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Retention of Metals Leached From Municipal Solid Waste Incineration (MSWI) Bottom Ashes in Soils

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Utilization of bottom ash in road construction may lead to a release of contaminants that can affect the soil of the swales constructed along these roads. Column tests were performed to evaluate the retention behavior of Cu, Cr, Zn, and Pb, originating from municipal solid waste incineration (MSWI) bottom ash leachate, in two substrates: peat and mould (a cultural soil). A chemical sequential extraction method was used to predict the risk associated with the release of the retained elements with modifications of environmental conditions. Apart from the dissolution of organic matter (OM), ash leachate properties hindered the metal transport from peat. Mould was efficient only in removing Zn, making it a less favorable substrate for the leachate control along the roads. Readily soluble forms made up a minor fraction of the retained metals in peat, reducing the risk of metal release due to ion exchange and pH drop. Changes in redox potential might be the main cause of Zn desorption from peat as the Fe-Mn oxides were the main scavengers for this metal. Oxidation of OM would be the primary reason of Cu and Cr release, while for Pb both fractions (Fe-Mn oxides and OM) might equally contribute to the metal discharge.

Keywords Pollutants, peat, roadside swales, sequential extraction, dissolved organic carbon

Introduction

In urban construction, the utilization of bottom ash from municipal solid waste incineration (MSWI) is increasing in European countries such as Sweden, the Netherlands, Denmark, Germany, France (RVF, 2002; van der Sloot *et al.*, 2001), though its usage may pose a risk to the environment if pollutants such as heavy metals are leached out. A test road was built in Northern Sweden during the summer of 2001 using bottom ash from MSWI as a substitute for gravel (Aberg *et al.*, 2006). The road passes through a peat bog, i.e. is in a vicinity of the organic soil. The soil of grassed swales constructed along the road is the recipient that can modify the mobility of the leached contaminants.

Ash leachate formed by the percolation of rainwater trough the road has high alkalinity and solution ionic strength. The former can lead to the dissolution of soil organic matter (OM) present in the swales, and thus mobilization of metals in the form of soluble metal-OM complexes. The high solution ionic strength, on the other hand, could result in flocculation of Ca-humic acids-metal complexes (ionic strength effect) (Impellitteri *et al.*, 2002) and therefore prevent metal transport from the swales.

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The main objective of this study was to evaluate the retention behaviors of Cu, Cr, Zn and Pb, originating from the MSWI bottom ash leachate, in two substrates: peat and mould. A chemical sequential extraction method was used to predict the risk associated with the release of the retained elements with modifications of environmental conditions, e.g. soil acidification, dominance of oxidizing or reducing conditions.

Materials and Methods

A column test using leachate from bottom ashes was performed in a laboratory to investigate the retention capacity of two substrates for Cu, Cr, Zn and Pb. These metals were included into the investigation due to their high concentrations in ash leachate collected from the experimental road. The leachate used in the laboratory experiment was generated from the MSWI bottom ashes according to the standard leaching test (NT ENVIR 002) at liquid to solid ratio (L/S) of 1 l/kg using double distilled water (ddH_2O, the first distillation by Seradest RO-25 Compact, Seral, Germany, the second by Fistreem Cyclon, Sanyo Gallenkamp PLC, England) acidified with HNO_3 (pro analysis, 65%, Merck, Germany) to pH 4 to simulate rain-water. The bottom ash was of the same origin as the one used for the road.

Five operationally defined metal fractions were extracted from the substrates before and after the column test. The sequential chemical extraction method developed by Tessier *et al.* (1979) was adopted to define four fractions of the metals in the substrates: (I) exchangeable (1 M MgCl, pH 7.0, for 1 h), (II) bound to carbonates (acido-soluble) (1 M NaOAc adjusted to pH 5.0 with acetic acid, for 5 h), (III) bound to Fe-Mn oxides (reducible) (0.04 M NH_2OH-HCl in 25% (v/v) HOAc (pH $\sim$ 2), for 6 h at 96°C) and (IV) bound to organic matter (oxidizable) (0.02 M HNO_3+30% H_2O_2 adjusted to pH 2 with HNO_3 at 85°C for 2 h and at 65°C for 3 h). The residual fraction (V) was determined by dissolving the residue from IV fraction in HNO_3 (65%) at 120°C for 2 h. The total metal concentration of mould and peat was calculated by the sum of the concentrations of the five fractions.

