observed between Cd concentrations in the branches (collected in week 49) and in leaves collected in week 18 (R^2 = 0.849) or week 42 (R^2 = 0.702). Positive correlation was markedly lower for Zn in branches and in week 18 (R^2 = 0.302) or week 42 (R^2 = 0.187), and not significant for Mn in branches and in week 18 (R^2 = 0.109)) or week 42 (R^2 = 0.062).

The results of the foliar samplings suggest that Cd is strongly translocated from stems to leaves in the early growing season, while stem cuttings collected at the end of the growing season contained high Cd levels and point at high bioavailability of Cd at this time (Table 3.13). Cd:Zn ratio in the stems is 2 to 4 times as high as Cd:Zn ratio in leaves (Fig. 3.15), illustrating the differences between Cd and Zn in translocation behaviour from stems to leaves.

4. Conclusions

After emergence, soils showed variable rates of oxidation, visualised in differences in topsoil structure. A crumbly soil structure due to soil drying accelerates oxidation, while a waterlogged, muddy structure allows the anaerobic soil conditions to prevail. Submersion and waterlogging in the first weeks of the growing season resulted in normal foliar Cd and Zn concentrations, but emergence then sharply increased foliar concentrations to levels comparable with the plots already emerged at the beginning of the growing season.

Variability in foliar Cd and Zn concentrations was lower between sampling years than between individual trees and sampling period. Tree individuality must therefore be acknowledged for when using willow leaves for biomonitoring. Bioconcentration for Cd, Cu and Zn in the leaves was highest for the sandy-textured soil with relatively low contamination levels. Foliar Cd and Zn concentrations were highest for the soil initially submerged but with the shortest submersion period and the most rapid emergence and oxidation.

Submersion conditions in the previous growing season seem to determine at least partly the foliar Cd concentrations for *Salix cinerea* through translocation from stems to leaves. Hydrological regime aiming at wetland creation is a potential management option for reducing bioavailability and thus for establishing a safe management of wetlands polluted with metals as long as submersion can be maintained till the end of the growing season.

Part 4. Ecosystem effects

Chapter 4.1. Rates of forest floor decomposition and soil forming processes as indicators of forest ecosystem functioning on a polluted dredged sediment landfill

Results of intensive monitoring of forest floor decomposition rates and soil forming processes after afforestation of a calcareous upland dredged sediment landfill with an oxidised topsoil are presented. Nutrient status of the sediment substrate favours tree growth and allows for afforestation and thus integration in the landscape. Soil processes on the landfill resulted in small differences between the topsoil and the deeper soil layer, although higher soil organic carbon and Cd concentrations in the topsoil were observed. Relative to the uncontaminated covered part of the site and to general references, forest floor decomposition was found to be relatively fast for sycamore maple and pedunculate oak. Despite the pollution status of the dredged sediment landfill, the sediment substrate was favourable for forest floor decomposition. This might indicate that the soil nutrition status and the high carbonate status override the negative impact of soil pollution with metals and other pollutants. Application of an uncontaminated cover topsoil resulted in lower Cd concentrations in earthworms, but concentrations were still higher than for references. We conclude from the observations that polluted but fertile soils allow for afforestation and for regular forest floor decomposition with normal or slightly elevated metal concentrations. Only long-term observations of such new forests will lead to a correct site-specific assessment of the actual ecological risks, but after 16 years of landfilling and 12 years of afforestation no adverse effects were observed.

1. Introduction

1.1. Afforestation of dredged sediment landfills

Afforestation of calcareous, polluted, dredged sediment landfills has several environmental benefits such as soil stabilisation and visual buffering. Research on afforestation techniques initially focused on pioneer tree species (willow and poplar species), especially for biomass production, phytoextraction and phytoremediation (Vervaeke *et al.*, 2001). Both willows and poplars showed elevated foliar Cd and Zn concentrations relative to uncontaminated sites (Mertens et al., 2001; Vandecasteele et al., 2002c; Vandecasteele et al., 2003b), even when an uncontaminated cover topsoil was established (Vandecasteele et al., 2003b). In afforestation of polluted, dredged sediment landfills, questions were raised concerning metal cycling and how the adverse effects of the cycling may be minimised by an adapted management. An appropriate tree species choice was shown to be very important in controlling metal uptake in leaves (Vandecasteele and De Vos, 2002).

Rather than trying to clean the soil by phytoextraction, afforestation attempts to reduce and control the metal fluxes through the ecosystem. Earlier research on a landfill site along the Leie river revealed excellent tree growth and normal foliar metal concentrations for common ash, pedunculate oak and sycamore maple (Vandecasteele and De Vos, 2002). These results demonstrated that polluted dredged sediment landfills could be easily revegetated in contrast to other polluted sites or landfills where a vegetative cover could only be established after considerable efforts (Bleeker et al., 2002; Ye et al., 2002).

1.2. Forest floor decomposition

The forest floor acts as a sink for contaminants and determines their fate, as was demonstrated for deposition and degradation of polycyclic aromatic hydrocarbons (Howsam et al., 2001). The forest floor mass and especially the forest floor decomposition rate can be considered an important indicator of long-term adverse effects of metal pollution (Martin *et al.*, 1982; Martin and Bullock, 1994, Laskowski *et al.*, 1995; Gillet and Ponge, 2002).

Beyer et al. (1990) concluded that leaching determines the availability of metals to the food chain on acid dredged sediment landfills, whereas on calcareous landfills, the accumulation of metals in the forest floor is the prevailing factor. In areas receiving atmospheric deposition from smelters, forest floor and humus were the most important sinks for metals (Bengtsson and Tranvik, 1989). Under these particular circumstances, Martin and Bullock (1994) found that earthworms, millipedes and woodlice in contaminated woodlands were significantly less abundant than in unaffected woodland. In the contaminated woodland, high Cd and Zn concentrations were found in earthworms, woodlice, snails and slugs and high litter accumulation was attributed to interferences with the natural decomposition process by elevated Cd and Zn concentrations.

1.3. Aims and objectives

On dredged sediment landfills, litter accumulation may result in soil acidification and may eventually increase metal availability. The variation of forest floor decomposition within a climatically homogeneous region is mainly explained by litter characteristics and, thus, by tree species (Muys and Lust, 1992; Muys, 1995). As a consequence, it is proposed that the forest floor decomposition capacity of a polluted soil as determined by the tree species choice is important for the development of an environmentally safe afforestation strategy for dredged sediment landfills. In this paper we present the results of intensive monitoring of forest floor decomposition rates and soil forming processes after afforestation of a calcareous upland dredged sediment landfill with prevailing aerobic conditions in the upper soil horizons.

Objectives of this study are:

1. Determination of the rates of soil forming processes in the sediment topsoil
2. Comparison of the forest floor decomposition rates and metal concentrations in the forest floor for two tree species with different litter characteristics: sycamore maple (intermediate decomposition) and pedunculate oak (slow decomposition).
3. Evaluation of the use of an uncontaminated cover topsoil based on metal concentrations in earthworms.

Based on these results the concept of controlled afforestation of polluted dredged sediment landfills is refined.

2. Materials and methods

2.1. Site description

The Meigem landfill site is situated on the left bank of the Leie Derivation Canal in Deinze, Belgium. The site was established for the disposal of sediments from maintenance dredging operations. In 1985 the site was raised 2 m with sediments dredged at the intersection of the Leie and the Derivation Canal at Deinze after the original soil material was used for dike construction. In spring 1987, the landfilled sediment was covered with a thin uncontaminated layer (40 cm) by pushing the landfill dikes inside. However, in November 1987, the central part of the basin was still swampy, and no cover topsoil was present here (De Vos, 1989). The whole site was afforested in 1990-1992 with mainly common ash (*Fraxinus excelsior* L.), pedunculate oak (*Quercus robur* L.), sycamore maple (*Acer pseudoplatanus* L.) and hybrid poplars (*Populus trichocarpa x deltoides* clones) in a regular pattern of 30 x 30 m plots laid out partly in covered, partly in uncovered zone (Fig. 4.1). The landfill is divided by alleys lined with poplars per eight plots. Ten years later, soil (December 2001), forest floor (from December 2001 till December 2002) and earthworms (April 2002 and November 2002) were sampled.

2.2. Soil forming processes in the sediment topsoil

Soil processes were monitored in 16 uncovered plots (= plots in the central part without cover topsoil) under different tree species (pedunculate oak: 2 plots, sycamore maple: 5 plots, common ash: 4 plots, hybrid poplar: 3 plots, unforested reference: 1 plot). In each plot the soil was sampled in December 2001 for detailed comparison of top and deeper soil layer. Five points selected at random in each plot were sampled at two depths: 0-15 cm and 15-30 cm (75 samples for each depth). Each sample was a composite of 5 subsamples taken in a circle of 1 m diameter around the sampling point.

Soil data for the uncovered plots were pair-wise compared for both sampling depths with the paired t-test. The deeper soil material is expected to be less influenced by soil forming processes, and a comparison between topsoil and deeper soil can reveal the dynamics of soil forming processes. Interpretation of differences in chemical composition between top and deeper soil are based on the variogram (Webster and Oliver, 2001). A difference might be found to be highly significant, but the absolute difference might be less relevant from an environmental point of view. For each soil property a variogram was constructed for the 15-30 cm soil data. The nugget effect was employed as a measure for the variance which cannot be explained by the spatial pattern and is thus the result of both short-distance variance, and sampling and laboratory errors.

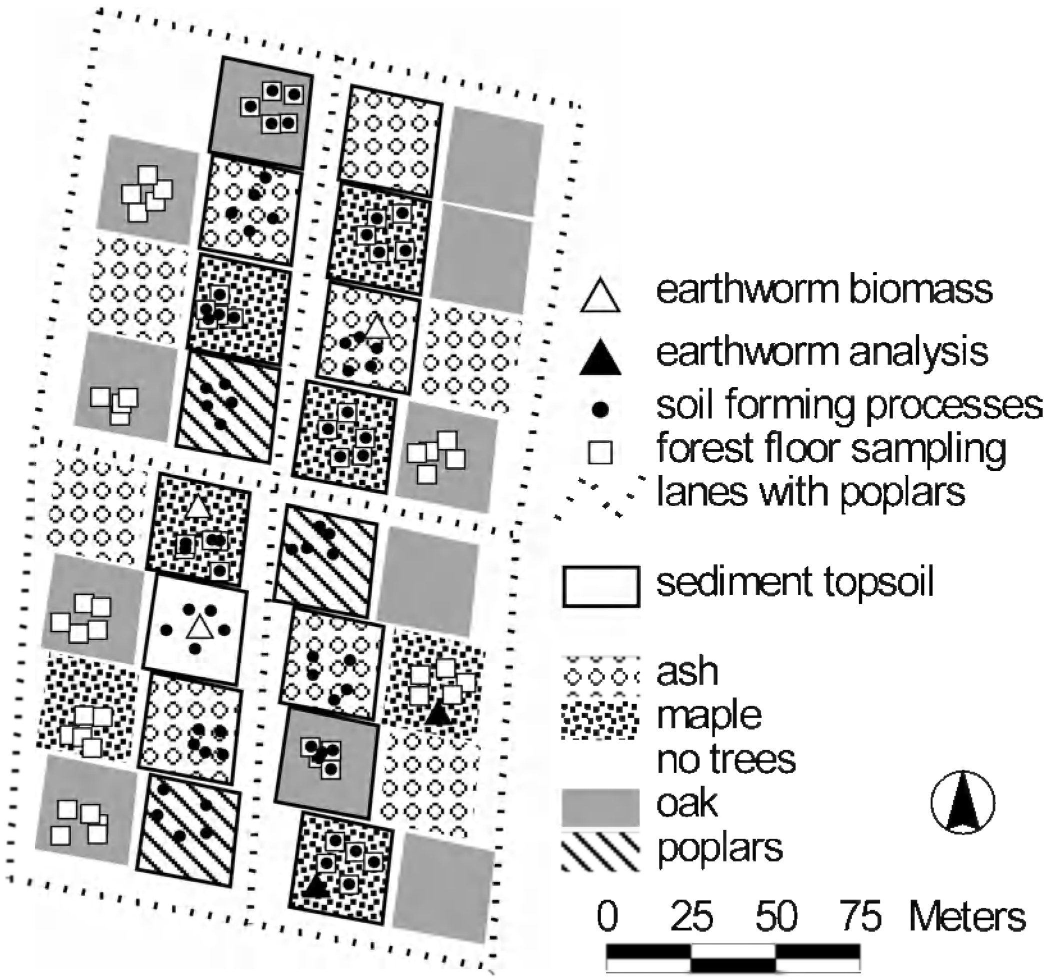

Fig. 4.1. The Meigem site with sampled plots selected for (1) determination of soil forming processes, (2) forest floor decomposition and (3) earthworm sampling.

2.3. Forest floor decomposition rates and metal concentrations

For site-specific monitoring of forest floor accumulation and decomposition with the focus on forest floor mass, forest floor samples were collected two-monthly in the field during a year. Forest floor was sampled at 5 subplots on a surface of 0.5 x 0.5 m for 7 plots with pedunculate oak and 7 plots with sycamore maple (Fig. 4.1). Standing crop for maple in 2001 was clearly higher than for oak (238 ± 50 versus 74 ± 18 m^3 ha^{-1}). Mixed litter input, a commonly observed process found to affect decomposition processes (Taylor et al., 1989; Zimmer, 2002), continuous litter input over the growing season and forest floor being affected by previous years were accounted for in this sampling strategy.

The forest floor was sampled the first time in December 2001, and the sampled plots were marked with a small stick. Each new sampling was executed 1m northwards from the previous sampling. The forest floor samples were selected for leaf litter only: twigs, bark pieces and other non-foliar materials were rejected. The forest floor samples were weighed in the field. They were transported to the lab for dry weight determination and analysis after drying in a ventilated oven at 40° C.

The topsoil of the selected plots for forest floor sampling was sampled for general characterisation with the same methodology as described for the topsoil of the uncovered plots. Soil sampling allowed for a topsoil classification for all sampled points in three classes: sediment topsoil, cover topsoil and a mixed transitional topsoil between both types.

The forest floor samples in October and December 2002 were manually divided in a fraction of poplar leaf material and a remaining fraction, consisting mainly of leaves from the dominant tree. Cd and Zn concentrations in both fractions were compared with ANOVA with 'sampling period', 'dominant tree species' and 'litter fraction' as three fixed factors. No interaction between these factors in the second or third order terms was observed.

Contamination of the forest floor with soil particles as a consequence of splash erosion and invertebrate bioturbation might partly mask the forest floor mass loss as a decomposition indicator. Therefore, we took only the organic fraction of the forest floor into account, measured as total organic carbon (%TOC). Organic carbon mass in the forest floor per m^2 (g OC m^{-2}) was calculated as the product of forest floor mass (g DW m^{-2}) and the percentage TOC.

Average forest floor decomposition rates expressed as weight loss per month per surface unit for both dry weight (g DW m^{-2}) and organic carbon (g OC m^{-2}) were calculated for a period with distinct net forest floor decomposition. To calculate decomposition rates for the forest floor under maple, the period between December and June was selected. For oak, the period between February and June was focused on.

Forest floor weight expressed as organic carbon per surface unit (g OC m^{-2}), C:N ratio and forest floor dry weight content (% DW) for four periods (December 2001, February, April, June 2002) and two topsoil types (cover topsoil and sediment topsoil) were compared with two-way ANOVA for both maple and Oak dominated plots. No interaction between sampling period and topsoil type was found. Multiple comparison for the sampling periods was performed with the Sidak test at a 0.95 confidence level (Insightful Corporation, 2001). pH_{CaCl2} in the forest floor under oak and maple in April were compared with the t-test.

Before metals in the forest floor of oak and maple were measured, forest floor samples were pooled per tree species, per plot and per topsoil type: 10 composite samples for oak and 9 composite samples for maple per sampling period (December 2001, February, April and

June 2002) were analysed. Forest floor quality data were compared for sampling period and dominant tree species with two-way ANOVA. Multiple comparison of Cd concentrations and amounts in the forest floor for several sampling periods was executed with the Sidak method (Insightful Corporation, 2001).

2.4. Earthworm biomass and effect of cover topsoil on earthworm metal concentrations

Earthworm biomass and density were determined in April 2002 on a plot under maple, a plot under Ash and an unforested plot in the uncovered, central part of the site with six replicates per plot (Fig. 4.1). Earthworms in the litter layer sampled in a (50 x 100) cm^2 area were separated by hand sorting in the lab. Four successive waterings were performed on the same area with a diluted formalin solution at increasing concentration (0.125%, 0.125%, 0.25%, 0.25%). Earthworms were preserved in a 5% formalin solution. Within a week after sampling, total earthworm biomass per sample was determined after drying for 1 min on a filter paper at room temperature.

Earthworms for metal analysis were collected with formalin extraction on both an uncovered and a covered plot under maple in November 2002 (Fig. 4.1). Extracted earthworms were washed directly in demineralised water, and starved for 24 h on wet filter-paper before weighing after drying for 1 min on a filter-paper. For each plot, five adult *Lumbricus rubellus* with a weight between 0.188 and 0.292 g fresh weight (FW) and 5 juvenile lumbricids with a weight between 0.157 and 0.490 g FW were selected for metal analysis. The animals were killed in demineralised water at 40 ° C and dried in a ventilated oven at 40° C.

