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ORIGINAL ARTICLE

Influence of compost maturation time on Cu and Zn mobility (M_F) and redistribution (I_R) in highly contaminated soil

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Abstract This study examined how maturation time (3, 6 and 12 months) of sewage sludge compost added to soil contaminated with heavy metals (HMs), i.e., Cu and Zn affects: (1) the redistribution pattern of HMs; (2) metal mobility (as mobility factor, M_F) and metal stability (as reduced partition index, I_R); (3) rate constants of metal transformations in exchangeable and acid-soluble ($F1$) fraction and rate constants of metal redistribution in all fractions ($F1$–$F4$). Soil without amendments was used as a control. It was shown that compost maturation time did not affect metal redistribution. After 12 months of incubation in non-amended soil, Cu still showed medium mobility ($M_F = 16.5$ %), while in amended soil it had low environmental risk ($M_F = 6.2$ %). In contrast, Zn was highly mobile ($M_F = 43.4$ %) in all treatments. Compost addition favored only Cu redistribution into more stable fractions. For both metals, the rate constants of redistribution were an order of magnitude lower than rate constants of metal transformations in the $F1$ fraction.

Keywords Sewage sludge compost · Maturation time · Metal redistribution · Mobility factor (M_F) · Reduced partition index (I_R)

Introduction

Soil contamination is a worldwide problem. The number of contaminated sites is still increasing and depends on the degree of industrialization of a given country. For comparison, the estimated maximum area of current potentially contaminating industry in Germany is 17,851 km^2, whereas in Poland it is 9381 km^2 (Perez 2012). In many polluted sites, heavy metals are one of the predominant contaminants. Due to the properties of degraded soils, i.e., low content of organic matter and clay minerals, metals can show excessive mobility and bioavailability. Therefore, such areas need urgent remediation.

Metal stabilization in soil through the addition of organic amendments is one of the existing remediation methods for reducing harmful effects in contaminated sites (Soler-Rovira et al. 2010). This technique is typically carried out in situ and can even be used for sites with a large area. An effective amendment should reduce metal bioavailability in soil as much as possible. For this purpose, industrial waste (e.g., coal fly ash), sewage and paper mill sludge, gypsum- and lime-rich industrial by-products have been used (Kumpiene et al. 2008).

In the last decade, the use of composted organic amendments such as agricultural waste (e.g., animal manure, green waste) (Liu et al. 2009) or municipal solid waste (Farrell and Jones 2010) has been extensively investigated. Some authors also used sewage sludge compost as it is cost-effective and readily available because of the increasing volume of sewage sludge and the necessity for its management (Vaca-Paulín et al. 2006).

The application of composted organic amendments is especially necessary for metal redistribution from labile (water-soluble and exchangeable) to less-available fractions (organic and residual) (Pérez-Esteban et al. 2012). In

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particular, the concentrations of humic substances (HS) and proportion between humic acids (HAs) and fulvic acids (FAs) in organic amendments are important (Clemente and Bernal 2006; Pérez-Esteban et al. 2012). This is because HS provide numerous non-specific and specific sites for metal adsorption. Formation of insoluble organometallic complexes with metals can decrease its mobility and uptake by plants (Udom et al. 2004).

Until now, most studies of metal immobilization in soils amended with compost have focused on how metal distribution is influenced by composts made from different organic materials. Various compost origins directly affect their physico-chemical properties (i.e., pH, organic matter, humic substances) which, in turn, influence the efficiency of metal transformations among individual fractions in amended soils (Pérez-Esteban et al. 2012). However, it is known that compost properties greatly depend on the duration of the maturation phase during composting. The length of the maturation phase varies, because it depends on the type of composted waste and technological parameters of composting. However, the optimum time for compost maturation has not been clearly specified.

Organic amendments used for metal immobilization should characterize suitable pH, cation exchange capacity, content of organic matter and humic substances (fulvic and humic acids). Less-mature compost has a lower total content of humic substances and a lower HAs-to-FAs ratio. As compost matures, the content of HS and the ratio between humic and fulvic acids increases (Paredes et al. 2002; Jouraiphy et al. 2005; Kulikowska and Klimiuk 2011). A higher content of HS has been shown to favor the stability of cadmium (Tapia et al. 2010).