The leachates and extracts were analyzed for metals by inductively coupled plasma optical emission spectroscopy (ICP-OES, Perkin Elmer Otima 2000 DV). Loss on ignition of the substrates was determined by dry combustion at 550°C for 2 h.

The ionic strength of the leachate was calculated from the electrical conductivity (EC) using the Marion-Babcock equation adopted from Chirenje *et al.* (2002): $\log I = 1.159 + 1.009 \log \kappa$, where κ is EC (dS/m).

Dissolved organic carbon (DOC) content was determined according to the European Standard (EN 1484); organic carbon in solution was oxidized to carbon dioxide through boiling and the amount of CO_2 was measured by infrared spectroscopy. Inorganic carbon (IC) of the bulk substrates was determined after the pre-treatment with HCl to eliminate carbonates using a TOC analyzer (TOC-V CSH Shimadzu). Texture of mould was estimated by the hydrometer sedimentation method (SIS, 1992).

The pH, redox potential (Eh) and electrical conductivity (EC) of the substrates were measured in solid-ddH_2O (1:2 w/v) suspensions using a 3M KCl liquid-filled electrode, a Pt-electrode with 3M KCl reference system and a standard-conductivity cell (TetraCon 325), respectively.

The computer software *Statgraphics Plus 5.0* and *SIMCA-P 9.0* (Umetrics) were used respectively for the analysis of variance (ANOVA) and multivariate data analysis (MVDA). In the latter case, projections to the latent structures by means of partial least squares (PLS) method was used to simultaneously relate all the factors with responses by a linear multivariate model (Eriksson *et al.*, 2001).

Table 1
Main properties of the analyzed substrates

	Unit	Substrate	
		Peat	Mould
pH $_{(H2O)}$	—	3.9	6.3
Electrical conductivity (EC)	μS/cm	100	32
Redox potential (Eh)	mV	404	243
Loss on ignition (LOI)	%	94.9	6.4
Organic carbon (OC)	”	74.8	6.2
Texture	”	79*	
Sand	”	ND	38.5
Silt	”	ND	42.0
Clay	”	ND	19.5
Metals			
Cr	mg/kg dw	6.0	22.4
Cu	”	17.3	22.1
Pb	”	8.0	9.8
Zn	”	21.5	50

*Total fraction <2 mm.
ND—not determined.

Soil Substrates

Two soil substrates, peat and mould, obtained from Norrlandsjord, Sweden, were studied (Table 1). Several sections of the swales along the experimental road were covered with mould (a commercially available substrate) and the remaining fractions with peat taken from the adjacent peat bog. According to the von Post peat decomposition classification (Day *et al.*, 1979), the analyzed peat was “almost completely unhumified, containing plenty of fibers.” Mould, or a cultural soil, is an upper layer of podsol enriched in organic matter by the addition of peat and compost. The column test was run using a soil fraction <4 mm. The analyses were performed after sieving and splitting of the air-dried samples. Larger than 4 mm particles made up 2% in mould and 15% in peat and were excluded from the samples.

Column Test

The column test aimed at analyzing the retention capacity of the substrates with the continuous addition of leachate to the columns. The polyethylene columns (bed height was 4 times the internal diameter of the column) were filled with peat and mould and simultaneously run in triplicates (Figure 1). The ash leachate generated in the laboratory and used as a leachant in the column test contained considerably lower concentration of Cr than that from the field experiment (Douard, 2002). Therefore the leachant was spiked with hexavalent chromium (CrO_3 GR for analysis, Merck, Germany) up to 0.4 mg Cr/l to mimic the concentration and speciation of Cr in the first leachate collected from the road. The metal concentrations in the ash leachate are presented in Table 2.