Cd and Zn concentrations in juvenile and adult lumbricid earthworms were compared for a covered and an uncovered plot under maple with ANOVA. Both factor 'stage' (juvenile or adult) and 'soil type' and an interaction term were included. No significant influence of soil type was found, while 'stage' resulted in significantly different weights. In neither case an interaction for metal concentrations between 'stage' and 'soil type' was observed.

2.5. Soil, forest floor and earthworm analyses

The forest floor samples were mechanically ground (Pulverisette 14, Fritsch, Idar-Oberstein, Germany). Total N in the forest floor was measured by the Kjeldahl method. Total forest floor metal concentrations were extracted with HNO_3 (p.a. 65%) and H_2O_2 (ultrapur) in a 3:1 ratio using microwave digestion (Milestone 1200 MS Mega), and measured with ICP-AES (Varian Liberty Series II, Varian, Palo Alto, CA). Forest floor pH was measured in a 1:5 soil to water suspension. Methods for soil analysis are described in Chapter 1.1.

For Cd and Zn determination in field-collected earthworms, a method adapted from Tack et al. (2000, method 4) was used. Samples were weighed into 100 mL pyrex beakers and treated with 10 mL ultra-pure 65% HNO_3. The beaker was covered with a watch-glass and the suspension was heated up to 130°C for 1 h. A total amount of 4 mL 20% H_2O_2 was added in aliquots of 0.5 mL. After cooling, the destruate was filtered (S&S, blue ribbon) in a 25-mL volumetric flask and diluted to the mark. Cd and Zn contents were measured in the extracts using flame (Varian SpectrAA-1475, Varian, Palo Alto, CA) atomic absorption spectrometry.

3. Results

3.1. Soil

The sediment topsoil is characterised by high clay and organic carbon contents, high nutrient contents (illustrated by the low C:P ratio) and markedly high carbonate contents and electrical conductivity (Table 4.1). The sediment topsoil is mainly contaminated with Cd and Zn. The cover topsoil is a calcareous loamy sand textured soil with high TOC contents (Table 4.1).

Pair-wise comparison of the topsoil layer with the deeper soil layer for 75 points revealed no significant difference between both layers for clay, silt, sand, P, S and Pb. In the upper layer, significantly higher values for Cd, N_{soil} and TOC ($p < 0.0001$) were found (Table 4.2). Lower values were found in the topsoil for Cu, Cr, Ni, pH_{H2O} and pH_{CaCl2} ($p < 0.0001$), Zn, EC ($p = 0.0297$), and $CaCO_3$ ($p = 0.0076$) (Table 4.2). The short range variability calculated from the variogram indicates that the observed differences for Cd, TOC, N_{soil} and pH_{CaCl2} are most relevant.

Table 4.1. Topsoil properties (0-15 cm) of the uncovered plots (90 samples) and the plots with cover topsoil (26 samples) at the Meigem site. Data are means ± standard deviations

		Uncovered	Covered
clay	%	34.1 ± 1.7	20.2 ± 7.7
silt	%	56.0 ± 1.1	31.0 ± 6.8
sand	%	9.9 ± 1.9	48.8 ± 14.2
Dry weight	%	67.6 ± 1.9	68.2 ± 8.7
$CaCO_3$	%	9.0 ± 0.8	8.9 ± 2.9
pH_{H2O}		7.6 ± 0.1	7.9 ± 0.1
pH_{CaCl2}		6.9 ± 0.1	7.2 ± 0.1
EC	$\mu S\ cm^{-1}$	250 ± 181	187 ± 42
TOC	%	4.9 ± 0.4	8.4 ± 4.1
N_{soil}	%	0.34 ± 0.03	0.51 ± 0.25
P	(mg kg^{-1} dry soil)	3421 ± 264	814 ± 329
S	(mg kg^{-1} dry soil)	1270 ± 216	2353 ± 1390
Cu	(mg kg^{-1} dry soil)	150 ± 15	20 ± 10
Cr	(mg kg^{-1} dry soil)	246 ± 24	40 ± 15
Pb	(mg kg^{-1} dry soil)	211 ± 20	25 ± 14
Ni	(mg kg^{-1} dry soil)	50 ± 4	18 ± 5
Zn	(mg kg^{-1} dry soil)	1306 ± 120	143 ± 89
Cd	(mg kg^{-1} dry soil)	11.4 ± 1.0	1.4 ± 0.9
C:N		10 ± 0.7	11 ± 1.1
C:P		10 ± 1.0	69.2 ± 22.3

Table 4.2. Average values for the 0-15 (sediment topsoil: T) and the 15-30 cm (deeper sediment: D) layer for the uncovered plots (75 sampling points), significance of the difference with pair-wise comparison and short range variance based on the variogram

	Value for T	Value for D	Mean difference T-D	Short range variance
Cu (mg kg^{-1} dry soil)	149.6	160.2	-10.6	10.5
Cr (mg kg^{-1} dry soil)	244.6	263.6	-19.0	17.0
Ni (mg kg^{-1} dry soil)	50.2	52.5	-2.3	2.7
Zn (mg kg^{-1} dry soil)	1269.0	1301.0	-32.0	89.0
Cd (mg kg^{-1} dry soil)	12.8	11.4	1.4	0.9
TOC (%)	4.5	3.9	0.6	0.3
EC (µS cm^{-1})	179.9	200.8	-20.8	20.0
N_{soil} (%)	0.397	0.337	0.060	0.025
$CaCO_3$ (%)	8.8	9.0	-0.28	0.6
pH_{H2O}	7.6	7.7	-0.099	0.055
pH_{CaCl2}	6.9	7.1	-0.125	0.037

Table 4.4. Cd concentrations and total Cd contents per m² for the forest floor under oak and maple for the Meigem site and reference data from De Vos (1998). Means that are not significantly different are denoted with the same letter (Sidak multiple comparison of means at the 95% level of significance).

	Forest floor concentration (mg kg^{-1} DW)		Forest floor content (mg m^{-2})	
	Oak	Maple	Oak	Maple
December 2001	4.1 b	3.4 b	2.3 b	1.1 b
February 2002	7.0 a	7.1 a	3.7 a	1.8 a
April 2002	6.5 a	7.6 a	1.9 b	0.6 b
June 2002	7.3 a	8.0 a	1.3 b	0.3 b
Reference data	0.1-4.2	0.4-2.3	0.2-19.3	0.4-5.0

3.2. Forest floor decomposition

Forest floor accumulation started between October and December (Fig. 4.2), and only small changes were observed between December and February. Forest floor mass under oak was the highest at all sampling times. Net decrease of forest floor mass started only after February for oak, while for maple highest forest floor mass was observed in December. The average decomposed forest floor amounts per month for the period with net forest floor mass decrease were 87.8 g DW m^{-2} $month^{-1}$ or 36.6 g OC m^{-2} $month^{-1}$ for oak dominated plots and 60.0 g DW m^{-2} $month^{-1}$ or 24.7 g OC m^{-2} $month^{-1}$ for plots under maple. The difference between highest and lowest forest floor mass for oak (360.1 g DW m^{-2}) and maple (291.4 g DW m^{-2}) are comparable with measurements of yearly total tree leaf litter production for Flemish forests (Muys, 1995), ranging between 209 and 424 g DW m^{-2} for maple and oak dominated forests. The plots under maple had hardly any leaf litter left in summer, and forest floor consisted exclusively of the L layer, while forest floor included also an F-layer for several samples under oak.

Table 4.3. Forest floor mass (g DW m^{-2}) for the Meigem site and reference data for Flemish forests. Values in parentheses are standard deviations. (1) Neirynck et al. (2000), (2) Muys (1995), (3) data collected on 64 plots with oak and 4 plots with maple as dominant tree species in several forests during the Flemish forest soil inventory conducted between 1996 en 2000 (De Vos, 1998).

	Oak	**Maple**
Meigem, average litter layer biomass in winter	578.8 (228.9)	313.4 (56.5)
Meigem, average litter layer biomass in summer	218.7 (201.5)	22.0 (17.7)
Reference data for litter layer biomass in April (1)	2200-3500	1500
Reference data for litter layer biomass August (2)	719 - 12956	1052
Reference data for litter layer biomass (3)	497-16290	494-5388

Both for oak and maple dominated plots, no difference was detected by ANOVA in forest floor weight expressed as OC for the factor ‘topsoil’, while the effect of sampling period was highly ($p < 0.0001$) significant (Fig. 4.3). Forest floor decomposition was thus similar for the sediment topsoil and the covered topsoil. The variance in forest floor mass is larger for oak than for maple (Fig. 4.2, Fig. 4.3). The topsoil type did not influence the dry weight content (%DW) and the C:N ratio of the forest floor both for oak and maple for any of the sampling periods.

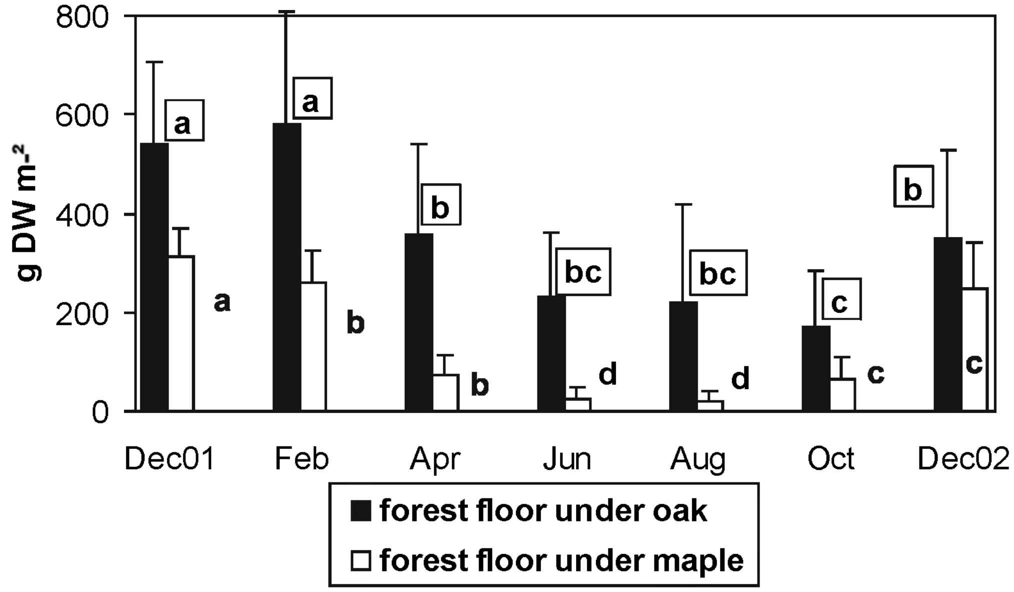

Fig. 4.2. Evolution of average forest floor mass (g DW m^{-2}) for oak and maple on the Meigem site. Standard deviation is indicated as error bar. Different letters indicate a significant difference for oak and maple dominated plots (Sidak multiple comparison).

A comparison was made between the pH_{CaCl2} for oak and maple forest floor samples collected in April. The pH was significantly lower ($p < 0.0001$) for the forest floor under oak (5.77 ± 0.25) than for the maple dominated plots (6.05 ± 0.16).

Especially the poplars lining the lanes had a significant contribution to the forest floor composition under oak and maple (11-22% DW in October and 3.7-4.2% in December). Forest floor samples in October and December 2002 were split up in a poplar fraction and the remaining fraction, and analysed separately. Effect of litter fraction and dominant tree species was tested with ANOVA. Only a significant influence of the factor 'litter fraction' ($p = 0.0043$ for Cd and $p = 0.0003$ for Zn) was detected, with average values for the poplar fraction being 3.8 mg Cd kg^{-1} DW and 241 mg Zn kg^{-1} DW higher than the remaining fraction.

Average Cr, Cu, Ni and Pb concentrations in the forest floor were respectively 49.3, 24.6, 19.4 and 33.8 mg kg^{-1} DW and were in the normal range for forest floors in Flemish forests (De Vos, 1998). However, Cd and Zn concentrations in the forest floor were slightly elevated. For Zn, no significant influence of dominant tree species or sampling period was detected with two-way ANOVA. Average forest floor concentration was 340 mg Zn kg^{-1} DW, while concentrations in Flemish forest soils ranged between 30 and 200 mg Zn kg^{-1} DW. As forest floor Zn concentration remained constant and forest floor mass decreased during decomposition, the Zn amount (mass x concentration: mg m^{-2}) in the forest floor decreased. Cd concentrations in the forest floor were not affected by dominant tree species, but concentrations significantly ($p < 0.001$) increased during decomposition (Table 4.4). The Cd amount in the forest floor was determined by both dominant tree species and sampling period without interaction between these factors, and was significantly higher in February than in the other sampling periods (Table 4.4).

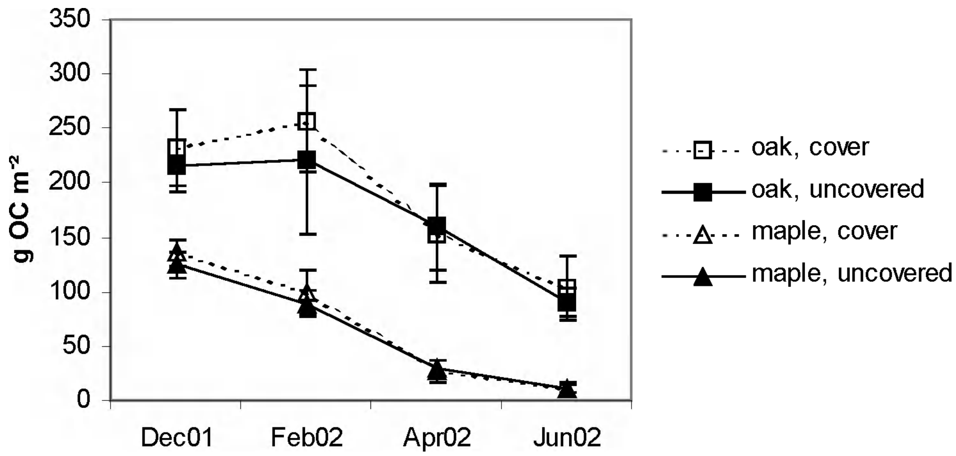

Fig. 4.3. Comparison of the evolution of the forest floor mass (g OC m^{-2}) for oak and maple on the Meigem site for cover topsoil and sediment topsoil. Standard deviation is indicated as error bar.

3.3. Earthworms

For the three sampled plots in the uncovered part of the landfill, the sampled earthworms consisted mainly (> 95% of total biomass) of epigeic species with a high potential for fast colonisation. Average earthworm biomass and density were 28.8 g FW m^{-2} and 117 individuals m^{-2} for the plot under maple, 17.0 g FW m^{-2} and 123 individuals m^{-2} for the plot under Ash and 13.0 g FW m^{-2} and 95 individuals m^{-2} for the unforested plot. Other litter decomposers such a snails and isopods were also abundant on the landfill.

Earthworms were collected on both a plot with a sediment topsoil and a cover topsoil for metal analysis. Adult *L. rubellus* were lower in weight than the juvenile earthworms. The Cd concentration in earthworms was not influenced by the factor ‘stage (juvenile *vs.* Adult)’, but concentrations were significantly lower ($p = 0.0011$) for the covered plot (37.9 mg Cd kg^{-1} DW) than for the uncovered soil (62.6 mg Cd kg^{-1} DW). In contrast, earthworm Zn concentrations were only influenced by stage ($p = 0.0006$) with higher values for the adult lumbricids (517 mg Zn kg^{-1} DW) than for the juvenile lumbricids (370 mg Zn kg^{-1} DW).

4. Discussion

The interest of this study lies in the fact that the studied landfill is a new soil at the time of disposal, where relatively uncontaminated foliar litter is decomposed on a polluted soil. Forest floor decomposition studies in polluted areas often focus on sites with high atmospheric input of metals. This pathway of pollution results in a forest floor with high metal concentrations (Fig. 4.4) and soil pollution that is localised in the upper cm of the soil profile (Bengtsson and Tranvik, 1989, Martin and Bullock, 1994). A developed soil profile is gradually polluted from the top layer down through atmospheric deposition. In the landfill studied here, the new soil profile is established at once over a larger thickness (> 150 cm in the studied site), starting from an initially reduced sediment layer. The soil profile is initially very homogeneous, and characterised by high metal, clay, organic carbon and $CaCO_3$ contents.

4.1. Soil forming processes in the sediment topsoil

In short, soil forming processes on the landfill site are characterised by apparently slow dynamics. Relative to the short range variance, the higher Cd, TOC and N_{soil} and the lower value for pH_{CaCl2} in the uncovered sediment topsoil are the main trends. Results for TOC and N_{soil} indicate a net organic carbon increase in the sediment topsoil. C accumulation in the soil was also reported by Peeters and van den Berg (1999) 26 years after afforestation of a polluted dredged sediment landfill, by Barajas Aceves et al. (1999) for soils affected by mine waste deposits and by Dai et al. (2004a) 70 years after metallurgic waste disposal. C accumulation might be related to variation in microbial communities and microbial activity due to metal contamination (Barajas Aceves et al., 1999; Dai et al., 2004a) or to stabilization of organic matter by calcium (Gillet and Ponge, 2002).