It is known that during composting, transformations of organic matter, including its humification, occur most intensively over the few months of the process and then the rate of humification (formation of humic substances) decreases. However, in this time, polymerization may take place, which means the formation of complex molecules of HA from more simple molecules of FA. Because humic acids form more stable complexes with heavy metals, it is reasonable to examine whether long-term compost maturation (which influences the degree of polymerization and, consequently the increase in humic acids content), improves the efficiency of metal transformations in soil.

The content of the exchangeable and acid-soluble fraction dictates metal mobility and may be described by mobility factor (M_F). However, metal transformations in all individual fractions concern their redistribution and are expressed by reduced partition index (I_R). The usability of both indices has already been confirmed to assess metal redistribution in contaminated soils (Miretzky et al. 2011; Zheng and Zhang 2011). However, in most studies, the efficiency of metal immobilization in soils amended with different materials is evaluated mainly on the basis of standard leaching tests using specific extractants such as $CaCl_2$ (Houben et al. 2013), DTPA and $Ca(NO_3)_2$ (Khan and Jones 2009) or analysis of the metal distribution in soil at the beginning and the end of incubation (Gadepalle et al. 2009; Hanc et al. 2009). In numerous studies, the effect of soil amendments was assessed through metal phytoavailability (van Herwijnen et al. 2007; Liu et al. 2009; Hanc et al. 2009). Only a limited number of studies have dealt with the M_F or I_R indices to measure metal transformations in amended soils (Han et al. 2003; Gusiatin 2012). Moreover, the rate constants of heavy metal transformations in soil, i.e., mobility and redistribution have not been calculated, despite the fact that some authors emphasize that all metal transformations are time-dependent.

In the context of the above, the aims of this study were: (1) verification of whether compost maturation time (3, 6, 12 months) affects Cu and Zn transformations in amended soil during one year incubation; (2) estimation of changes in metal mobility (as M_F) and their stability (as I_R) over time (3) calculation of the rate constants of metal transformations in the exchangeable and acid-soluble fraction and the rate constants of metal redistribution.

Materials and methods

Soil sampling and contamination

The uncontaminated surface soil samples (depth 0–30 cm) were collected from an agricultural area in Wanguty, Warmia and Mazury Province in north-eastern Poland. The soil classified as sandy clay loam (48 % sand, 24 % silt, 28 % clay) in texture was air-dried and ground to pass through a 1-mm sieve and was then homogenized and spiked with Cu and Zn.

The soil spiking procedure was adapted from Thawornchaisit and Polprasert (2009). A mixture of 3.6602 g $Cu(NO_3)_2 \cdot 2.5\ H_2O$ and 9.0963 g $ZnN_2O_6 \cdot 6H_2O$ (Sigma-Aldrich) was dissolved in 1 L of distilled water and added to 1 kg of soil. To homogenize the soil–water mixture, it was shaken overnight (100 rpm) on a mechanical, horizontal shaker. The mixture was then left at room temperature for a month with frequent, thorough mixing and rinsing with distilled water. Finally, the contaminated soil was air-dried, ground manually and passed through a 1-mm sieve and stored for analysis. In all, 10 kg of soil was contaminated with metals.

Organic amendments

Compost from municipal sewage sludge and lignocellulosic materials (feedstock composition: 60 % sewage sludge, 5 % inoculation in the form of compost matured for 12 months, 15 % wood chips, 15 % straw rape, 5 % grass) was used as soil organic amendment. The composting process was conducted in a two-stage system: the first system used an aerated bioreactor and the second used a periodically turned windrow. A detailed description of composting technology with the scheme of the bioreactor was given by Kulikowska and Klimiuk (2011). In present study, three sewage sludge composts were used, differing in maturation time in the windrow (3, 6 and 12 months). The compost samples, collected at different time intervals, were air-dried and ground in a RETSCH SM-100 cutting mill (0.5 mm) to homogenize them before they were characterized.