The ash leachate was added with inflow through the bottom of the column (saturated flow) by means of a peristaltic pump (Alitea-XV, Sweden) at the flow rate of L/S

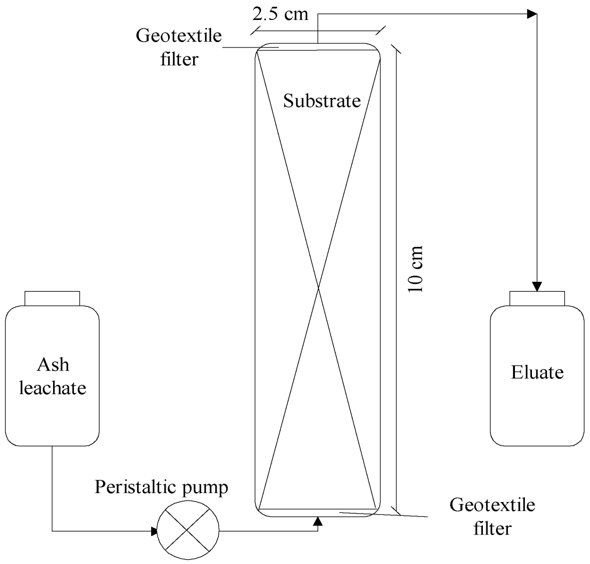

Figure 1. The lay-out of the column leaching test used in the experiment.

10/day on the dry substrate basis (i.e. 10 volumes of the leachant (l) per one mass of dry soil (kg) per day). The columns were operated for 72 h of continuous flow. Geotextile filters (Naue Fasertechnik, Secutex 301 GRK5, Germany) were placed inside the tops and bottoms of the columns to prevent particle washout. Eluates were collected

Table 2
Characteristics of the ash leachate used in the column test

Ash leachate (L/S 1)	Unit	Amount
pH	—	12.7
Electrical Conductivity (EC)	mS/m	29.9
Ionic strength (I)	mol/dm^3	0.42
Metals*		
K	mg/l	1100
Na	”	3740
Ca	”	661
Mg	”	0.3
Cr**	”	0.4
Cu	”	3.8
Pb	”	1.4
Zn	”	0.8
Ni	”	0.06
Cd	”	0.001
Mn	”	<
Fe	”	<

*Permissible levels for Cu and Cr in drinking water in Sweden are <0.05 mg/l; Pb <0.01 mg/; Zn <0.3 mg/l (SNV, 1994).
**Cr concentration after spiking with CrO_3. <-below instrument detection limit.

every 12 h, filtrated through a 0.45 μm nitrocellulose membrane filters (Schleicher & Schuell, Germany) and analyzed with ICP-OES. In total, 140 ml of eluates were collected from each peat column and 1190 ml from each mould column. The amounts of metals retained by substrates were computed by mass balance in mg/kg of dry weight substrate.

Results

Column Test Using Ash Leachate of L/S 1

A continuous load of the ash leachate to the columns did not decrease the metal retention of peat, except for Cu (Figure 2). The retention of Cu reached its lowest point (91%) after 48 h, and increased by a 1% unit at the end of the experiment. In average, 6% of the total added Cu was transported through the columns during the experiment. Pb and Cr concentrations in the eluates were just above the instrument detection limits (4 and 0.4 μg/l, respectively) indicating that retention of these metals was close to maximum. Zn behaved somehow differently from the other metals. The retention of Zn continuously increased during the experiment from 24% to 84%. Mould, in turn, showed a decreasing retention capacity for all analyzed metals (Figure 2).

The concentration of the metals loaded to the columns in all cases had a linear relationship to the cumulative metal retention. The coefficients of the linear regression equations were calculated by a best-fit line method and presented in Table 3.

The eluates from the peat columns had low pH (2.9) at the beginning of the experiment and increased to 11.7 with continuous additions of ash leachate (Figure 3). Despite the changes in pH, the retention of the metals was rather constant. Among the studied metals, only Zn had a positive correlation between the metal retention and the pH (correlation coefficient = 0.8).

The lowered electrical conductivity (EC) in the column eluates compared to the initial EC of the ash leachate indicates the removal of salt ions from the solution. Peat showed relatively constant capacity for salt removal, while mould retained salts poorly (Figure 3). Concentration of Ca in the leachates from peat and mould had a reverse pattern, i.e. the

Table 3

Regression equation coefficients (linear model: $Y = a + b*X$) describing the relationship among the total concentration of the metals added to the columns and the retained amount of the metals (mg/kg dw)

Substrate	Cr	Cu	Pb	Zn
Peat				
Intercept (a)	−0.15	2.4	0.01	−3.83
Slope (b)	0.98	0.92	0.99	0.97
R^2	0.99	0.99	1	0.99
Mould				
Intercept (a)	0.55	6.26	0.34	0.69
Slope (b)	0.70	0.60	0.98	0.91
R^2	0.99	0.99	0.99	0.99