Organic matter mineralization is a major cause of acidification in soils, leading to a higher availability of metals. The rate and impact of soil decalcification and subsequent acidification must be further monitored, as it results in higher bioavailability (Ma and Van der Voet, 1993; Tack et al., 1998) and leaching risks of metals in polluted soils (Tack et al., 1999). The higher Cd concentration in the topsoil is remarkable, and further research may explain this observation.

4.2. Forest floor decomposition rates and metal concentrations

Average forest floor mass on the landfill in the first week of August was low compared to references for forests (Table 4.3), and revealed low oak and maple forest floor accumulation. Especially for oak relatively fast forest floor decomposition was observed. Probably the optimal soil nutrient status of both covered and uncovered topsoil was responsible for this feature, in contrast with Flemish forest soils generally characterised by acid conditions (Muys, 1995). The net yearly forest floor disappearance is comparable with values of of 338 g m^{-2} in a soil with mull humus reported by Van der Drift (1963). The loss in dry weight for the forest floor in a pedunculate oak stand with a mull litter type was only 17.4% after 6 months (Williams and Gray, 1974), while in Meigem a loss of 43% after 6 months was measured for oak. Forest floor quantities accumulated on forest soils around smelters where forest floor decomposition is inhibited, are also high relative to data for the Meigem site: Martin et al. (1982) reported forest floor quantities for a polluted oak-hazel woodland of 177 g m^{-2}, 3422 g m^{-2} and 6964 g m^{-2} for the L, F and H layer respectively.

The application of a thin uncontaminated cover topsoil on the dredged sediment did not result in a faster or slower forest floor decomposition. Comparison is allowed as no significant differences were observed in both soil (Table 4.1, expressed as gravimetric dry weight content) and forest floor water content between covered and uncovered plots.

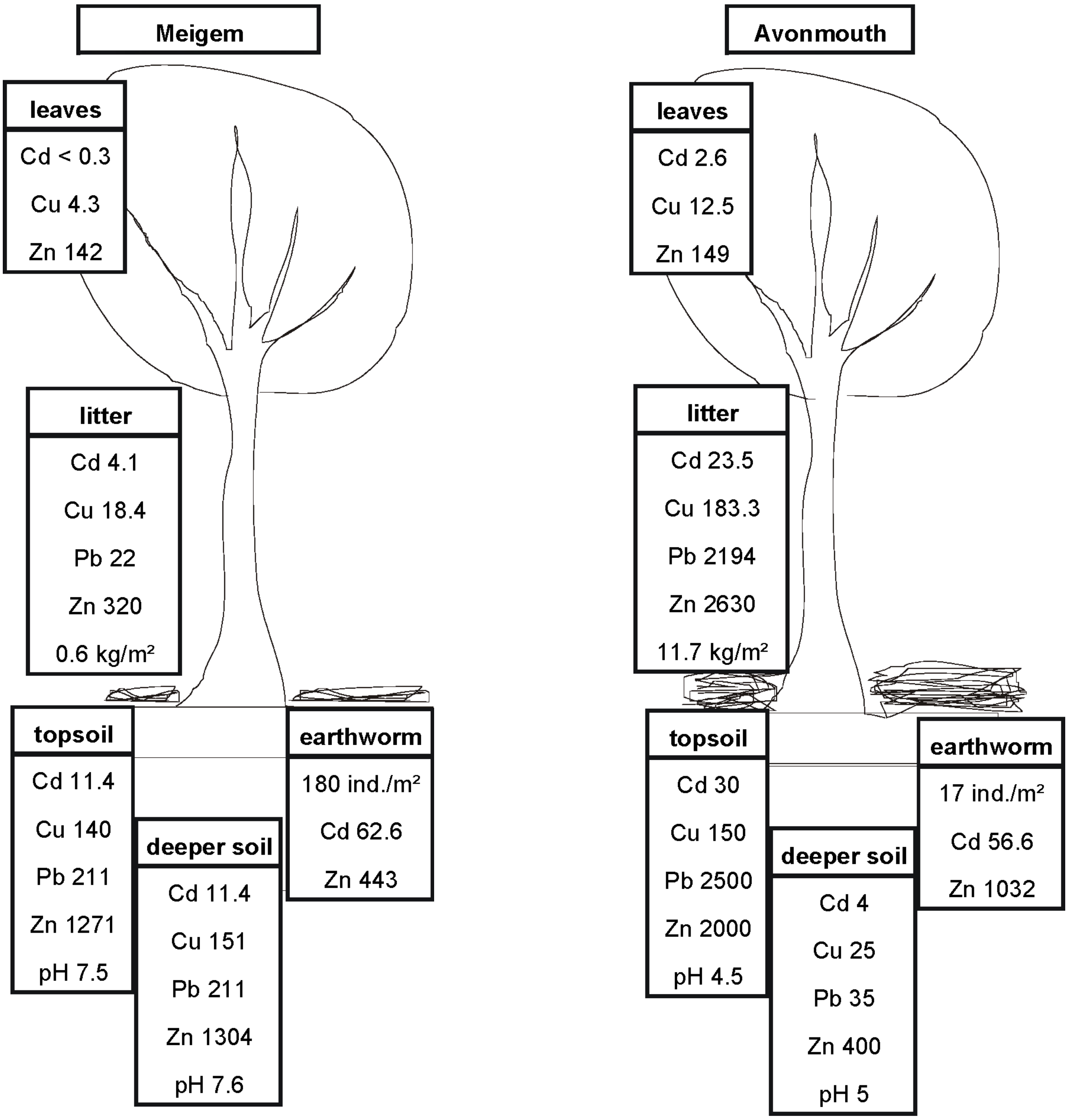

Fig. 4.4. Comparison of several compartments in forests for Meigem and the Avonmouth area (Martin and Bullock, 1994). All values except pH are expressed as mg kg^{-1} DW unless otherwise indicated. All data are expressed on a dry weight basis. Both sites are classified as heavy clay soils.

Forest floor metal concentrations for Cd and Zn are slightly lower than maximum allowable concentrations without adverse effects reported by Bengtsson and Tranvik (1989). Concentrations are substantially lower than values reported by Martin and Bullock (1994) for oak woodlands near zinc smelters with extreme litter accumulation (39-112 mg Cd kg^{-1} DW and 1900-3500 mg Zn kg^{-1} DW) and by Grelle et al. (2000) for the most polluted sites near a smelter with undecomposed litter layers (300 mg Cd kg^{-1} DW, 30000 mg Zn kg^{-1} DW and 5000 mg Pb kg^{-1} DW). Forest floor metal concentrations were not in the toxic range for soil invertebrates and thus no detrimental effects for soil invertebrate abundance as observed by Spurgeon et al. (1996) and Grelle et al. (2000) are expected.

The slightly elevated Cd and Zn concentrations in the forest floor for the Meigem site is at least partly a consequence of elevated concentrations in poplar leaves in the forest floor, but might also be due to topsoil-litter mixing or atmospheric deposition, or a combination of these factors. Foliar concentrations measured in poplar samples taken in the second half of August were 12.2 mg Cd kg^{-1} DW and 640 mg Zn kg^{-1} DW, while concentrations in oak and maple leaves were < 0.35 mg Cd kg^{-1} DW and < 150 mg Zn kg^{-1} DW (Vandecasteele and De Vos, 2002). Both for plots under oak and maple, the total metal amount in the forest floor clearly decreased during forest floor decomposition. The forest floor obviously does not act as a sink for metals on this dredged sediment landfill.

4.3. Earthworm biomass and effect of the cover topsoil on metal concentrations in earthworms

Sixteen years after landfilling the dredged sediment, the uncovered part of the landfill was found to be colonised by mainly epigeic earthworms. Earthworm biomass was found to be low to moderate relative to the soil properties, but this has to be explained by the absence of more heavy endogeic and anecic species due to their low colonisation rate (Vandecasteele et al., 2004b).

In theory, the cover topsoil results in an uncontaminated environment for soil organisms. For the earthworm *Lumbricus rubellus*, the application of a cover topsoil did result in lower body concentrations for Cd, but body concentrations were still slightly higher than for reference situations (Heikens et al., 2001). Dai et al. (2004b) concluded that differences in metal bioaccumulation between soil feeding and leaf litter feeding earthworm species on polluted soils was related to food selectivity. From an ecological point of view, forest floor quality and pollution status is more relevant for some soil organisms than soil pollution status (Hopkin, 1994). The forest floor metal concentrations at Meigem were slightly higher than normal concentrations (Table 4.4), and may have affected metal concentrations in the litter-dwelling species *L. rubellus*. In conclusion, the application of an uncontaminated cover topsoil is a management option which does not seem to be essential in the case of dredged sediment landfills.

4.4. Conclusions

Establishing a forest on dredged sediment landfills without a thick covering layer of uncontaminated soil or without the use of impermeable liners and capping layers allows for taking full profit of the high nutrient status of the dredged sediment substrate. Nutrient status favours tree growth and allows for reforestation and thus an integration of dredged sediment landfills into the landscape. However caution is advised in notation to possible adverse effects of metals on forest functioning. We conclude from the observations in Meigem that 10 years after afforestation of a polluted but fertile soil, forest floor decomposition with normal or slightly elevated metal concentrations in the forest floor is normally functioning.

Chapter 4.2. Earthworm biomass as complementary information for risk assessment of metal biomagnification: a case study for dredged sediment-derived soils and polluted floodplain soils*

The important role of earthworms in the biomagnification of metals in terrestrial ecosystems is widely recognised. Differences in earthworm biomass between sites are mostly not accounted for in ecological risk assessment. These differences may be large depending on soil properties and pollution status. A survey of earthworm biomass and colonisation rate was carried out on dredged sediment-derived soils (DSDS). Results were compared with observations for the surrounding alluvial plains. Mainly grain size distribution and time since disposal determined earthworm biomass on DSDS, while soil pollution status of the DSDS was of lesser importance. Highest earthworm biomass was observed on sandy loam DSDS disposed at least 40 years ago.

1. Introduction

Metals may enter the food web via soil dwelling organisms living on dredged sediment-derived soils (DSDS) and contaminated floodplains. Earthworms constitute the largest terrestrial faunal biomass. In the transfer of pollutants towards other trophic levels, they occupy a key position (Kreis et al., 1987, Granval and Muys, 1995). Earthworms were found to have a high potential for Cd accumulation in polluted floodplains (Hendriks et al., 1995). They are considered useful for assessing metal pollution in soils (Menzie et al., 1992) because earthworm biomass and abundance seem more sensitive to pollution in comparison with other indicator taxa (Spurgeon et al., 1996). The presence and abundance of earthworms can be a determining factor for the occurrence of higher organisms. Presence of Little Owl (*Athene noctua*) in Flanders could be predicted based on landscape and soil characteristics. The highest occurrence of Little Owl was on locations with soils optimal for large earthworm populations (Van Nieuwenhuyse et al., 2001).

Metal pollution can induce two major effects on the ecosystem level: (a) accumulation of e.g. Cd can lead to risks of secondary poisoning, while (b) earthworms disappear already at

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low levels of soil Cu which can cause food scarcity for earthworm predators (Abdul Rida, 1992; Klok et al., 2000).

A major factor for both metal uptake and toxicity (Ma et al., 1983), and earthworm abundance (Muys and Granval, 1997) is the soil pH. The pH of the soil material decreases during gut passage in *L. terrestris* Linnaeus (Heine and Larink, 1993). Brzóska and Moniuszko-Jakoniuk (1998) found a strong interaction between Cd and Ca in feed, resulting in a lower toxicity of Cd at high Ca intakes. Earthworms are very sensitive to Cu (Ma, 1982; Ma et al., 1983), but Cu accumulation by earthworms is particularly unpredictable (Edwards et al., 1998). Besides pH, soil characteristics like OM content and CEC are influencing metal availability and uptake by *L. rubellus* Hoffmeister (Ma, 1982; Ma et al., 1983). Beyer et al. (1990) presumed that the acidity (pH 3.0-5.5) of the dredged sediment substrate at four confined disposal facilities was responsible for the absence of earthworms, as on an older landfill with a higher pH, earthworms were found.

Modelling of uptake kinetics and accumulation of metals by earthworms as a biological reference system and as a key process in trophic transfer for risk assessment on polluted sites is a current topic (Abdul Rida, 1992; Beyer and Stafford, 1993; Kooistra et al., 2001). *In situ* observation of biomass and population dynamics of earthworms can be a means to determine long term effects. The importance of earthworms for metal biomagnification in terrestrial ecosystems is widely recognised. Knowledge of earthworm metal tissue concentrations is essential for risk assessment of metal biomagnification. Large differences in earthworm biomass may be encountered on different locations as a function of soil properties and soil pollution status, but are also related to the rate of colonisation/recolonisation. Despite this, earthworm biomass is mostly not incorporated in site-specific ecological risk assessment and homogeneous earthworm biomass is usually assumed.

To allow for risk assessment on DSDS and floodplains affected by overbank sedimentation, we will focus in this paper on earthworm biomass and population dynamics relative to the surrounding unaffected alluvial plains. The pollution level of the floodplain soils and several characteristics of the DSDS (time since disposal, pollution level, physicochemical properties) will be included in the analysis. The originality of this study lies in the fact that the studied DSDS were new soils at the time of disposal and are expected to be slowly invaded by earthworms. We feel that most research about earthworm populations in polluted areas focuses on superficially located pollution, concentrated in the upper centimeters of the soil profile. This is the case for forests around smelters (Bengtsson and Tranvik, 1989, Martin and Bullock, 1994), floodplain soils (Hendriks et al., 1995; Ma et al., 1997), areas polluted by agricultural activities (Ma, 1988; Filser et al., 1995; Didden, 2001), or by use of timber preservatives (Yeates and Orchard, 1994). All these case studies have in common that a developed soil profile has gradually been polluted from the top layer, existing earthworm populations were subjected to an increasing environmental stress and consequently, earthworm population changes might be measured. In such locations, soil pollution is relatively heterogeneous and organisms can survive through avoidance (Eijsackers, 1987; Ma, 1988; Yeates and Orchard, 1994).

In the DSDS studied here, the polluted soil profile is established at once over a larger thickness (> 80 cm at least in this study). The reduced sediment was hydraulically pumped on the site and caused the earthworms from the original soil to disappear. During development of the sediment layer, subsoil is not a cleaner refuge for earthworms. Suter et al. (2000) stress the importance of selecting adequate reference locations for soil biological surveys since high variation in quantities of soil biota from location to location were regularly observed. We will make an attempt to link earthworm biomass and density with ecological and pollution factors and use these data for a general risk assessment.

2. Materials and methods

2.1. Study area

All sampling plots in this paper were located along the Scheldt and Leie river and the Canal Ghent-Bruges (Fig. 4.5). The presented study was executed in two steps. In a first exploratory step, total biomass and density of earthworms, and the relative distribution over the ecological categories were compared for three unaffected alluvial soils (ALL), four alluvial soils polluted due to overbank sedimentation (overbank sedimentation zones = OSZ) and five soils affected by dredged sediment disposal (dredged sediment-derived soils = DSDS) (Table 4.5, Table 4.6). Plots were selected to allow for pair-wise comparison of sites of a different soil type located close to each other. In the second step, the influence of soil physical conditions, pollution status and general landfill characteristics on the biomass and density of earthworms on 19 DSDS (Table 4.7, Table 4.8) was determined. Five of the 19 sites were already sampled for the research goal of step 1. All sampled alluvial soils were under pasture, while the sampled DSDS were used for pasture or forestry, or developed as thicket.

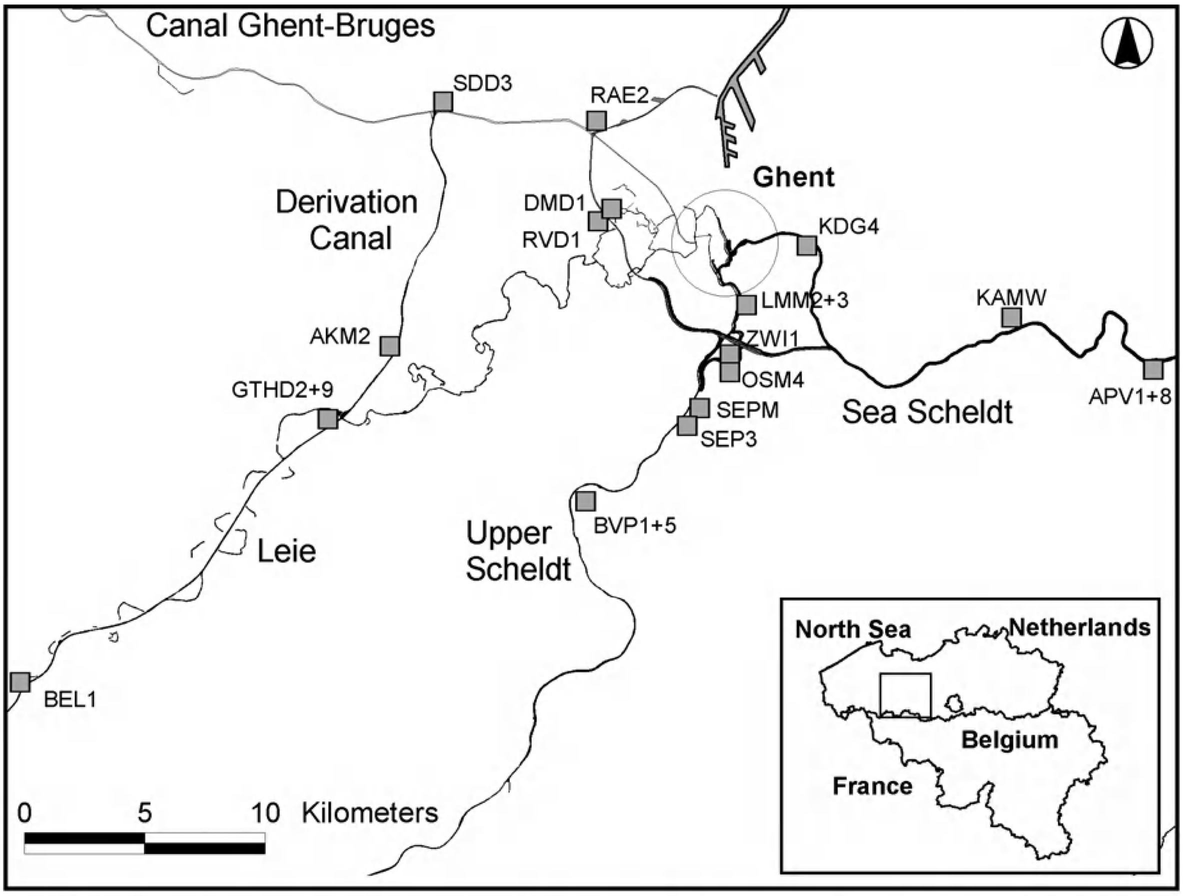

Fig. 4.5. The study area with the sampled locations. Each sampled location is referred to with a unique code based on a local toponym.