Pot experiments

The spiked soil was subsequently amended with composts in 3.5-L plastic vessels covered with porous plastic membrane. The mixtures contained 80 % soil and 20 % compost (w/w, equivalent to 600 t/ha). Four experimental series were performed: non-amended soil (series 1), soil with compost of 3-month maturation (series 2), soil with compost of 6-month maturation (series 3), and soil with compost of 12-month maturation (series 4).

After compost addition, the samples were mixed for 2 h using a mechanical shaker, deionized water was then added and the pot experiments were continued at ambient temperature. The soil mass, amendments and water volume used in individual series are given in Table 1. During incubation, the treated soils were maintained at 60–70 % of the maximum water holding capacity with distilled water by regular weighing of the vessels and adding an appropriate volume of distilled water (once a week). About 400 g of soil sample (wet weight) was collected from each pot at time intervals of 0; 1; 3, 6; 9 and 12 months. The soil samples were dried at 105 °C, and ground to pass through a 1-mm sieve before physico-chemical characterization. Each treatment was performed in three replicates.

Physico-chemical analysis

Soil, amendments and the prepared mixtures were analyzed for several properties. The pH was determined in 1 M KCl extracts (1:2.5 w/w for soil and 1:5 w/w for amendments) using a pH-meter (Hanna Instruments). Cation exchange capacity (CEC) for soil and composts were calculated as the sum of the hydrolytic acidity (in 1 mol/L $Ca(CH_3COO)_2$) and the exchangeable bases (in 0.1 mol/L HCl) (Ostrowska et al. 1991). Total organic matter (OM) was measured by ignition of samples at 550 °C (PN-Z-15011-3:2001); total organic carbon was measured with TOC analyzer (VCSN, Shimadzu). Humic substances (HS), humic acids (HAs) and fulvic fraction (FF) were isolated according to conventional procedure described by Boratyński and Wilk (1965). Before extracting the HS, the samples were extracted with distilled water to eliminate soluble non-humic substances (e.g., sugars and proteins) (Amir et al. 2006). The samples were then defatted with a mixture of chloroform and methanol in a microwave oven according to the procedure provided by Jouraiphy et al. (2005).

The total metal concentrations in all samples were determined through digestion with aqua regia in a microwave oven (MARSXpress, CEM USA). For this purpose, 1 g of non-amended/amended soil or 0.25 g of composts (dried at 105 °C) were weighed and placed in polytetrafluoroethylene vessels, treated with $HCl:HNO_3$ mixture (Sigma-Aldrich) at 3:1 ratio (v/v) and 2 ml of 30 % H_2O_2 and were then heated using a one-stage microwave program ($T = 170$ °C, $P = 800$ W, $t = 30$ min). After cooling, the extracts were filtered through Whatman 42 filter papers (pore size 8 µm) into 50-mL glass flasks and filled to the mark with distilled water. Metal concentrations in the extracts were measured by a flame atomic absorption spectrometer (FAAS) (Varian, AA28OFS).

For metal fractionation in non-amended/amended soil, a modified BCR sequential extraction procedure (Pueyo et al. 2008) was adopted. Four operationally defined fractions were determined for each metal: exchangeable and acid soluble (*F*1), reducible (*F*2), oxidizable (*F*3) and residual (*F*4). All analyses were performed in triplicate.

Calculations

Humification progress

To assess the humification degree of organic matter in composts and non-amended/amended soil, humification

Table 1 Design of pot experiments

Series	Water holding capacity (%)	Mass of contaminated soil (g)	Mass of organic amendment (g)	Volume of added water (ml)
1	88.6	2400	0	1382
2	96.2	1920	480	1501
3	98.0	1920	480	1541
4	96.5	1920	480	1505

indices were calculated using the following equations (Sánchez-Monedero et al. 1999; Paredes et al. 2002; Bustamante et al. 2008):

$$\text{Humification ratio HR} = (C_{\mathrm{HS}}/C_{\mathrm{TOC}}) \times 100 \quad (1)$$

$$\text{Humification index HI} = (C_{\mathrm{HAs}}/C_{\mathrm{TOC}}) \times 100 \quad (2)$$

$$\text{Percentage of HA } P_{\mathrm{HA}} = (C_{\mathrm{HAs}}/C_{\mathrm{HS}}) \times 100 \quad (3)$$

$$\text{Degree of polymerization DP} = C_{\mathrm{HAs}}/C_{\mathrm{FAs}} \quad (4)$$

where C_{HS}, C_{HAs}, C_{FAs}, C_{TOC} is concentration of humic substances, humic acids, fulvic acids and total organic carbon, respectively.