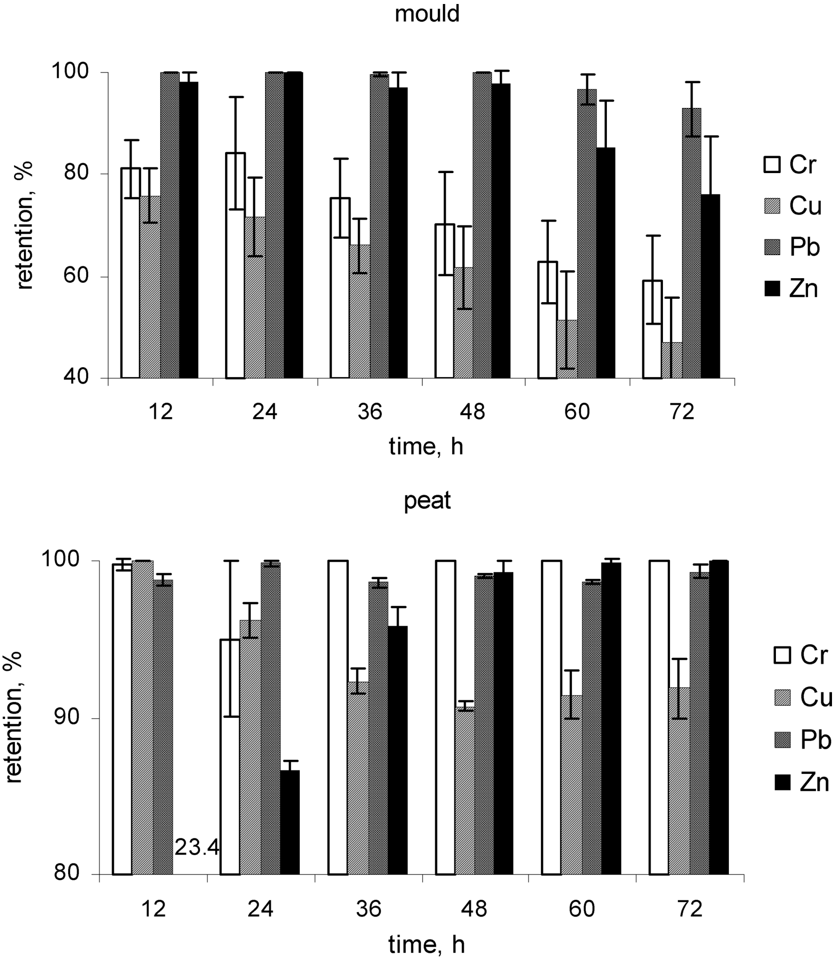

Figure 2. Metal retention efficiency of mould and peat, expressed in percent of the metal concentration added to the columns. Average metal load per 12 h in mg/kg dw was: Cr = 4.8; Cu = 45.6; Pb = 16.3; Zn = 9.2 to the peat columns and Cr = 2.6; Cu = 24.3; Pb = 8.7; Zn = 4.9 to the mould columns. *Note:* Zn retention in peat after 12 h was 23.4%. The error bars represent the standard deviations of the means (n = 3).

leaching of Ca from peat decreased from 406 to 157 mg/l and was the lowest after 36 h of leaching (50 mg/l), while Ca leaching from mould gradually increased throughout the experiment from 184 to 729 mg/l.

The leaching of DOC depended on the organic matter content of the substrates, i.e. higher OM yielded higher DOC in the column eluates (Figure 4). Mould was, however, more depleted in OM than peat; 15.8% of the initial OC was washed-out in the form of DOC from mould, while only 3.6% from peat.

Chemical Sequential Extraction

The fractional distribution of the metals in peat prior to the column test was quite similar for all the metals (Figure 5). The dominating fraction was reducible (bound to Fe-Mn oxides). After the column test, considerable changes in the metal fraction distribution occurred.