2.2. Earthworm and soil sampling

The ecological categories of earthworms could be distinguished functionally or evolutively (Bouché, 1972; Muys and Lust, 1992). Functionally, three groups are distinguished: epigeic, endogeic and anecic earthworms. The epigeic earthworms live in compost or in litter, and are adapted through more expressed pigmentation. The litter constitutes their food and the function of this group is to fragment and digest the soil organic matter. The endogeic earthworms burrow horizontal galleries into soils rich in humus. This group consists of humus feeders which are not involved in the litter decomposition. It is however an important group for the bioturbation of the upper 30 cm of the soil. The anecic earthworms burrow vertical galleries in the soil and take up food at the soil surface, especially at night.

Earthworm sampling was done on all sites with six replicates between September and November 2001 (13 sites), in April 2002 (2 sites) and between September and November

2002 (11 sites) according to the combined method of Bouché and Aliaga (1986). At the time of sampling, soil moisture content was at field capacity, and differences in soil moisture conditions between sites can be addressed to soil type and hydrological conditions of the sites. When selecting the sampled sites for the DSDS, care was taken to stay at least 15 m from the landfill edge to exclude fast migration of earthworms as a confounding factor influencing the biomass. As no litter layer was found on any site, sampling of the litter layer was unnecessary. Sprinkling the two nested subplots (0.5 m^2 each) with formalin solutions (2 times 10 L of a 0.05% solution and 2 times 10 L of a 0.1% solution at 10 minute intervals) yielded the first earthworm fraction. The second fraction was obtained after wet washing and sieving 20 dm^3 of a soil sample (0.1 m^2) from every subplot after pretreatment during 48 h in a solution of 10 L water, 100 mL sodium hexametaphosphate and 800 mL formalin. Both earthworms from formalin application and wet soil sieving were preserved in a 37% formalin solution. Formalin extraction and formalin conservation caused irritation and intense movements resulting in a certain defaecation. Within a week after sampling, total earthworm biomass and density was determined after drying for 1 min on a filter paper at room temperature. Collected earthworms from all fractions were identified, counted and weighed. Fresh earthworm biomass in g/m^2 was calculated according to:

[(mass earthworms soil × 2) + (mass earthworms soil after sieving × 10)]

Juveniles, parts of earthworms and non-identifiable species were proportionally attributed to species. To reduce the time-consuming species identification step, we limited species identification to a number of replicates until at least 40 g of field-collected fresh weight (FW) was determined. For sites with less fresh material, all subsamples were used. Earthworm biomass per ecological category (Bouché, 1972) was calculated for a general description of the sampled earthworm communities.

After the first sampling on DSDS (step 1), it was concluded that additional soil core sampling after formalin extraction did not result in additional information about species diversity or ecological categories. Only a low biomass gain (at most 16%) was obtained by additional soil core sampling. Furthermore the soil sampling, transportation, dispersion and sieving is a very time-consuming step with an important impact on the sampled area. For this reason, the determination of the relation between soil characteristics and earthworm populations on DSDS (step 2) was based on formalin extraction data only. Data for both formalin extraction and soil sampling were available for APV1 and APV8.

Before earthworm sampling started, soil samples (0-20 cm horizon) for separate chemical analysis were collected outside the sampling frame, on a distance of 20 cm. The methods used for chemical soil analysis are described in Chapter 1.1. Soil total contents of Cd, Cr, Cu, Ni, Pb, S, P and Zn are actually pseudo-total *aqua regia* extractable contents measured with ICP-AES after microwave digestion. Soil physical properties on the DSDS were estimated based on measurement of soil bulk density, penetration resistance and saturated hydraulic conductivity (Ksat) and calculation of the ripening factor. On each DSDS, four core samples of 100 cm^3 were taken at the soil surface and used for determination of saturated hydraulic conductivity with an ICW permeameter (Eijkelkamp Agrisearch Equipment, Giesbeek, The Netherlands) and bulk density calculation (ISO 11272). At four points per DSDS, penetration resistance was measured when soil was at field capacity using an Eijkelkamp penetrometer (Eijkelkamp Agrisearch Equipment, Giesbeek, The Netherlands), capable of recording values at intervals of 1 cm to a depth of 80 cm. A 1 cm^2 base area cone was used with an angle of 60°. The penetration in the soil occurred at a rate of 20 ± 5 mm/s. For the measurements of the penetration resistance, the maximum values for the 0-40 cm (measurements in 1 cm intervals) layer for the four replicates was used as value for the site. For bulk density and permeability, average resp. median values for the four replicates were used. For all soil samples on DSDS, the ripening factor as used by de Haan et al. (1998) was calculated as a measure of the ripening rate.

Ripening factor = $[A - p \times (100 - L - H)] / [L + b \times H]$ with:

A: water percentage (g/100 g DM)

H: organic matter percentage (g/100 g DM)

L: clay fraction (g/100 g DM)

b: ratio between water absorption capacity of organic matter and absorption capacity of clay (usually b = 3)

p: moisture bound by non-colloidal material (usually p = 0.2 g/100 g non-colloidal material)

2.3. Data handling and statistics

All earthworm biomass data were expressed on a fresh weight base (g/m^2 FW), and abundance was expressed as N/m^2. As individual earthworm weight varied between <0.01 g FW and > 4 g FW for the sampled species, earthworm biomass rather than earthworm density was chosen to represent food availability for predators. For two replicates on the APV8 site, earthworm biomass was very low. The reason for this observation was unknown but the site was recently affected by tree cutting, which may have caused soil compaction. For the RVD1 site, two replicates were situated on soils with deviant soil properties relative to the other four replicates. For these two sites, deviant replicates were excluded from further analysis.

The three soil categories (ALL, OSZ and DSDS) were compared for soil properties and earthworm parameters with one-way ANOVA after grouped variables were tested for normality and homoscedasticity. Variables included in the analysis were earthworm biomass (expressed as g FW/m^2) and density (expressed as N/m^2), soil pollution status (Cr, Cu, Cd, Pb and Zn) and soil properties (gravimetric dry matter content (DM), TOC, pH, total S, total P, $CaCO_3$, N, grain size distribution). Data for $CaCO_3$, Cd, Cr, Zn, Cu were log-transformed prior to ANOVA. Additionally, the heaviest adult *L. rubellus* was recorded as a measure of the appropriateness of sites for earthworms.

A linear model was built for interpretation of DSDS earthworm biomass as a function of soil properties. To test which factors have an impact and to assess their magnitude a multivariate regression model was built in two steps. First, a reference model was constructed with the variables "grain size" and "time since disposal" (TSD), because these factors were recognised as dominant during prior exploratory data analysis. This second step was aimed to assess whether on top of this basic model other additional factors were important. First, the residuals of the reference model plotted as a function of the other soil variables and pollution status were inspected to check for patterns. Next, a stepwise regression at a significance level of 0.05 searched for the best subset of variables against which residuals showed a pattern. For this model only the final result is shown here. Full details of the model are given in the 'results'-section. During statistical model building, both RVD1 and RAE2 sites were observed to be outliers. Data on biomass were square root-transformed before statistical analysis.

3. Results

3.1. Comparison between DSDS and surrounding alluvial soils

Unaffected alluvial soils (ALL), dredged-sediment derived soils (DSDS) and overbank sedimentation zones (OSZ) did not significantly differ in grain size distribution (clay, silt, sand), EC and soil organic matter content (SOM) (Table 4.5, results for silt and sand not shown). Gravimetric water content was significantly ($p < 0.001$) higher for OSZ, while values for DSDS and ALL were comparable. P concentrations in DSDS were higher than in OSZ and ALL ($p < 0.001$). Pb concentrations were higher in DSDS and OSZ compared with ALL ($p < 0.001$). For $CaCO_3$ ($p < 0.0001$), Cd ($p = 0.0009$), Cu ($p < 0.0001$), Zn ($p < 0.0001$), Cr (p =

0.0003) and S ($p < 0.0001$), concentrations in DSDS were significantly higher than for OSZ, which in turn were significantly higher than for ALL (Table 4.6). For earthworm density, only a significant difference was found between ALL and OSZ ($p = 0.032$). However, for earthworm biomass, values for ALL were significantly higher ($p < 0.0001$) than values for both DSDS and OSZ (Table 4.5). In conclusion, general soil characteristics between the three soil types are similar, but chemical properties of both DSDS and OSZ deviate from the ALL characteristics.

Biomass of earthworms for the selected pairs of sites is shown in Fig. 4.6b. Biomass is highest for unaffected alluvial soils, intermediate for polluted floodplains and low for the DSDS. Standard deviation (SD) on six replicates is for most sites considerable. When results on density of earthworms is displayed (Fig. 4.6a), differences between the sites are less clear, but most DSDS are characterised by low densities.

Table 4.5. Properties of the sampled unaffected alluvial soils (ALL), the dredged sediment-derived soils (DSDS) and overbank sedimentation zones (OSZ). Earthworm density and biomass (based on a combination of formalin extraction and soil core excavation) are given in the last columns. Values in parentheses are standard deviations for six replicates. NA: not assessed

Site	River	Soil type	Land use	clay %	TOC %	EC µS/cm	pH-H_2O	$CaCO_3$ %	DW soil %	Density N/m²	Biomass g/m²
BEL1	Leie	DSDS	willow brushwood	39 (5)	8.1 (1.5)	286 (56)	7.6 (0.2)	10.6 (4)	61.1 (4.2)	165 (70)	20.9 (8.6)
		ALL	oak plantation	33 (4)	8.5 (1.2)	224 (88)	5.7 (0.2)	1.9 (0.3)	67.2 (3.6)	164 (67)	88.9 (26.7)
BVP1	Upper Scheldt	DSDS	willow brushwood	28 (5)	3.9 (1.3)	246 (95)	7.5 (0)	10.3 (1.3)	63 (1.9)	166 (41)	20.9 (8.6)
		ALL	pasture	26 (1)	6.1 (0.6)	197 (21)	6.9 (0.2)	2.2 (0.8)	63.6 (2)	485 (253)	157.5 (141)
SEPM	Upper Scheldt	DSDS	alder plantation	32 (1)	13.8 (0.6)	177 (20)	7.5 (0.1)	5.3 (0.9)	64.5 (2.4)	139 (41)	13.4 (5.5)
		OSZ	pasture	35 (1)	13 (1.2)	198 (14)	6.7 (0.5)	2.5 (0.4)	57.8 (5.5)	119 (38)	31.1 (10)
DMD1	Leie	DSDS	pasture	15 (4)	10 (3.5)	132 (28)	7.4 (0.1)	4.3 (0.8)	72.4 (3.8)	35 (13)	8.2 (2.9)
		OSZ	pasture	25 (2)	10.9 (0.5)	249 (54)	6.5 (0.2)	2.3 (0.4)	53 (3.7)	374 (185)	78.9 (44.8)
GTH9	Leie	DSDS	elder brushwood	42 (3)	8.7 (1.9)	257 (37)	7.4 (0.1)	11.1 (1)	63.5 (2.7)	45 (45)	9.9 (11.8)
		OSZ	pasture	19 (5)	1.8 (1)	312 (181)	7.9 (0.3)	4.4 (1.9)	NA	215 (95)	47.2 (18.7)
SEP3	Upper Scheldt	OSZ	pasture	36 (3)	10.7 (1.4)	227 (40)	7.3 (0.2)	4.6 (1.1)	66.4 (3.6)	30 (44)	10.8 (15.2)
		ALL	pasture	31 (4)	5.1 (2.2)	219 (54)	7.2 (0.2)	2.4 (0.4)	72.1 (5.9)	48 (31)	24.9 (9.4)

Table 4.6. Elemental contents in the sampled unaffected alluvial soils (ALL), the dredged sediment-derived soils (DSDS) and overbank sedimentation zones (OSZ). Cd, Cu Cr, Pb, Zn, P and S are *aqua regia*-extracted and are expressed as mg kg^{-1} dry soil. Values in parentheses are standard deviations for six replicates

Site	Soil type	Cd mg/kg	Cr mg/kg	Cu mg/kg	Pb mg/kg	Zn mg/kg	P mg/kg	S mg/kg
BEL1	DSDS	9.2 (2.7)	216 (31)	209 (30)	216 (36)	1558 (269)	5279 (988)	1905 (300)
	ALL	1.6 (0.3)	88 (13)	47 (8)	122 (22)	276 (39)	1364 (215)	1337 (218)
BVP1	DSDS	6.8 (0.9)	320 (16)	156 (53)	126 (21)	810 (178)	2870 (318)	1780 (382)
	ALL	0.9 (0.1)	66 (3)	21 (2)	43 (5)	132 (11)	976 (40)	1092 (106)
SEPM	DSDS	11.4 (2)	269 (33)	136 (12)	408 (35)	2053 (197)	2105 (165)	1692 (177)
	OSZ	6.2 (0.5)	156 (11)	89 (12)	278 (29)	1112 (198)	1182 (126)	1530 (142)
DMD1	DSDS	5.7 (2.6)	429 (232)	89 (39)	197 (59)	889 (413)	2034 (614)	1611 (405)
	OSZ	1.4 (0.2)	83 (7)	120 (14)	518 (74)	352 (18)	1438 (167)	1409 (80)
GTH9	DSDS	23 (2.2)	515 (43)	332 (29)	474 (36)	2742 (163)	5087 (334)	2116 (376)
	OSZ	4 (3.4)	118 (82)	65 (53)	104 (83)	524 (433)	1306 (817)	629 (389)
SEP3	OSZ	34.3 (12.2)	816 (309)	106 (21)	341 (103)	2042 (495)	3485 (969)	1603 (88)
	ALL	0.7 (0.1)	69 (10)	20 (6)	80 (59)	149 (15)	1974 (789)	844 (228)

Table 4.7. Soil physical properties, earthworm density and biomass for the dredged sediment-derived soils (DSDS) where earthworms were sampled. Earthworm density and biomass are based on formalin extraction only. Highest body weight for an adult *L. rubellus* is recorded in the column '*L. rubellus*'. Values in parentheses are standard deviations for six replicates (TSD = time since disposal, P.R. = penetration resistance, NA = not assessed)

Site	River/Canal	Land use	Sampling	Clay	DW soil	Density	Biomass	*L. rubellus*	TSD	P.R.	Bulk density	Permeability	Ripening
				%	%	N/m²	g/m²	g	(year)	MPa	kg/m³	cm/day	factor
APV8	Sea Scheldt	poplar plantation	Fall 2001	14 (2)	81.9 (6.5)	244 (26)	128.0 (56.6)	1.800	70	2.44	1016	10	0.21
DMD1	Leie	pasture	Fall 2001	15 (4)	NA	28 (13)	7.6 (3.1)	1.633	30	2.91	1287	5	0.35
SDD3	Canal Ghent-Bruges	poplar plantation	Spring 2002	20 (3)	80.6 (2.9)	97 (25)	118.5 (56.7)	2.587	60	2.16	1167	213	0.30
KDG4	Sea Scheldt	pasture	Fall 2002	21 (7)	79.3 (6.9)	55 (29)	32.7 (31.3)	2.268	14	3.09	1296	1529	0.24
OSM4	Upper Scheldt	pasture	Fall 2002	25 (4)	76 (1.6)	48 (9)	48.1 (8.1)	0.487	40	2.42	1140	24	0.23
APV1	Sea Scheldt	poplar plantation	Fall 2001	26 (2)	61.1 (10.8)	154 (60)	59 (30)	1.110	70	0.83	1193	36	1.04
BVP1	Upper Scheldt	willow brushwood	Fall 2001	28 (5)	63 (1.9)	156 (40)	20 (8.3)	1.006	6	0.75	1099	4	0.96
SEPM	Upper Scheldt	alder plantation	Fall 2001	32 (1)	64.5 (2.4)	117 (46)	11.4 (6.1)	0.421	40	1.06	1013	4	0.43
RVD1	Canal Ghent-Bruges	willow brushwood	Fall 2002	41 (2)	54.5 (2.5)	53 (25)	11.3 (4.5)	0.569	25	2.07	867	4	0.72
AKM2	Leie	ash plantation	Fall 2002	35 (3)	69.8 (4.7)	148 (51)	19.2 (10.7)	0.716	16	1.56	1058	4	0.50
LMM3	Upper Scheldt	ash plantation	Fall 2001	37 (1)	53.8 (3.1)	381 (216)	33.5 (9.3)	0.728	40	1.6	899	56	0.84
GTH2	Leie	willow brushwood	Fall 2002	37 (1)	60.3 (6.8)	110 (46)	8.5 (2.8)	0.461	22	1.12	989	30	0.70
LMM2	Upper Scheldt	ash plantation	Fall 2001	38 (3)	54.8 (3.9)	145 (39)	9.8 (4.4)	1.062	40	2.46	896	181	0.71
BEL1	Leie	willow brushwood	Fall 2001	39 (5)	NA	125 (52)	15.7 (7.1)	0.586	16	3.5	800	356	0.67
KAMW	Sea Scheldt	pasture	Fall 2002	40 (3)	57.7 (3.2)	89 (34)	33.1 (8.1)	0.614	70	1.12	850	83	0.68
BVP5	Upper Scheldt	willow brushwood	Fall 2002	41 (2)	66.1 (5.6)	113 (42)	10.8 (5.3)	0.278	6	1.04	995	29	0.55
GTH9	Leie	elder brushwood	Fall 2001	42 (3)	63.5 (2.7)	32 (22)	7.3 (6.8)	0.963	25	1.14	882	490	0.55
RAE2	Leie	willow brushwood	Spring 2002	43 (2)	62.7 (2.9)	3 (5)	0.4 (0.7)	NA	6	0.87	743	595	0.67
ZWI3	Upper Scheldt	abandoned arable land	Fall 2002	45 (2)	69 (0.9)	175 (31)	27.2 (6.8)	0.497	20	2.06	962	79	0.43