Metal transformations in soils

In order to access the relative metal mobility and bioavailability in soil, the M_{F} factor was used, defined as the ratio of the metal concentration in the mobile fraction to the sum of the metal concentration in all fractions:

$$M_{\mathrm{F}} = \frac{F1}{F1 + F2 + F3 + F4} \times 100\ \%, \quad (5)$$

where $F1$–$F4$ is the metal concentration in individual fractions (mg/kg) according to the BCR procedure.

Based on the M_{F}, metal mobility is classified into four categories: low ($1\ \% \le M_{\mathrm{F}} \le 10\ \%$), medium ($10\ \% < M_{\mathrm{F}} \le 30\ \%$), high ($30\ \% < M_{\mathrm{F}} \le 50\ \%$) and very high ($M_{\mathrm{F}} > 50\ \%$) (Rodríguez et al. 2009).

The relative binding intensity of metals based on their fractionation is expressed by reduced partition index (I_{R}) using the following equation (Han et al. 2003):

$$I_{\mathrm{R}} = \frac{\sum_{i=1}^{k} i^2 F_i}{k^2}, \quad (6)$$

where i is the index number of the extraction step, progressing from 1 for $F1$ fraction to 4 for $F4$ fraction (in the BCR procedure, $k = 4$), F_i is the percentage content of the considered metal in fraction i.

To simulate the transformations of metal mobility and their redistribution in non-amended and amended soils, regression equations were used:

$$M_{\mathrm{F}} = a_{\mathrm{m}} \times t + b_{\mathrm{m}} \quad (7)$$

$$I_{\mathrm{R}} = a_{\mathrm{r}} \times t + b_{\mathrm{r}}, \quad (8)$$

where a_{m} is the rate constant of transformations for $F1$ fraction (metal mobility), a_{r} the rate constant of redistribution for $F1$–$F4$ metal fractions (metal stability); for linear relationship (0 order kinetic), rate constant of transformations is equal to transformation rate, b_{m} is the initial metal mobility in soil, b_{r} the initial metal stability in soil, t the stabilization time.

Data were statistically evaluated using STATISTICA 9.0 (StatSoft, Inc.). Duncan's test was used and $p < 0.05$ was considered to be significantly different.

Results and discussion

Properties of soil, composts and mixtures of soil with composts

Table 2 shows the main properties of soil, composts and mixtures of soil with composts.

The soil characterized with acidic pH, organic matter content was 10.1 % and a cation exchange capacity (CEC) 20.5 cmol(+)/kg. The content of humic substances (HS) was relatively low; while concentrations of humic acids (HAs) and fulvic fraction (FF) were comparable. The DP (i.e., $C_{\mathrm{HAs}}/C_{\mathrm{FAs}}$), reflected the ratio between complex molecules (HAs) and more simple molecules (FAs) was on the level of 1.01, whereas HR (i.e., $C_{\mathrm{HS}}/C_{\mathrm{TOC}} \times 100$) was 51.6. In non-contaminated soil the concentration of Cu and Zn was 32 (±3.5) and 67 (±3.4) mg/kg, respectively. After spiking, total Cu and Zn concentrations increased to 992.3 (±44.9) mg/kg and 2062.1 (±41.4) mg/kg, respectively (Table 2). These concentrations exceeded permissible values for industrial areas according to Polish law (OME 2002), indicating high metal contamination.