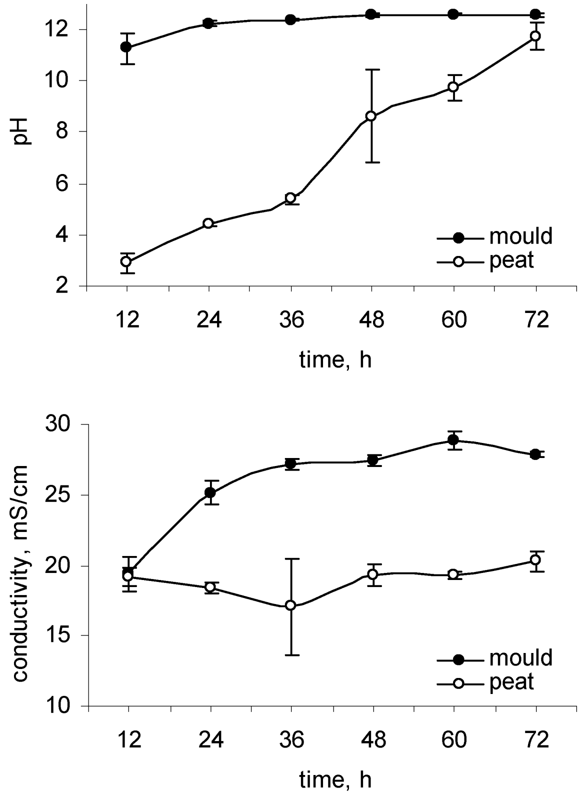

Figure 3. The pH and electrical conductivity (EC) of the eluates from the analyzed substrates: (●) mould and (○) peat. Initial ash leachate EC = 29.9 mS/cm and pH = 12. The error bars represent the standard deviations of the means (n = 3).

The fraction of Cu, Cr, and Pb bound to organic matter significantly increased, especially for Cu, while for Zn it changed insignificantly. The largest fraction of Zn remained the same as prior to the column test, i.e. bound to Fe-Mn oxides. The acido-soluble (operationally defined as carbonate) fraction was more significant for Zn and Pb than for Cu and Cr.

After the column test with mould, the largest concentrations of Cu, Pb, and Zn were bound to Fe-Mn oxides. Cr occurred mainly in the residual fraction both prior to and after the ash leachate percolation through the columns. The increase in the fraction bound to OM was most considerable for Cu, while carbonate fraction was significant for the retention of all the metals passing through the mould columns.

Lead was the only metal undetected in the exchangeable fraction of both substrates after the column test.

Discussion

Metal Retention in the Substrates

Metal retention in the roadside swales depends on the properties of the soil covering them. Since the road passes a peat bog, the interaction of the metal ions with OM is of particular

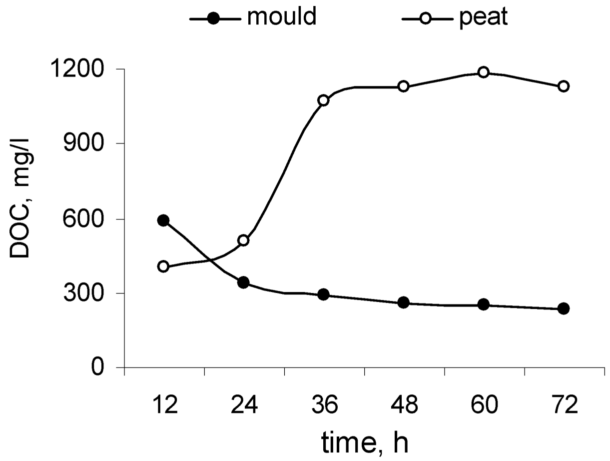

Figure 4. Leaching of dissolved organic carbon (DOC) from the columns filled with (●) mould and (○) peat.