Table 4.8. Properties of the dredged sediment-derived soils (DSDS) selected for earthworm sampling. Cd, Cu, Cr, Pb, Zn, P and S are *aqua regia*-extracted and are expressed as mg kg^{-1} dry soil, TOC, $CaCO_3$ are expressed in %. Values in parentheses are standard deviations for six replicates

Site	$CaCO_3$	TOC	EC	pH-H_2O	Cd	Cr	Cu	Pb	Zn	P	S
	%	%	µS/cm		mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
APV8	2.7 (1.1)	2.8 (0.4)	154 (17)	7.5 (0.2)	1.6 (0.5)	66 (18)	31 (9)	62 (10)	350 (67)	841 (146)	624 (187)
DMD1	4.3 (0.8)	10 (3.5)	132 (28)	7.4 (0.1)	5.7 (2.6)	429 (232)	89 (39)	197 (59)	889 (413)	2034 (614)	1611 (405)
SDD3	6 (1.5)	1.3 (0.6)	1447 (1519	7.6 (0.2)	3.3 (1.2)	109 (36)	53 (22)	98 (45)	513 (285)	1242 (287)	1545 (2447)
KDG4	4.4 (2)	3.4 (1.6)	170 (73)	7.5 (0.2)	6.3 (5.5)	600 (474)	319 (221)	235 (115)	1321 (936)	4124 (2449)	1373 (904)
OSM4	5 (1.1)	10.7 (1.3)	193 (69)	7.6 (0.2)	4.9 (0.9)	378 (60)	226 (48)	457 (71)	1805 (348)	2592 (518)	1735 (148)
APV1	6.3 (0.7)	5.2 (0.8)	235 (19)	7.5 (0.1)	3.2 (0.2)	157 (28)	96 (7)	161 (5)	661 (30)	1536 (77)	849 (91)
BVP1	10.3 (1.3)	3.9 (1.3)	246 (95)	7.5 (0)	6.8 (0.9)	320 (16)	156 (53)	126 (21)	810 (178)	2870 (318)	1780 (382)
SEPM	5.3 (0.9)	13.8 (0.6)	177 (20)	7.5 (0.1)	11.4 (2)	269 (33)	136 (12)	408 (35)	2053 (197)	2105 (165)	1692 (177)
RVD1	2.7 (0.9)	11.4 (1.1)	1490 (894)	6.7 (0.3)	27.7 (0.7)	1100 (125)	661 (29)	1077 (143)	5686 (170)	7146 (286)	10300 (6492)
AKM2	8.3 (0.4)	5.4 (1.3)	198 (30)	7.2 (0.1)	13.6 (1)	242 (15)	138 (6)	224 (18)	1310 (105)	4328 (1429)	1253 (122)
LMM3	13 (1.8)	10.2 (1.3)	269 (36)	7.4 (0.1)	28.7 (1.7)	1933 (108)	229 (10)	405 (10)	2953 (114)	5724 (343)	2334 (91)
GTH2	9.4 (0.7)	8.2 (1.1)	359 (78)	7.2 (0.1)	12.4 (0.9)	227 (13)	133 (18)	283 (128)	1339 (108)	3849 (1077)	1452 (259)
LMM2	5.3 (0.6)	12.6 (1.1)	277 (23)	7.1 (0.1)	18.2 (3.2)	1121 (240)	159 (19)	347 (29)	2243 (275)	3891 (420)	2017 (129)
BEL1	10.6 (4)	8.1 (1.5)	286 (56)	7.6 (0.2)	9.2 (2.7)	216 (31)	209 (30)	216 (36)	1558 (269)	5279 (988)	1905 (300)
KAMW	4.9 (0.7)	10.3 (0.9)	361 (57)	7.4 (0.1)	4.3 (0.5)	422 (15)	210 (49)	336 (133)	1205 (69)	3145 (842)	1622 (199)
BVP5	11.5 (0.7)	6.6 (1.3)	286 (51)	7.5 (0)	13 (1.2)	498 (79)	129 (19)	138 (22)	1089 (184)	4310 (280)	1877 (188)
GTH9	11.1 (1)	8.7 (1.9)	257 (37)	7.4 (0.1)	23 (2.2)	515 (43)	332 (29)	474 (36)	2742 (163)	5087 (334)	2116 (376)
RAE2	9.2 (0.5)	6.3 (0.8)	1793 (437)	7 (0)	8.5 (0.7)	308 (48)	182 (15)	219 (19)	1641 (126)	4215 (343)	5052 (1434)
ZW3	10.1 (0.2)	7.3 (0.6)	238 (29)	7.3 (0)	23.5 (0.7)	1487 (96)	119 (4)	576 (172)	2747 (95)	6123 (1300)	2023 (51)

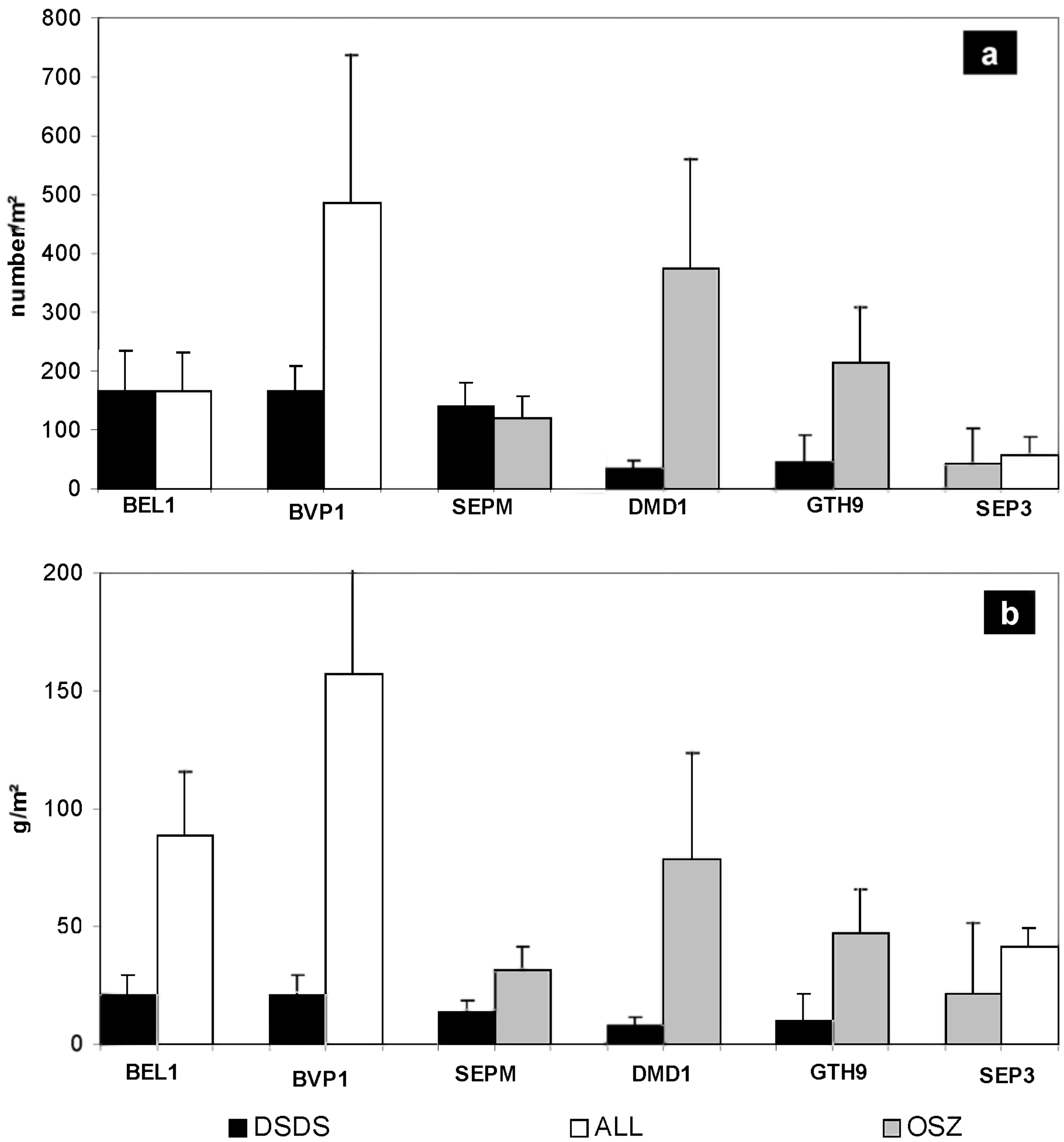

Fig. 4.6. (a) Earthworm density (N/m^2) and (b) biomass (g/m^2 FW) for the pairwise sampled sites (unaffected alluvial soils = ALL, dredged sediment-derived soils = DSDS and overbank sedimentation zones = OSZ).

Biomass distribution over the ecological categories is shown in Fig. 4.7a. It is obvious that on the DSDS endogeic and anecic earthworms are low in biomass or are absent. Relative to the unpolluted alluvial soils, polluted floodplain soils have a higher endogeic biomass. The highest weight recorded for an adult *Lumbricus rubellus* is displayed in Fig. 4.7b. For DSDS, the values tended to be lower than for the ALL or OSZ soils, but the difference was not significant ($p > 0.05$).

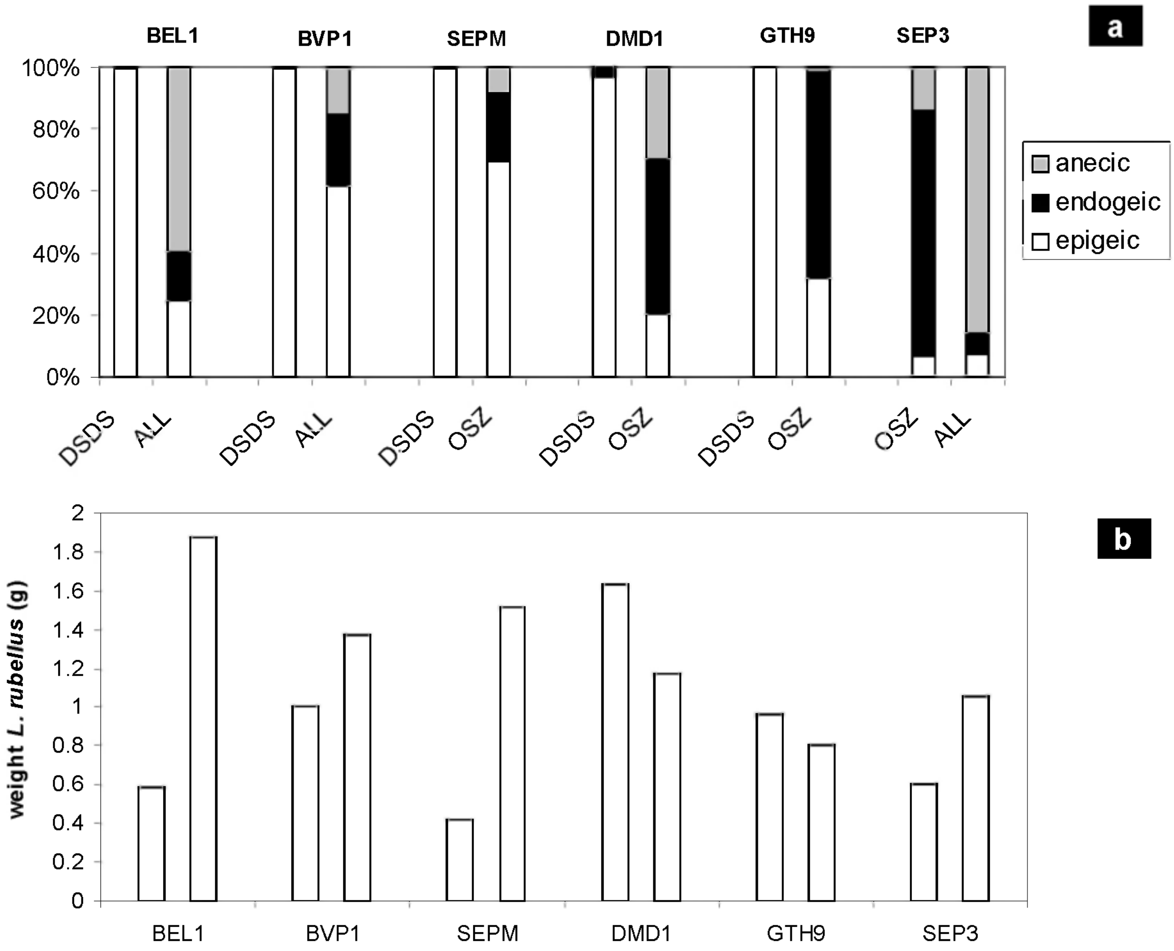

Fig. 4.7. (a) Relative distribution of the earthworm biomass over the ecological categories for the pairwise sampled sites and (b) highest weight recorded for adult *L. rubellus* (unaffected alluvial soils = ALL, dredged sediment-derived soils = DSDS and overbank sedimentation zones = OSZ). Descriptive data for the sites are given in Table 4.5 and 4.6.

3.2. Influence of time since disposal and DSDS properties on earthworm populations

RVD1 and RAE2 were recognised as outliers for earthworm biomass. RAE2 is a recent DSDS where hardly any earthworms were found, while RVD1 was landfilled 25 years ago with strongly contaminated dredged sediments, especially with Cu and Zn (Table 4.8). The RVD1 site was clearly observed to be an outlier based on the principal components analysis (PCA) of soil data for the sampled DSDS (data not shown). Highest earthworm biomass was observed at the slightly polluted APV8 site and SDD3 site, both with a sandy loam soil, and the APV1 site with a heavy clay soil (Table 4.7). For the APV8 and APV1 sites, soil cores also were excavated and hand-sorted after washing. Total earthworm biomass was 152 g/m² FW for APV8 and 80 g/m² FW for APV1. Highest biomass recorded at the alluvial soils was 158 g/m² FW.

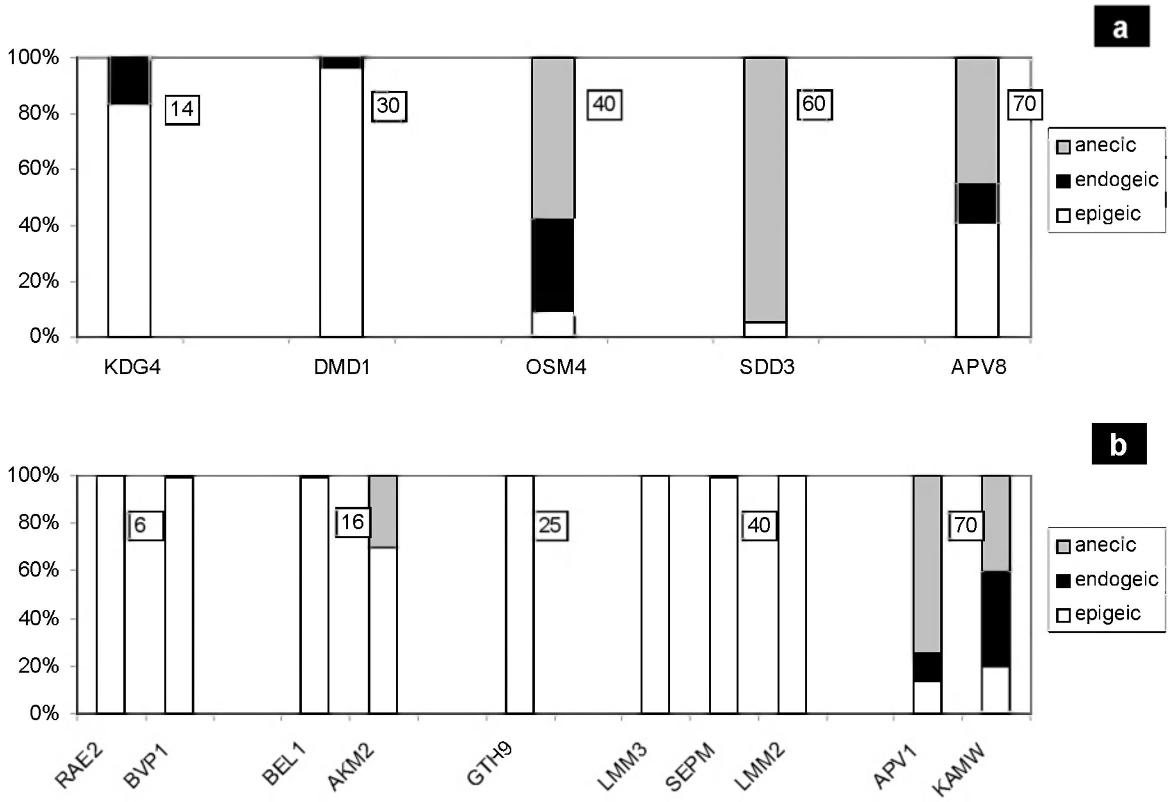

Fig. 4.8. Relative distribution of the earthworm biomass over the ecological categories for (a) sandy loam DSDS and (b) heavy clay DSDS. Descriptive data for the sites are given in Table 4.7 and 4.8. Time since disposal (TSD) is given in the boxes.