All organic amendments had pH close to neutral and contained over fivefold higher organic matter than non-amended soil (Table 2). They contained also more humic substances (HS): from 98.5 mg C/g (3-month compost) to 114.4 mg C/g (12-month compost). With the increase of maturation time, the content of HAs and values of DP (i.e., $\mathrm{C_{HAs}/C_{FAs}}$) and HI (i.e., $\mathrm{C_{HAs}/C_{TOC}} \times 100$) increased in all composts (Table 2). This means that humification process proceeded during the whole maturation time of compost (12 months). Taking into account that the highest increase in the content of HS in 3- and 6-month compost was observed (compared to feedstock, data not shown), it may be assumed that the process of HS formation proceeded most intensively for 6 months of maturation. With compost maturation time up to 12 months, there was no increase in concentration of HS. However, in this time, complex molecules of HA, from more simple molecules of FA, were formed. This was confirmed by increase of degree of polymerization in compost (based on DP index). Because complexes between heavy metals and humic acids are more stable than those with fulvic acids, the usability of compost used for soil amendment is affected not only by humic substances content, but also by humic acids.

Table 2 Properties of composts used as amendments, soil and soil mixed with composts (mean ± standard deviation, $n = 3$)

Properties	Unit	Compost of 3-month maturation	Compost of 6-month maturation	Compost of 12-month maturation	Non-amended soil (series 1)	Soil with compost of 3-month maturation (series 2)	Soil with compost of 6-month maturation (series 3)	Soil with compost of 12-month maturation (series 4)
pH_{KCl}	–	6.7 (±0.3)	6.8 (±0.4)	6.8 (±0.3)	5.9 (±0.2)	6.17 (±0.4)	6.07 (±0.3)	6.11 (±0.3)
OM	%	57.2 (±0.65)	54.8 (±0.42)	52.2 (±0.81)	10.1 (±0.32)	21.8 (±0.9)	22.8 (±0.75)	21.7 (±0.82)
TOC	mg/g	295.0 (±25)	302.0 (±32)	291.0 (±27)	32.9 (±3.6)	78.6 (±8.4)	83.4 (±9.3)	78.5 (±7.5)
CEC	cmol(+)/kg	42.1 (±2.4)	46.3 (±3.5)	49.5 (±4.1)	20.5 (±1.2)	–	–	–
HS	mg C/g	98.5 (±4.1)	113.4 (±6.2)	114.4 (±7.6)	17.0 (±1.3)	31.5 (±2.3)	34.8 (±2.8)	33.9 (±2.5)
FF	mg C/g	53.8 (±2.4)	58.6 (±1.3)	53.7 (±2.0)	8.4 (±0.6)	16.7 (±1.4)	18.1 (±1.6)	16.6 (±1.4)
HAs	mg C/g	44.7 (±3.1)	54.8 (±2.8)	60.7 (±3.3)	8.5 (±0.7)	14.8 (±1.1)	16.7 (±1.4)	17.3 (±1.6)
DP	–	0.83	0.94	1.13	1.01	0.89	0.92	1.04
HR	%	33.4	37.6	39.3	51.6	40.1	41.7	43.2
HI	%	15.1	18.2	20.9	25.9	18.8	20.0	22.0
P_{HA}	%	45.4	48.3	53.1	50.3	47.0	48.0	51.0
Cu	mg/kg	61.3 (±5.3)	54.1 (±4.6)	57.6 (±7.6)	992.3 (±44.9)	871.6 (±46.4)	888.8 (±46.4)	846.3 (±49.0)
Zn	mg/kg	255.4 (±17.1)	237.6 (±27.4)	253.7 (±28.6)	2062.1 (±41.4)	1725.1 (±86.6)	1698.5 (±66.9)	1709.1 (±88.5)