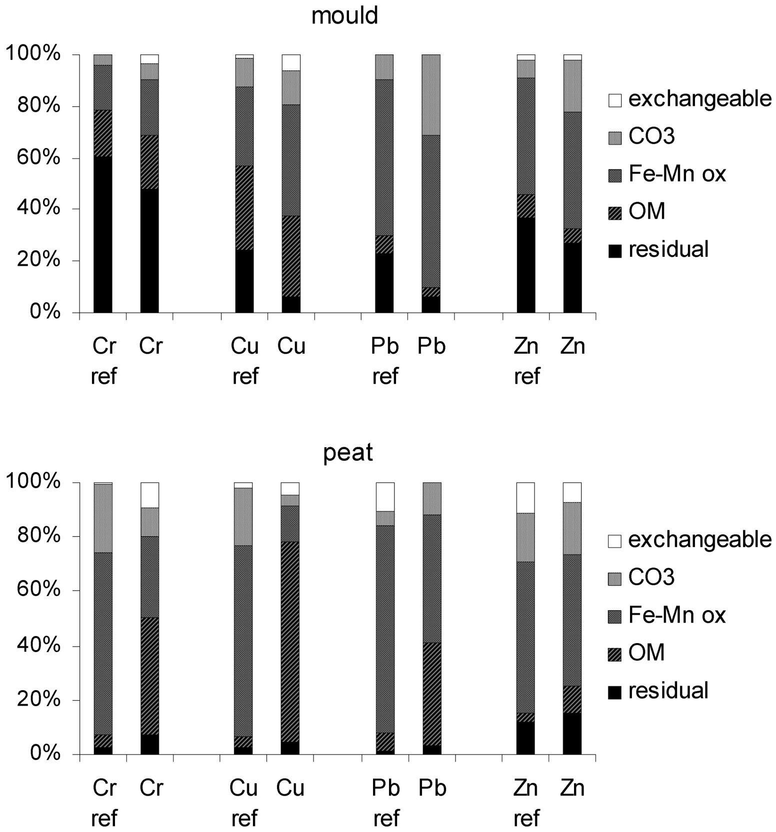

Figure 5. The metal distribution between the operationally defined fractions in the substrates prior to (ref) and after the column test.

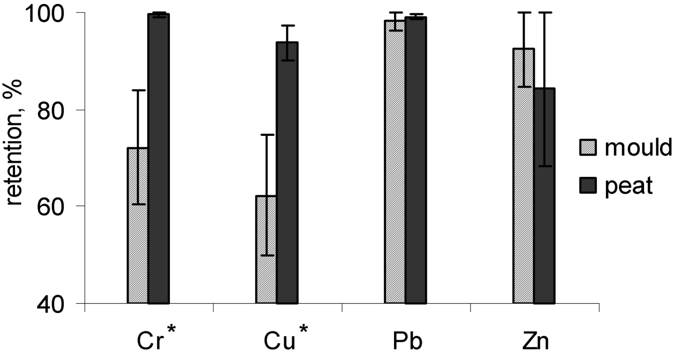

Figure 6. Comparison of average metal retention in mould and peat. Symbol * represents a significant difference between two substrates in the metal retention ($\alpha = 0.05$). The error bars represent the standard deviations of the means (n = 3).

importance. Pb and Cr has just as high an affinity for OM as Cu, therefore considerable portions of Pb, Cr and Cu were retained by peat even at pH 3 (Figures 2 and 3). The sequential extraction confirms the importance of OM for Pb, Cr and Cu binding in peat (Figure 5). Solid metal-organic complexes of Zn are reported to be unstable below pH 5 (Brümmer *et al.,* 1986), explaining the lowest Zn retention in peat during the first 24 h of the experiment when pH of the eluates was <5 (Figures 2 and 3). According to Kabata-Pendias and Pendias (2001), the adsorption of Zn can be reduced at pH <7 by competing cations, resulting in the mobilization and leaching of Zn from acid soils (e.g. peat). Besides, the higher peat selectivity for Pb and Cu could weaken the Zn retention, especially when Zn concentrations in the ash leachate were exceeded by that of Cu 6 and by Pb 2 times (Table 2). The order of the Pb, Cu and Zn retention was in agreement with that observed by Jang *et al.* (2005) in an organic substrate tested as a filter material for urban stormwater runoff.

Contrary to peat, mould was most efficient in Zn retention (Figure 6). Carbonates and Fe-Mn oxides were the main scavengers of Zn from the solution. The order of the metal retention strength in mould was as follows: Pb > Zn > Cr > Cu (Figure 6). Usually, the stability of metal-humic complexes reverses the order of Cu and Zn, confirming that inorganic constituents of the mould were more active in the Zn retention.

Leaching of elements from the road decreases with time as their available fraction in ashes becomes depleted (Aberg *et al.*, 2006). The time period simulated by the column test, which was calculated by converting L/S into time scale of average water infiltration (200 mm/y), would correspond to approximately 150 years of the swale operation time. In this experiment, the columns were overloaded with regard to the expected contaminant discharge from the road by continuously adding the ash leachate highly concentrated with heavy metals. However, a point of breakthrough of peat was not reached. By the end of the experiment, in contrast to mould, the retention of Cr and Pb by peat was at its maximum and for Zn and Cu continued to increase. Therefore, peat exhibited better properties than mould as filling material for roadside swales when considering the retention of the investigated metals in a long-term perspective.