In DSDS a large range in metal concentrations was observed. Total contents of different metals were strongly correlated. From the selected soil physical measurements and calculations, the ripening factor allowed for the clearest distinction between sites. Lowest values were found for the sandy loam DSDS, while the heavy clay sites were split up in sites with values lower than 0.7 (‘completely ripened’ according to de Haan et al. (1998)) and values between 0.7 and 1.0 (‘nearly ripened’, de Haan et al. (1998)). Values for bulk density and penetration resistance were highest for the sandy loam DSDS. Penetration resistance increased with depth. Averaged results per site were comparable with data for sediment-derived soils in Illinois (Darmody and Marlin, 2002). Measured penetration resistance was clearly lower than values measured by Muys (1989) for compacted forest soils where lower earthworm biomass was observed. The Ksat values for most sites were low compared to data for storm water facilities (Massman and Butchart, 2000) and for dredged sediment disposal sites (Van Driel and Nijssen, 1988). Only for the heavy clay DSDS LMM2, BEL1, GTH9 and RAE2 and the sandy loam KDG4 and SDD3 site, Ksat values were normal to high. For the other sandy loam DSDS, Ksat values were low.

When the relative importance of the ecological categories was displayed as a function of the TSD for both grain size subsets (Fig. 4.8), it is concluded that epigeic earthworms dominate during the first period of 30 (sandy loam soils) or 40 (heavy clay soils) years. For AKM2, in one replicate one heavy *L. terrestris* was found, strongly influencing the results when *L. terrestris* was classified as an anecic species. However, the species is generally described as an epi-anecic species.

The highest negative correlation was found between the highest weight recorded for an adult *L. rubellus* and clay content. Linear regression yielded the equation weight(*L. rubellus*) = 2.63 – 0.051 * % clay ($R^2 = 0.568$, $p = 0.0003$).

A linear model was built for interpretation of DSDS earthworm biomass as a function of soil properties. Based on visual interpretation of scatterplots, both grain size distribution and TSD were recognised as main determining factors. Three classes were defined based on the grain size and the ripening factor: average biomass was 13.2, 25.7 and 68.5 g FW/m² for the completely ripened heavy clay soils, the nearly ripened heavy clay soils and the sandy loam soils. Since no difference was found between both 'heavy clay' soil classes, the dataset with all DSDS was split up in two subsets based on grain size distribution: sites with more than 30% sand were grouped as 'sandy loam' sites (DMD1, APV8, KDG4, SDD3, OSM4, RVD1), the other DSDS were the sites with 'heavy clay' soils. Data for the time since disposal (TSD) were categorised in 4 classes: DSDS constructed (1) before 1950, (2) between 1950 and 1970, (3) between 1970-1982 and (4) since 1982. Average biomass for these classes was (1) 68.5, (2) 24.2, (3) 14.5, (4) 23.3 g FW/m². However, during the construction of the model, results indicated that a clustering of data in two periods (1 *vs.* 2-3-4) was appropriate.

The selected linear model for earthworm biomass on DSDS is presented in Table 4.9. The biomass is mainly determined by TSD and grain size class (GSC), with earthworm biomass being lowest for recent heavy clay DSDS and highest for sandy loam DSDS with a TSD higher than 40 years. The negative influence of Cu pollution on earthworm biomass is very low relative to both TSD and grain size and might be compensated by the positive influence of high P concentrations as indicated by the linear model. Correlation between Cu and P is positive and significant ($R^2 = 0.47$, $p = 0.001$). In Fig. 4.9 the low relevance of adding the Cu pollution status to the basic model is clearly demonstrated. Neither soil chemical, nor soil physical variables had an additive significant influence on the model.

Table 4.9. Coefficients and descriptives for the linear model describing the sqrt(earthworm biomass (g/m² FW)) as a function of DSDS properties. Value for TSD is 0 when time since disposal < 50 year and 1 when > 50 year. GSC (Grain size class) is 0 when sand content < 30% and 1 when sand content > 30%. Cu and P are expressed as mg kg^{-1} dry soil

	Value	Std. Error	t-value	Pr(>|t|)
(Intercept)	2.7072	0.6047	4.4767	0.0000
GSC	1.8407	0.4933	3.7318	0.0003
TSD	3.5382	0.5769	6.1334	0.0000
GSC:TSD	2.4179	0.8831	2.7380	0.0074
Cu	-0.0040	0.0020	-2.0085	0.0475
P	0.0004	0.0001	3.2326	0.0017

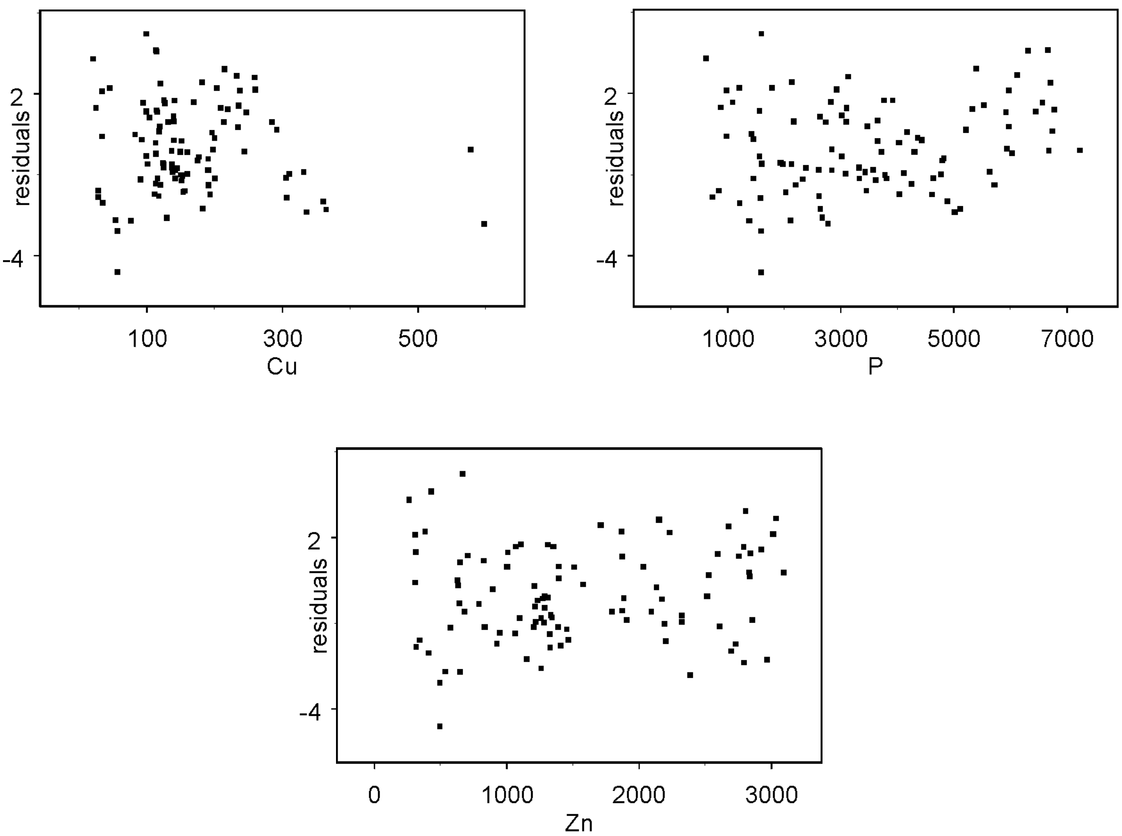

Fig. 4.9. Relation between residuals of the basic model: SQRT(earthworm biomass) = TSD + GSC + TSD:GSC as dependent, and Cu, P and Zn (mg kg^{-1} dry soil) as independent variables, with TSD = time since disposal and GSC = Grain size class.

4. Discussion

4.1. Comparison between DSDS and surrounding alluvial soils

Compared to available data for Flanders (Muys and Lust, 1992; Neirynck et al., 2000), and for alluvial soils in the Netherlands (Faber et al., 2000), normal to high earthworm biomass was found in the polluted OSZ and the sandy loam DSDS, and rather low biomass was observed in the heavy clay DSDS. Muys and Lust (1992) reported values in forest soils between less than 1 and 133 g FW/m². Neirynck et al. (2000) found on a loamy acid brown forest soil a minimum biomass of < 1 g FW/m² and a maximum of 37 g FW/m². The biomass was strongly related to dominant tree species. Both authors used the combined formalin extraction and soil core excavation method. Faber et al. (2000) reported values for floodplains excavated for clay reclamation along the Waal and Rhine river (Netherlands) based on hand sorting of excavated soil cores. Biomass ranged between 52.7-84.5 g FW/m² for the higher parts, 2.9-26.1 g FW/m² for the lower parts and 22.2-49.1 g FW/m² for the reference sites. Didden (2001) observed an average biomass of 80.4 g FW/m² and a density of 384 N/m² on grassland soils (20 sites) in the Netherlands, based on hand sorting of excavated soil cores.

Data of Yeates and Orchard (1994) suggested that earthworms in superficially contaminated areas were also feeding on less polluted soil material from deeper in the profile, and in the highest polluted sites closest to smelters, only the endogeic species *Aporrectodea caliginosa* Savigny survived (Bengtsson and Tranvik, 1989). This behaviour may also explain the relatively high earthworm biomass found at polluted OSZ. In strongly degraded and acidified forest soils in Flanders, only epigeic earthworms can survive (Muys and Lust, 1992). Spurgeon et al. (1996) determined earthworm biomass and abundance in a gradient in the vicinity of a smelter. No earthworms were found on the sites closest to the smelter with accumulation of undecomposed leaf litter, only *Lumbricus* species were found in the intermediate sites and both *Lumbricus* species and endogeic species were found farthest away. Morgan and Morgan (1999) focused on the importance of the vertical distribution of metals within the soil profile for metal exposure and uptake by earthworms occupying different ecological niches with specific food preferences. Hence, the high relative proportion of endogeic earthworms on the polluted OSZ might be a consequence of the concentration of pollutants in the upper soil horizons. For the slightly polluted floodplains (Cd < 6.8 mg kg^{-1} dry soil, Zn < 739 mg kg^{-1} dry soil, Cu < 133 mg kg^{-1} dry soil) of the Waal river (Netherlands) the pollution was found to be of less importance for species composition and diversity of the functional groups of the invertebrate fauna (Ma et al., 1997).

4.2. Influence of time since disposal and DSDS properties on earthworm populations

Soil characteristics on DSDS are, apart from the pollution status, optimal to very prosperous for earthworms: high SOM content, mull-type humus forms, high carbonate contents and subsequent optimal pH. Earthworm biomass can not be directly linked to metal pollution, as the sediment substrate is also enriched by PCBs, PAHs and other pollutants. However, in accordance with the Flemish Decree on Soil Sanitation (VLAREBO, 1996), metal contamination in DSDS with especially Cd, Cr and Zn was considered more severe than for other pollutants (Vandecasteele et al., 2000). Earthworms are very sensitive to Cu pollution (Ma, 1982; Ma et al., 1983). Higher soil Cu concentrations result in a longer period before subadult stage and adulthood is reached or even in the inability to reach adulthood (Ma, 1983; Spurgeon and Hopkin, 1996). In areas with topsoils contaminated with Cu, Cr and As as a result of the use of timber preservatives, lower earthworm biomass was found at higher Cu concentrations. No higher tissue concentrations were encountered excluding food chain effects for Cu (Yeates and Orchard, 1994). In optimal conditions with abundance of food and absence of predation, populations of juvenile *L. rubellus* treated with 362 mg Cu kg^{-1} dry soil showed negative growth rates as the minimum body weight to reach adulthood was never reached (Ma, 1984). Klok and De Roos (1996) calculated that the critical threshold for major danger of extinction on the population level for *L. rubellus* was 200-300 mg Cu kg^{-1} dry soil. Stress due to sublethal toxicant concentrations for earthworms results in a decreased metal detoxification (Hönsi et al., 2003), a reduced cocoon production (Siekierska and Urbanska-Jasik, 2002) or a failure to reach adulthood (Ma, 1984). Filser et al. (1995) concludes from a literature review that higher SOM content reduces Cu toxicity to a great extent. Our results did not indicate a large influence of soil Cu concentrations on earthworm biomass, not even for the most polluted site (RVD1, > 600 mg Cu kg^{-1} dry soil). Interpretation of this observation is not straightforward. The DSDS might be characterised by a low Cu bioavailability. Alternatively, the rate of colonisation and/or soil physicochemical properties may be the more important limiting factors than pollution status for earthworm biomass. DSDS are quickly colonised by epigeic earthworms, since even for sites that are flooded during the winter and a large part of the spring (BVP5, GTH2, SEP3) earthworms were found.

In contrast to the initially fast colonisation, it takes a rather long time (at least 40 years) to reach a biomass higher than 30 g FW/m².

The soil profile of DSDS deviates from normal soil profiles, as the soil organic matter content is high throughout the whole soil profile, while it is concentrated in the topsoil for normal alluvial soils. On the DSDS, the earthworm population was dominated by epigeic species, especially by *L. rubellus*. The pollution status of the soil did not prevent the cocoons to become juveniles and juvenile earthworms to reach adulthood. The absence of endogeic and anecic earthworms on recent DSDS might be a consequence of both the ecological stress and the colonisation strategy. Both categories are known to have a slow colonisation rate (K-strategy). Endogeic worms are also known to feed on large amounts of soils and consequently have a more intense contact (higher exposure) with the soil pollution. Both the pollution and the lower oxygen availability in the profile can be a reason for their absence. Earthworms play an important role in SOM decomposition and subsequent incorporation in the mineral soil (Ma, 1984; Edwards and Fletcher, 1988). The slow colonisation or the adverse effects of polluted soils on soil organisms can result in a hampered SOM decomposition (Yeates and Orchard, 1994).

4.3. Ecological risk assessment

In risk assessment based on trophic chains of several species inhabiting alluvial plains and floodplains, earthworms will play an important role as they can form up to 80% of the soil faunal biomass. A general trend is that sandy loam DSDS seem very prosperous for earthworms, while heavy clay DSDS are less suited. In general, it can be concluded that relative to the surrounding environment, earthworm biomass is four times lower at heavy clay DSDS. However, total biomass might be a biased indicator, as not all ecological categories of earthworms are evenly susceptible to predation. Epigeic earthworms live on the soil surface, endogeic earthworms stay in the soil, while anecic earthworms are only feeding at the soil surface at night.

In general, Cd is the most important pollutant for food chain transfer, while Cu is the most important element limiting earthworm survival. This means that for soils with low Cu pollution but with high Cd pollution there is a larger risk as more earthworms with higher Cd concentrations are available for predation, while in the opposed case less earthworms can survive. The results of this study point at the colonisation rate of a polluted site as another factor in the risk assessment of DSDS. Risk assessment models focus on the metal uptake and transfer by earthworms. Knowledge of earthworm metal tissue concentrations is essential for risk assessment of metal biomagnification. Before using an appropriate model for a selected area, differences in earthworm biomass related to different soil properties must be considered. If no large differences exist between the reference situation (in this case ALL) and the polluted site, a model without correction for biomass can be used, i.e., an equal biomass at all sites is inherently assumed. However, if differences between the reference and the polluted site are large, the food availability is subject to spatial variability. Spatial patterns in food availability must be included in risk assessment, as is the case for spatial patterns of soil pollution (Kooistra et al., 2001) and for differences in feeding behaviour of target animals (Heikens et al., 2001). Both earthworm tissue concentrations and data on earthworm biomass might thus be necessary for a good ecological risk assessment of biomagnification.

Secondary poisoning can result in changes at higher organisation levels of the biological system. An indirect effect of soil pollution is a possible food shortage for higher levels in the food chain (Hörnfeldt and Nyholm, 1996; Klok et al., 2000) or a changed, less optimal diet (Van den Brink et al., 2003). Secondary poisoning is highly dependent on the configuration of the polluted area and ecology of the target species (Menzie et al., 1992). Sandy soils, waterlogged soils, swamps and lakes, forests and urbanisation were found to have a negative impact on Little Owl presence. As the preferred soil types are also optimal for earthworms, food availability was thought to be a major factor for the Little Owl habitat (Van Nieuwenhuyse et al., 2001).

5. Conclusions

We focused on earthworm biomass determination as additional information for risk assessment of metal biomagnification through the foodweb on DSDS relative to the surrounding alluvial plains. In general it can be concluded that relative to the surrounding environment, earthworm biomass is four times lower for contaminated dredged sediment-derived heavy clay soils and comparable to alluvial soils for sandy loam DSDS. Risks for secondary poisoning at the more polluted heavy clay DSDS are thus partially compensated by the lower earthworm biomass. However, not all ecological categories of earthworms are equally susceptible to predation. Endogeic earthworms stay in the soil, while anecic earthworms are only at night at the soil surface.

It was found that recent sediment landfills were colonised by epigeic earthworms rather fast, but larger earthworm populations were only observed in sites of more than 50 years old. Relative to polluted overbank sedimentation zones, impact of dredged sediment disposal on earthworm communities is large as data suggest that it takes more than 40 years until all ecological categories have colonised the sites. A clear difference was observed between heavy clay and sandy loam DSDS with a distinct higher earthworm biomass on the sandy loam DSDS.

For *L. rubellus*, a negative relation was found between the highest recorded body weight and the clay content of the DSDS. The highest recorded adult body weight per species is thus a potential habitat quality parameter. Mainly time since disposal and grain size distribution determined earthworm biomass on DSDS, while considerable levels of soil pollution were merely found to influence this important population characteristic in view of risk assessment. High soil Cu concentrations did thus not lower the risk for Cd biomagnification through a reduced earthworm population. For future ecotoxicological field research on earthworm populations, inclusion of a broad range of soil physical and chemical variables is worthy of consideration.

Synthesis, conclusions and research prospects

Synthesis and conclusions

The first goal of our study (Part 1) was to survey the alluvial plains of the Leie and the Derivation Canal, the Canal Ghent-Bruges and the Upper and Sea Scheldt for the presence of dredged sediment-derived soils (DSDS), and to measure and appraise the metal contamination at these sites. Results on geographical impact for the Leie river were presented in Chapter 1.1. An overview of the geographical results for the study area is given in Chapter 1.2.

In Part 2, land use and spatial planning objectives for dredged sediment-derived soils in the study area are summarised, and actual and potential risks for polluted DSDS are assessed. When polluted DSDS are assessed to be a serious threat for the environment, sanitation is mandatory. A concept of safe land use of polluted areas as an alternative remediation technique has been developed for uncovered dredged sediment-derived soils in nature conservation areas. Two frequent endpoints were selected for further research: an upland soil type afforested with appropriate tree species, and a wetland soil type with dominance of willow shrubs.

Bioavailability of metals for plants was the topic of Part 3 of the study. Foliar samples were collected for several current land uses on DSDS for assessment of metal bioavailability. Site and species effects on metal availability for *Salix alba* and *Salix cinerea* was focussed upon in Chapter 3.1, 3.2 and 3.3.

Effects of pollutants on organisms and processes for contaminated soils might vary considerably. In Part 4, research on ecosystem effects of metal pollution for forest floor cycling and soil-forming processes (Chapter 4.1) and earthworm populations (Chapter 4.2) are presented.

Conclusions about four issues were drawn from the results of this study: (1) knowledge of soil and sediment quality, (2) an integrated approach for ecological risks, (3) the safe management of polluted DSDS and (4) site-specific and regional biomonitoring.

1. Knowledge of soil and sediment quality

Former and current human activities are an obstruction or a limitation for river restoration. A soil survey and an archive query for reconstructing the history of dredging operations were conducted simultaneously. The geographical impact expressed as topographical changes and covering of the original soil profile and related processes and biota was large. The pollution status of dredged sediment-derived soils was found to be far from negligible. Results indicate the importance of soil quality assessment in alluvial plains for an integrated river management, rather than *a priori* assuming pristine soil conditions.

Flood events strongly influence the trace-metal content of the suspended matter by inducing resuspension processes and intensive transfer of solid material from the river system to the sea. The removal of large quantities of polluted sediment during dredging operations can be considered as a kind of clean-up operation: the removal of large amount of pollutants from the river system potentially protected the downstream tidal marshes, the Scheldt estuary and the North Sea.

Alluvial plains act as sink for nutrients and pollutants. Sediment-derived soils developed spontaneously as a consequence of overbank sedimentation or are human-made as disposal site for dredged sediments, and were characterised by metal pollution. Soil properties of sediment-derived soils clearly deviate from normal unaffected soils due to the pollution status and the high nutrient concentrations. Currently, a contradiction is found in legislation with stringent criteria for construction of new dredged sediment landfills while overbank sedimentation of polluted sediments in alluvial plains and constructed wetlands is considered to be a natural process without further constraints. Spatial and management planning for alluvial plains must start from the knowledge that pollution due to sediment dispersal might be present instead of ignoring environmental issues in nature management.

Soils in landfilled dredged sediments are high in organic matter, clay and calcium carbonate. Leaching of metals and subsequent ground water pollution is therefore of less environmental concern. However, metal bioavailability for plants and soil biota is a possible threat, in particular in the long-term. Both pollution levels and affected areas determine the extent of the ecological risk involved. Soil decalcification and acidification pose a long-term risk as these processes may lead to metal leaching. Both agriculture and nature rehabilitation on dredged sediment-derived soils can only be accepted after profound risk assessment, and management should focus on ecological risk reduction. Risk assessment should not be based solely on soil physical and chemical properties, but should also account for metal availability for plants and soil invertebrates. Therefore, risk assessment of historically polluted soils must be conducted for food webs relevant for the studied site.

An integrated regional approach of polluted sediments in rivers and in alluvial plains and tidal muds and flats is necessary. Soil quality data for the alluvial plain (pollution status), relative importance of each soil type in terms of areas and knowledge on relevant food webs are the basic needs for regional risk assessment and as a tool for management and planning. Food webs must be considered without simplification. The effect of other factors than soil pollution status on risk of biomagnification must be accounted for. The important role of earthworms in metal biomagnification in terrestrial ecosystems is widely recognised. Differences in earthworm biomass between sites are mostly not accounted for in ecological risk assessment. These differences may be large depending on soil properties and pollution status. A survey of earthworm biomass and colonisation rate was carried out on dredged sediment-derived soils and surrounding alluvial plains. Mainly grain size distribution and time since disposal determined earthworm biomass on dredged sediment-derived soils, while soil pollution status was of lesser importance.

2. Integrated approach for ecological risks

We are confronted with another contradictory reality: alluvial plains with polluted sediment-derived soils are protected for nature conservation, but pollution levels and related soil properties limit the habitat functioning and can be a threat through food web transfer and biomagnification. Management of these alluvial areas must focus on reducing metal biomagnification and this goal must have priority on other objectives about habitat type. Appropriate water management should prevent or slow down acidification and reduce bio-availability of pollutants. It is also important in development planning to give a correct destination to the dredged sediment-derived soils, as the polluted substrate cannot fulfil all the basic functions of a normal soil. There is a clear need for a frame of reference for nature management in polluted areas. Before selecting a target nature development goal, nature management objectives must be confronted with environmental management objectives and the feasibility of combining both objectives must be evaluated.

An important choice in risk assessment is the scale of the target area. In a first approximation of an area, one can require that each single part should fulfil all the prerequisites for optimal habitat function and should allow optimal biodiversity and sustainable habitat development. From a more moderate point of view, one can state that the polluted sediment-derived soils must guarantee the save disposal of pollutants and may not hinder the habitat functioning of the whole area, both in the current situation and in the future. From this point of view the relative portion of sediment-derived soils in the area will be an important element in the evaluation. Apart from any nature management objective, a normal functioning habitat with specific characteristics can develop on polluted sediment-derived soils. As for many organisms acclimation and adaptation mechanisms or avoidance behaviour for pollution is found, besides the processes in soil leading to ageing of pollution, it can be expected to keep on functioning over a long period. Translated to spatial planning, the whole area is selected as nature area in the first stringent case, while in the second case, polluted sites have an adjusted risk-reducing land use within a larger area.

Ecological risks of soil pollution are not always perceptible in the field, and the presence of ecotopes with certain ecological values on polluted sites hinder drastic remediation. Safe management aiming at ecological risk reduction for these sites is a potential strategy.

3. Safe management of polluted sediment-derived soils

It is obvious that sediment-derived soils are strongly deviant from the normal alluvial soils and thus cannot match the processes and habitat linked with alluvial soils. Besides the low probability of acute or chronic toxicity for plants and soil organisms in direct contact with the polluted substrate, a large uncertainty is involved with the risk assessment for secondary poisoning. There is currently no frame of reference for nature management in polluted areas. Pragmatism leads to management of polluted sediment-derived soils focusing on avoiding hinder for the functioning of the whole area. As many obstacles for expensive and drastic management options such as capping and excavation exist, future research should focus on the determination of the dynamics of soil forming processes on sediment-derived soils and the long-term consequences of less drastic risk-reducing management options such as hydrological management.

Field observations and results of the greenhouse experiment suggest that a hydrological regime aiming at wetland creation is a potential management option that favours reducing metal bioavailability for plants. The advantage of willows for research on metal bioavailability is that they reflect time-integrated accumulated concentrations for the studied soil profiles. Longer submersion periods in the field caused lower Cd concentrations in the leaves and the bark for the wetland plant species *Salix cinerea*. The wetland hydrological regime in the greenhouse experiment resulted in normal Cd and Zn concentrations in the leaves, and normal Cd and high Mn concentrations in the cuttings, while the upland hydrological regime caused elevated Cd and Zn concentrations in the leaves and elevated Cd concentrations in the cuttings. This would constitute a safe management option of metal-polluted, willow-dominated wetlands provided wetland conditions can be maintained throughout the full growing season. Especially for Cd, a transfer effect from one growing season to the next season was observed: submersion conditions in the previous growing season seem to determine at least partly the foliar concentrations for *Salix cinerea* through this translocation mechanism. Duration of the submersion period is a key factor since initially submerged soils emerging only in the second half of the growing season resulted in elevated Cd and Zn foliar concentrations.

Soil processes on a calcareous upland dredged sediment landfill after afforestation resulted in small differences between the topsoil and the deeper soil layer, although higher soil organic carbon and Cd concentrations in the topsoil were observed. Despite the pollution status of the dredged sediment landfill, forest floor decomposition was found to be relatively fast for sycamore maple and pedunculate oak. This might indicate that the soil nutrition status and the high carbonate status override the negative impact of soil pollution with metals and other pollutants. Application of an uncontaminated cover topsoil resulted in lower Cd concentrations in earthworms. No adverse effects were observed after 16 years of landfilling and 12 years of afforestation. It is concluded from the observations that polluted but fertile soils allow for afforestation and for regular forest floor decomposition with normal or slightly elevated metal concentrations.

4. Site-specific and regional biomonitoring for sediment-derived soils

Each pathway of pollution has specific characteristics. Several authors proved in field trials and in experiments under controlled conditions the large impact of pH, redox potential, clay and organic matter content, cation exchange capacity, aging effects on pollutants, metal speciation and heterogeneity of the pollution within the soil on metal availability. Interaction with living organisms is complex due to genetic variability, adaptation and avoidance mechanisms. These findings may strengthen the relevance of passive biomonitoring and site-specific assessment of pollution as an alternative for defining general permissible levels in soils and biota. Metal availability of plants is determined by a range of soil charactistics, but plant species is the dominant factor in determing metal availability in the first trophic level in food webs as large differences in metal accumulation between plant species exist.

Metal contamination in forests and nature conservation areas in Flanders is caused by atmospheric deposition due to industrial activities, emission from traffic, metals in fertilisers and deposition of waste and sediments. These pathways of soil contamination can be categorised as diffuse or point sources of pollution. Metal pollution due to dredged sediment disposal is a point source, with pollution being clearly confined within the boundaries of the site. Site-specific ecological risk assessment focuses on pollution effects observed within the boundaries of the site. However, effects and bioavailability of pollution may occur over the boundaries and may more or less persist on the level of the river basin or on a regional scale.

For site-specific biomonitoring, bioavailability and bioaccumulation must be recognised on one hand, and effects on biota, biodiversity and processes must be assessed. Presented results indicate the applicability of foliar and forest floor sampling for site-specific risk assessment and biomonitoring. However, several factors complicate the application for regional biomonitoring. Tree species composition (e.g. the presence of willows and poplars) was found to affect the metal concentrations in the forest floor. Foliar concentrations in willows were observed to be largely determined by tree individuality. A clear effect of duration of submersion and redox pontential on metal availability for willows was reported. These factors are mainly determined by site management, and may thus hinder regional biomonitoring. Regional biomonitoring should focus on bioaccumulation in higher trophic levels in the food web or should be highly standardised for the effect of land use and site management.

Future research needs and prospects

1. Inclusion of food web complexity, spatial patterns of food availability, spatial patterns of soil pollution and differences in feeding behaviour for target animals in ecological risk assessment
2. Inclusion of spatial patterns in plant concentrations, plant species composition and feeding behaviour of herbivorous target species in ecological risk assessment

3. Case studies on the effects of high plant concentrations (e.g. Cd and Zn concentrations measured in willows and poplars on polluted sediment-derived soils) on herbivorous organisms and interactions with higher trophic levels
4. Method development for monitoring of bioaccumulation in higher levels in the food web (birds, mammals)
5. Application of monitoring methods for higher levels in the food web (birds, mammals) for areas affected by different pathways of pollution
6. Assessment of the impact of scale on ecological risks: does slight contamination over large areas (diffuse pollution) include a higher risk than highly contaminated sites (point sources) scattered over a relatively uncontaminated area?
7. Use of grazing by herbivores as management option for polluted sediment-derived soils in nature conservation areas
8. Relation between soil physical properties and soil chemical properties for polluted sediment-derived soils in a submersion-emersion cycle
9. Determination of the dynamics of soil forming processes on sediment-derived soils
10. Long-term consequences of less drastic risk-reducing management options such as hydrological management

Policy recommendations

1. Harmonisation of legislation on dredged sediment-derived soils and other categories of sediment-derived soils
2. Development of a frame of reference for nature management in polluted areas, in which nature management objectives are confronted with environmental management objectives and the feasibility of combining both objectives is evaluated
3. Development of an integrated approach of polluted sediments from a watershed perspective
4. Regional biomonitoring of metal pollution focussing on bioaccumulation in higher trophic levels in the food web, with strong standardisation for the effect of land use and site management
5. Application of passive biomonitoring and site-specific assessment of pollution as an alternative for defining general permissible levels in soils and biota

Synthese, conclusies en toekomstig onderzoek

Synthese, conclusies en toekomstige onderzoek

De eerste doelstelling van dit werk (Deel 1) was het bepalen van de bodemkwaliteit van de alluviale gebieden en meer specifiek de baggergronden langs de Bovenschelde, de Zeeschelde, de Leie en het afleidingskanaal, en het kanaal Gent-Brugge, en het bepalen van de metaalverontreiniging op deze plaatsen. De resultaten over de geografische impact van de baggergronden voor de Leie werden voorgesteld in in Hoofdstuk 1.1. In Hoofdstuk 1.2. werden de geografische resultaten voor het volledige studiegebied samengevat.

In Deel 2 wordt het landgebruik en de geplande bestemming voor de baggergronden in het studiegebied beschreven, en worden de actuele en potentiële risico's van verontreinigde baggergronden samengevat. Wanneer geoordeeld wordt dat verontreinigde baggergronden een ernstige bedreiging vormen voor het milieu, dan is sanering verplicht. Er werd een concept van veilig beheer van verontreinigde gebieden ontwikkeld als een alternatieve saneringsstrategie voor verontreinigde baggergronden zonder afdeklaag in natuurgebieden. Twee veel voorkomende situaties werden geselecteerd voor verder onderzoek: enerzijds baggergstortterreinen die met de geschikte boomsoorten aangeplant werden, en anderzijds moerassige baggerstortterreinen met een dominante wilgenvegetatie.

De biobeschikbaarheid van metalen voor planten is het onderwerp van Deel 3 van dit werk. Bladstalen werden verzameld voor verschillende landgebruiksvormen op baggergronden voor het bepalen van de biobeschikbaarheid van metalen. Standplaats- en soorteffecten op de metaalbeschikbaarheid voor *Salix alba* en *Salix cinerea* werden onderzocht in Hoofdstuk 3.1, 3.2 en 3.3.

De effecten van polluenten op organismen en processen bij verontreinigde bodems kunnen sterk variëren. In Deel 4 werd het onderzoek naar de ecosysteemeffecten van metaalverontreiniging op de strooiselafbraak en bodemvormende processen (Hoofdstuk 4.1) en regenwormpopulaties (Hoofdstuk 4.2) voorgesteld.

De resultaten werden vertaald in besluiten rond 4 kwesties: (1) kennis omtrent bodem- en sedimentkwaliteit, (2) een geïntegreerde benadering van ecologische risico's, (3) het veilig beheer van verontreinigde baggergronden en (4) locatiespecifieke en regionale biomonitoring.