Dissolved Organic Carbon (DOC) and Metal Transport

Although alkaline conditions can lead to the dissolution of soluble organics and thus increase the mobility of metals by the formation of soluble metal-OM complexes, this was not

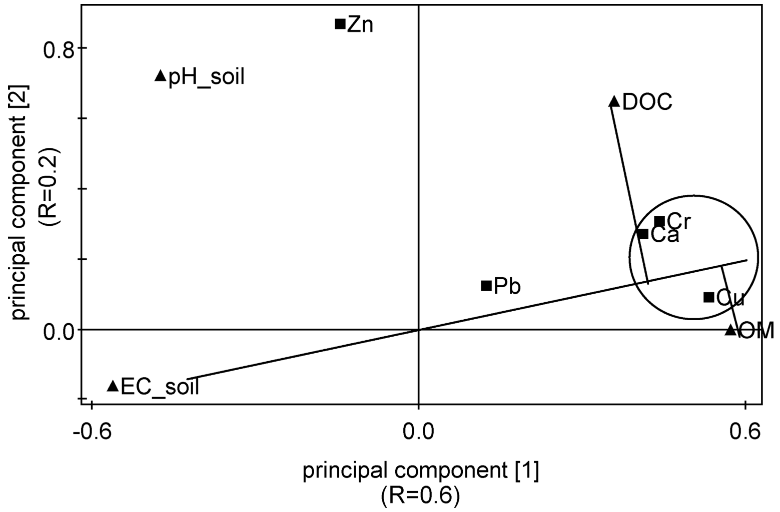

Figure 7. Loading scatter plot of PLS analysis of (■) retained metals in relation to (▲) the factors. OM—initial organic matter content of the substrate; DOC—dissolved organic carbon and EC—electrical conductivity of the column eluates.

observed in our study. By contrast, the increasing Cu and Cr retention with more organic carbon being dissolved was observed. The DOC as a factor and the retained Cu and Cr as responses are distant to the same direction from the origin, indicating a positive correlation between them (Figure 7). Such a pattern contradicts the results obtained by other authors (Hsu and Lo, 2000; Chirenje and Ma, 1999), who observed that the increase in DOC resulted in higher Cu leaching.

The high ionic strength of ash leachate can modify the metal mobility in the presence of elevated concentrations of DOC. High content of the major cations and especially Ca in solution might result in flocculation of humic acids-metal complexes and their precipitation (ionic strength effect), thus preventing metal transport (Chirenje *et al.,* 2002; Impellitteri *et al.*, 2002). Addition of Ca to the DOC extracts can flocculate up to 50% of the DOC originally present in the samples (Romkens and Dolfing, 1998). The observed positive correlation between the retention of Ca and the metals (Figure 7) may support the assumption of the ionic strength effect. Conversely, the retention of Ca at such a high pH range (11, 12) as it was observed at the end of the column test (Figure 3) can be due to the precipitation of calcium carbonates. However, the Ca leaching from mould was the highest at the end of the experiment (reaching 406 mg/l or 61% of the total loaded Ca), suggesting that Ca retention at a similar pH in peat was due to other mechanisms, possibly the above mentioned precipitation of Ca-DOM-metal complexes.

The retention of the metals at high pH can also be due to their precipitation. Cu and Pb can precipitate from the bottom ash leachates by forming hydroxides and low solubility (at $\text{pH} > 7.5$) having secondary minerals (e.g. tenorite CuO) (Meima *et al.*, 1999). The leaching of Cu and Pb usually reaches their lowest point at pH 9-10 and then increases again due to the formation of OH^- complexes (van der Sloot *et al.*, 1997). Since the leaching of Pb from peat was lower at pH 11.7 (9 μg/l, or 0.7 % of the total loaded Pb) than at pH 9.7 (18 μg/l), the formation of mixed metal-ligand complexes and co-sorption reactions in the substrate matrix may have accounted for the metal retention at high pH.

If the ionic strength effect was a mechanism responsible for the metal retention in our study, then it is possible that the increase in DOC of the peat eluates was only due to the increase in the lower molecular weight FA fraction that was not engaged in metal binding (except for Cu) and/or precipitation. Otherwise, much higher metal concentrations, e.g. of Pb, which is more readily bound to humic (HA) than fulvic acids (FA) (Jordan *et al.*, 1997), could be expected in the eluates. Since Cu has a high affinity for both humic substances and the only forms of dissolved Cu at high pH ($\text{pH} > 8$) are HA and FA complexes (Wu *et al.*, 2002), the transport of dissolved Cu (6% of total added Cu) at the high electrolyte concentrations likely occurred as complexes with FA. High molecular weight HA can coagulate easier and be expected to precipitate from the solution as Ca-DOM-metal complexes (Stevenson, 1994).

According to this investigation, potential negative effects on the release of metals due to the increase in DOC caused by the alkaline properties of the ash leachate were attenuated possibly through the leachate ionic strength. Precipitation of new mineral phases may have contributed to the metal retention at high pH.

Predictions of the Metal Desorption

Conditions in the swales may change over time, varying between dry and flooded, acidic (due to rain) and alkaline (due to ash leachate). De-icing salts or a new portion of ash leachate after a dry season can affect the ionic strength of the soil solution. As a result, the exchangeable fraction of the metals that made up 2–10% in the analyzed substrates (Figure 5) can be released.

Weathering of ashes and interaction with atmospheric CO_2 lowers pH of the leachate (Ecke, 2001). Consequently, the soil solution pH is also expected to decline. If pH gets below 5, e.g. due to extended period of acid rain, the fraction bound to carbonates may be released. In such a case, a relatively large fraction of Pb (31%) could remobilize from mould. In general, Pb and Zn would be more sensitive to the pH drop than Cr and Cu.

The main risk for release of the retained metals is related to changes of oxidizing-reducing conditions. Although the significant fractions of the metals in mould are associated with Fe-Mn oxides, it would take more than just soil saturation with water to release the bound metals. Redox potential in peat can more readily decrease than in mould due to higher OM content and microbial activity. In the case of redox drop when Fe and Mn oxides cannot be stable any longer due to the lack of oxygen, a release of the major Zn fraction would occur from both substrates. By contrast, the oxidation and decomposition of soil OM would be the main cause of the large quantities of Cu (73%), Cr (43%), and Pb (37%) to be released from peat. If a vegetation cover is established in the swales, the content of soil OM is likely to be maintained. An accumulation of OM is more likely in this geographical region due to relatively low temperatures, water excess and slow mineralization.

The residual fraction that was larger for Cr and Zn than for Cu and Pb (Figure 5) is usually considered as not available for leaching and therefore bears no risk for release of the metals defined in this fraction (Tessier *et al.*, 1979).

The highest retention of metals in roadside swales would be achieved using peat as a filling material. To maintain the filtering function of the swales, the establishment of vegetation cover promoting the accumulation of OM and prevention of strongly oxidizing conditions is recommended.

Conclusions

Although the substrates were not absolutely effective in the metal retention, the concentrations of the metals leached through the columns (in a range of μg/l) were much lower compared to those in the ash leachate. Apart from the dissolution of OM, ash leachate properties hindered the metal transport from peat. The decrease of the metal concentrations in solution could be due to the flocculation of metal-humic acid complexes, caused by the presence of a high concentration of Ca (ionic strength effect) in the ash leachate. Mould, however, is a less favorable substrate for the leachate control along the roads since it was only efficient in the removal of Zn, considered least toxic among the analyzed metals. Peat in turn was capable of reducing the concentrations of Cr, Pb, and Zn to permissible drinking water levels in Sweden; Cu concentrations though were not sufficiently low. High Cu affinity for OM still poses some risk of Cu leaching as complexed with DOC even under the effect of the ionic strength. Using peat as a cover substrate of the swales, additional Cu stabilizing material would be recommended. Readily soluble forms composed a minor fraction of the retained metals in peat, reducing the risk of metal release due to ion exchange and pH drop. A decrease in redox potential might be the main cause of Zn desorption from peat as the Fe-Mn oxides were the main scavengers for this metal. For Cu and Cr, the oxidation of OM would be a primary reason for the metal release, while for Pb both fractions (Fe-Mn oxides and OM) would equally contribute to the metal discharge.

Acknowledgments

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