1. Kennis van de bodem- en sedimentkwaliteit

Voormalige en huidige menselijke activiteiten kunnen een belemmering vormen voor rivierherstel. Gegevens over bodemverontreiniging met metalen bij voormalige baggerstortterreinen werden geconfronteerd met archiefgegevens van baggerwerken om de recente geschiedenis van de aanleg van baggerstortterreinen te kunnen reconstrueren. De geografische impact, uitgedrukt als topografische veranderingen en het bedekken van het oorspronkelijk bodemprofiel en hiermee verwante processen en biota, was groot. De verontreinigingsgraad van baggergronden bleek verre van verwaarloosbaar te zijn. Resultaten wijzen op het belang van kennis van de bodemkwaliteit in alluviale gebieden voor integraal waterbeheer, in plaats van *a priori* te veronderstellen dat er een normale bodemkwaliteit is.

Hoge waterstanden resulteren in resuspensie en verhoogd sedimenttransport van het riviersysteem naar de zee. Het verwijderen van grote hoeveelheden verontreinigd sediment tijdens baggerwerken kan beschouwd worden als een soort saneringsoperatie, waarbij stroomafwaarts gelegen schorren, het Schelde-estuarium en de Noordzee beschermd worden.

De alluviale vlakte fungeert als een sink voor nutriënten en polluenten. Baggergronden ontwikkelen zich spontaan door het afzetten van sedimenten of door de aanleg van baggerstortterreinen en worden gekarakteriseerd door metaalverontreiniging. De eigenschappen van baggergronden wijken duidelijk af van normale alluviale gronden door de aanwezigheid van verontreiniging en de hoge nutriëntconcentraties. Momenteel is er een tegenstrijdigheid in de wetgeving met enerzijds strenge criteria voor de aanleg van nieuwe stortterreinen voor baggerspecie terwijl anderzijds de afzetting van verontreinigde sedimenten in de alluviale vlakte en in gecontroleerde overstromingsgebieden als een natuurlijk proces beschouwd wordt zonder dat er hierbij verdere beperkingen zijn. Bij de ruimtelijke en beheersplanning van alluviale gebieden moet uitgegaan worden van de wetenschap dat verontreiniging door de aanwezigheid van sedimenten aanwezig kan zijn.

De bodems van baggergronden hebben een hoog organische stof-, klei- en calciumcarbonaatgehalte. Het uitlogen van metalen en eventuele grondwaterverontreiniging is in deze omstandigheden minder waarschijnlijk. Daarentegen is de biobeschikbaarheid van metalen voor planten en bodeminvertebraten een mogelijke bedreiging, in het bijzonder op lange termijn. Zowel de graad van verontreiniging als het relatieve belang van de oppervlakte aan verontreinigde bodems bepalen de omvang van het ecologisch risico. Bodemdecalcificatie en -verzuring vormt een risico op lange termijn aangezien deze processen wel kunnen leiden tot het uitlogen van de metalen. Zowel landbouw als natuurinrichting op baggergronden kan enkel aanvaard worden na een risicoschatting, en het beheer zou zich op de reductie van het ecologische risico moeten concentreren. Risicobeoordeling mag niet enkel gebaseerd zijn op bodemfysische en –chemische eigenschappen maar zou ook de metaalbeschikbaarheid voor planten en ongewervelde bodemdieren moeten in rekening brengen. Daarom moet de risicoschatting van historisch verontreinigde gronden gebaseerd zijn op voedselwebben die relevant zijn voor de bestudeerde locaties.

Een geïntegreerde regionale benadering van verontreinigde sedimenten in rivieren, slikken en schorren en in alluviale gebieden is noodzakelijk. Bodemkwaliteitsgegevens voor het alluviale gebied (verontreinigingsstatus), het relatieve belang van elk deelsysteem (slikken, schorren, baggergronden, …) uitgedrukt in oppervlakte, en kennis van relevante voedselwebben zijn de basisgegevens voor een regionale risicoschatting en voor beheer- en planningsdoelstellingen. Voedselwebben moeten zonder vereenvoudiging beschouwd worden. Het effect van andere factoren dan bodemverontreiniging op het risico voor biomagnificatie moeten in rekening gebracht worden. De belangrijke rol van regenwormen in de biomagnificatie van metalen in terrestrische ecosystemen is algemeen aanvaard. Verschillen in regenwormbiomassa tussen locaties worden meestal niet in rekening gebracht bij ecologische risicoschatting. Deze verschillen kunnen echter groot zijn, afhankelijk van bodemeigenschappen en verontreinigingsgraad. Gegevens van regenwormbiomassa en kolonisatiesnelheid op baggergronden werd vergeleken met gegevens voor de omringende alluviale gebieden. De regenwormbiomassa op baggerstortterreinen wordt hoofdzakelijk bepaald door de bodemtextuur en de tijdsduur sinds de aanleg van het baggerstortterrein, terwijl de graad van bodemverontreiniging weinig effect had op de regenwormbiomassa.

2. Geïntegreerde benadering voor ecologische risico's

We worden met een andere tegenstrijdige realiteit geconfronteerd: alluviale gebieden met verontreinigde baggergronden worden voor natuurbehoudsdoelstellingen beschermd, maar de verontreinigingsgraad beperkt het functioneren van het habitat en kan een bedreiging vormen door metaaltransfer doorheen het voedselweb via biomagnificatie. Het beheer van deze alluviale gebieden moet zich richten op het beperken van metaalbiomagnificatie en dit doel moet voorrang krijgen op andere doelstellingen. Een gepast waterbeheer moet bodemverzuring voorkomen of vertragen en de biobeschikbaarheid van metalen beperken. Het is ook belangrijk om bij de ruimtelijke planning een geschikte bestemming te geven aan baggergronden, aangezien de verontreinigde baggergronden niet alle basisfuncties van een normale bodem kunnen vervullen. Er is een duidelijke nood aan een referentiekader voor natuurbeheer in verontreinigde gebieden. Voor er een natuurdoeltype geselecteerd wordt, moeten de randvoorwaarden (o.a. de bodemverontreiniging) duidelijk beoordeeld worden en moet de haalbaarheid van de doelstelling geëvalueerd worden.

Een belangrijke keuze bij de risicoschatting is de schaal van het doelgebied. In een eerste benadering van een gebied kan er gesteld worden dat elk perceel aan alle voorwaarden voor het optimaal functioneren van het habitat moet voldoen en een optimale biodiversiteit en duurzame habitatontwikkeling moet toelaten. Vanuit een meer gematigd standpunt kan er gesteld worden dat verontreinigde baggergronden de metalen moeten vastleggen en het functioneren van het grotere gebied zowel in de huidige toestand als in de toekomst niet mogen belemmeren. Vanuit dit standpunt zal het relatieve belang van de oppervlakte aan verontreinigde baggergronden in het gebied een belangrijk onderdeel in de evaluatie zijn. Los van elke natuurbeheerdoelstelling kan er zich een normaal functionerend ecotoop met specifieke kenmerken ontwikkelen op verontreinigde baggergronden. Aangezien bij veel organismen acclimatie en aanpassingsmechanismen of vermijdend gedrag t.o.v. verontreiniging vastgesteld werd, naast de processen in de bodem die tot het minder beschikbaar worden van de verontreiniging leiden, kan verwacht worden dat deze ecotopen over een lange periode kunnen blijven functioneren. Vertaald naar ruimtelijke planning betekent dit dat het hele gebied is als natuurgebied ingekleurd wordt in het eerste strenge geval, terwijl in het tweede geval de verontreinigde locaties een aangepaste functie binnen het grotere gebied krijgen.

Ecologische risico's van bodemverontreiniging zijn niet altijd waarneembaar in het veld en de aanwezigheid van ecotopen met een zekere ecologische waarde op verontreinigde plaatsen belemmeren drastische saneringsingrepen. Het veilig beheer gericht op ecologische risicoreductie is een potentiële strategie voor deze locaties.

3. Het veilig beheer van verontreinigde baggergronden

Het is duidelijk dat baggergronden sterk afwijkend zijn van de normale alluviale gronden en de processen en habitats verbonden met alluviale gronden niet kunnen evenaren. Naast de lage kans op acute of chronische toxiciteit voor planten en bodemorganismen in rechtstreeks contact met de verontreinigde bodem, is er een grote onzekerheid bij de risicoschatting voor secundaire vergiftiging. Er is momenteel geen referentiekader voor natuurbeheer bij verontreinigde gebieden. Pragmatisme leidt tot een beheer van verontreinigde baggergronden die zich concentreert op het vermijden van negatieve effecten voor het functioneren van het hele gebied. Aangezien er vele praktische hindernissen zijn voor dure en drastische beheersopties zoals het afdekken en afgraven, zou het toekomstig onderzoek zich op de dynamiek van bodemvormende processen op baggergronden en de langetermijn gevolgen van minder drastische risicobeperkende beheersopties zoals hydrologisch beheer moeten concentreren.

Veldwaarnemingen en resultaten van het serre-experiment tonen aan dat een hydrologisch regime dat op moerascreatie gericht is, een potentiële beheersoptie is de biobeschikbaarheid van metalen voor planten kan verminderen. Langere perioden van overstroming in het veld veroorzaakten lagere Cd-concentraties in de schors en de bladeren van Grauwe wilg (*Salix cinerea*), een soort typisch voor moerassen. Het moeras-regime in het serre-experiment resulteerde in normale Cd- en Zn-concentraties in de bladeren en normale Cd-concentraties in de stekken, terwijl het vochtregime ‘veldcapaciteit’ hoge Cd- en Zn-concentraties in de bladeren en verhoogde Cd-concentraties in de stekken veroorzaakte. Het creëren of in stand houden van een moerassituatie zou een veilige beheersoptie kunnen zijn voor verontreinigde baggergronden met een wilgenvegetatie, op voorwaarde dat de moerasomstandigheden het hele groeiseizoen gegarandeerd kunnen worden. In het bijzonder voor Cd werd er een overdracht-effect naar het volgende groeiseizoen vastgesteld onder veldomstandigheden: de hydrologische omstandigheden in het vorige groeiseizoen blijken tenminste gedeeltelijk de bladconcentraties voor *Salix cinerea* te bepalen door dit translocatiemechanisme. De duur van de overstromingsperiode is een bepalende factor aangezien overstroomde bodems die in de tweede helft van het groeiseizoen droog kwamen te staan, gekenmerkt worden door verhoogde Cd- en Zn- concentraties in de bladeren.

Bodemvormende processen op een kalkrijk baggerstortterrein resulteerde na bebossing in lage verschillen tussen de bovengrond en de diepere bodemlaag, niettegenstaande hogere organische koolstofgehalten en Cd-concentraties in de bovengrond gemeten werden. Ondanks de verontreinigingsgraad van het baggerstortterrein, verliep de strooiseldecompositie betrekkelijk snel voor Gewone esdoorn en Zomereik. Dit kan er op wijzen dat de bodemvruchtbaarheid en het hoge carbonaatgehalte de negatieve effecten van de bodemverontreiniging met metalen compenseren. Het aanbrengen van een niet-gecontamineerde afdeklaag leidde tot lagere Cd-concentraties in regenwormen. Er werden 16 jaar na de aanleg van het baggerstortterrein en 12 jaar na de bebossing geen negatieve effecten of ongunstige evoluties vastgesteld. Een verontreinigde baggergrond laat bebossing toe, gekenmerkt door een normale strooiselafbraak met licht verhoogde Cd- en Zn-concentraties.

4. Locatie-specifieke en regionale biomonitoring voor baggergronden

Elke blootstellingsroute aan verontreiniging heeft specifieke kenmerken. Het grote effect van pH, redox-potentiaal, klei- en organische stofgehalte, kationenuitwisselingscapaciteit, metaalspeciatie en heterogeniteit van de verontreiniging binnen het bodemprofiel op de metaalbeschikbaarheid is algemeen bekend. De wisselwerking met levende organismen is complex tengevolge van genetische variabiliteit, aanpassings- en vermijdingsmechanismen. Deze vaststellingen beklemtonen de relevantie van passieve biomonitoring en locatie-specifieke beoordeling van verontreiniging als een alternatief voor het definiëren van toelaatbare concentraties in bodems en biota. De metaalbeschikbaarheid voor planten wordt door een reeks bodemeigenschappen bepaald. De plantensoort zelf is de dominante factor voor de metaalbeschikbaarheid in het eerste trofische niveau van voedselwebben, aangezien er grote verschillen in metaalopname tussen plantensoorten bestaan.

Metaalverontreiniging in bos- en natuurgebieden in Vlaanderen wordt veroorzaakt door atmosferische depositie tengevolge van industriële activiteit, uitstoot van verkeer, metalen in meststoffen en het storten van afval en sedimenten. Deze soorten bodemverontreiniging kunnen gecategoriseerd worden als diffuse of puntbronnen van verontreiniging. De metaalverontreiniging bij baggergronden is een puntbron, waarbij de verontreiniging duidelijk binnen de perceelsgrenzen beperkt is. Locatiespecifieke ecologische risicoschatting concentreert zich op de effecten van de verontreiniging binnen de grenzen van de locatie. Nochtans kunnen de effecten en biobeschikbaarheid van verontreiniging zich buiten de grenzen manifesteren en een effect hebben op het niveau van het stroomgebied of op een regionale schaal.

Voor locatie-specifieke biomonitoring moeten enerzijds biobeschikbaarheid en bioaccumulatie, en anderzijds effecten op biota, biodiversiteit en processen bepaald worden. De resultaten tonen de bruikbaarheid aan van blad- en strooiselstalen voor locatie-specifieke risicoschatting en biomonitoring. Bepaalde factoren belemmeren echter de toepassing voor regionale biomonitoring. De boomsoortensamenstelling (bijv. de aanwezigheid van wilgen en populieren) bepaalde gedeeltelijk de metaalconcentraties in de strooiselstalen. Bladconcentraties aan metalen in wilgen werden grotendeels door de individualiteit van de boom bepaald. Bij onderzoek onder veldomstandigheden werd vastgesteld dat de duur van overstroming en de redoxpotentiaal een grote invloed heeft op de beschikbaarheid van metalen voor wilgen. Deze factoren worden hoofdzakelijk door lokale omstandigheden en beheersmaatregelen beïnvloed, zijn variabel van jaar tot jaar, en kunnen de bruikbaarheid voor regionale biomonitoring belemmeren. Regionale biomonitoring zou zich moeten toeleggen op bioaccumulatie op hogere trofische niveau in het voedselweb, of moet zeer sterk gestandaardiseerd worden voor het effect van landgebruik en beheer.

Toekomstige onderzoeksnoden en vooruitzichten

1. Het inbouwen van de complexiteit van het voedselweb, de ruimtelijke variabiliteit van voedselbeschikbaarheid, de ruimtelijke variabiliteit van bodemverontreiniging en verschillen in het voedingsgedrag voor doelsoorten in modellen voor ecologische risicoschatting.
2. Het inbouwen van ruimtelijke variabiliteit in plantconcentraties, soortensamenstelling en voedingsgedrag van herbivore doelsoorten in modellen voor ecologische risicoschatting.
3. Gevalstudies rond het effect van verhoogde plantconcentraties (bijv. Cd en Zn concentraties gemeten in wilgen en populieren op verontreinigde baggergronden) op herbivore organismen en wisselwerkingen met hogere trofische niveaus.
4. De ontwikkeling van methodes voor het monitoren van bioaccumulatie bij hogere niveaus in het voedselweb (vogels, zoogdieren).
5. De toepassing van deze monitoringmethodes voor hogere niveaus in het voedselweb (vogels, zoogdieren) voor gebieden met een verschillende type bodemverontreiniging (blootstellingsroute).

6. Evalueren van de impact van het schaalniveau op ecologische risico's: leidt geringe verontreiniging over grote gebieden (diffuse verontreiniging) tot een hoger ecologisch risico dan sterk verontreinigde percelen (puntbronnen) verspreid over een relatief niet-gecontamineerd gebied?
7. Het inzetten van grazers als beheersoptie voor verontreinigde baggergronden in natuurgebieden.
8. De relatie tussen bodemfysische (dichtheid, structuur, permeabiliteit) en bodemchemische eigenschappen voor verontreinigde baggergronden met een variabel hydrologisch regime (afwisseling overstroming en droogvallen)
9. Bepalen van de dynamiek van bodemvormende processen op verontreinigde baggergronden
10. Lange-termijngevolgen van risicobeperkende beheersopties zoals een gepast hydrologisch beheer van verontreinigde baggergronden

Beleidsaanbevelingen

1. Harmonisatie van de wetgeving rond baggerstortterreinen en andere sedimentbodems
2. Ontwikkeling van een referentiekader voor natuurbeheer en natuurontwikkeling in verontreinigde gebieden, waarbij natuurbeheersdoelstellingen geconfronteerd worden met milieudoelstellingen, en de haalbaarheid van het combineren van beide doelstellingen geëvalueerd wordt
3. Ontwikkeling van een geïntegreerde benadering van verontreinigde sedimenten op het niveau van het stroomgebied
4. Regionale biomonitoring van metaalverontreiniging waarbij de klemtoon ligt op de bioaccumulatie in hogere trofische niveaus van het voedselweb, met een sterke standaardisatie voor het effect van het landgebruik en –beheer
5. Het toepassen van passieve biomonitoring en locatiespecifieke evaluatie van pollutie als alternatief voor het formuleren van algemene toelaatbare concentraties in bodems en biota

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