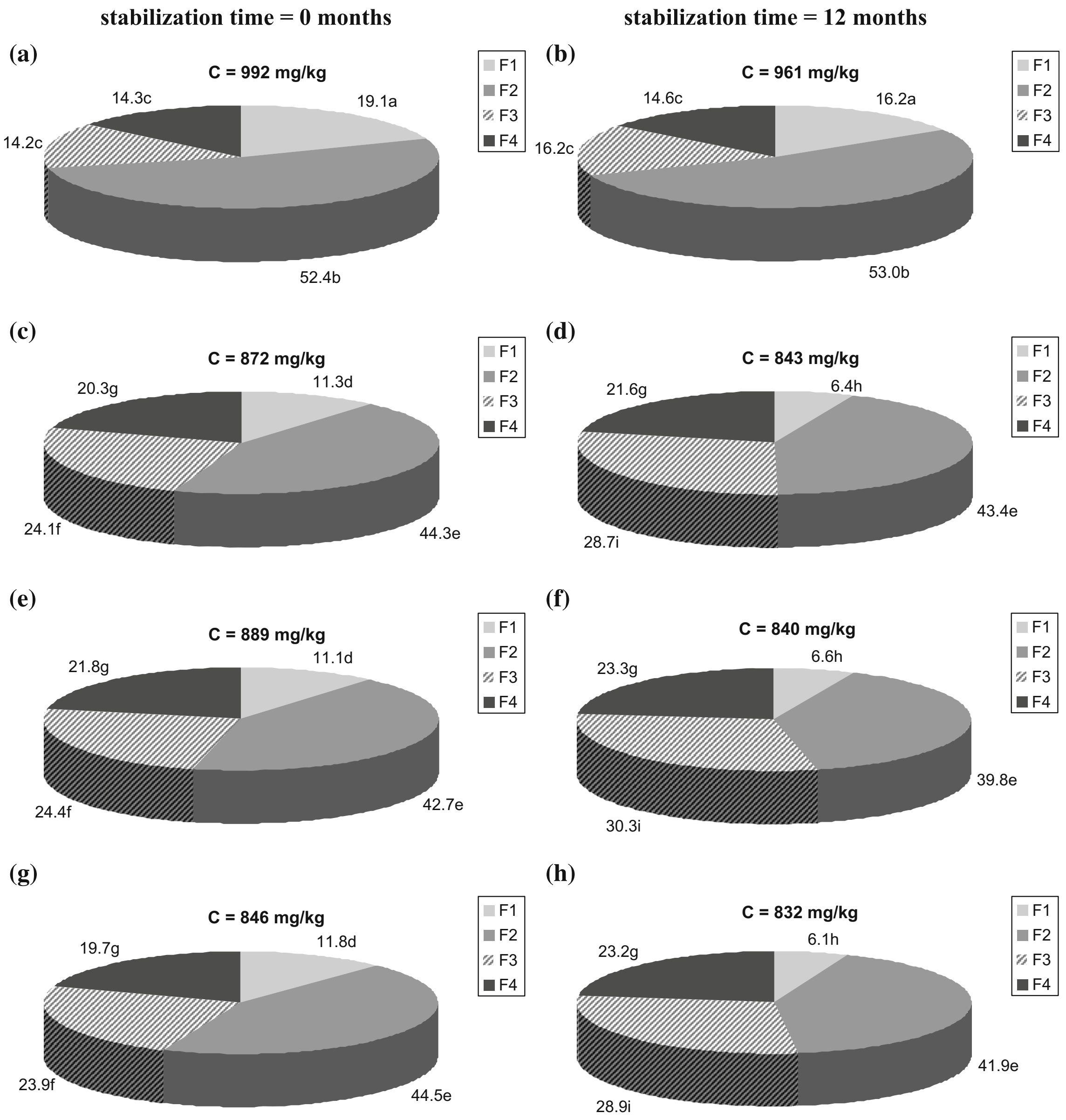

Fig. 1 Percentage content of Cu in individual fractions (*F*1–*F*4) in non-amended and compost-amended soils at the beginning and the end of stabilization: **a**, **b** non-amended soil (control, series 1); **c**, **d** soil with compost of 3-month maturation (series 2); **e**, **f** soil with compost of 6-month maturation (series 3); **g**, **h** soil with compost of 12-month maturation (series 4). Mean values ($n = 3$) between the same fractions at $t = 0$ and 12 months in individual series followed by the same letters do not differ significantly at $p < 0.05$

Total Cu and Zn were below guideline limits for organic fertilizers, similar to other metals. The average concentrations of Cd, Cr, Hg, Ni and Pb were 0.8 (±0.13), 53.5 (±3.4), 0.041 (±0.01), 24.1 (±3.1) and 9.1 (±0.8) mg/kg, respectively. Therefore, tested composts can be used as soil amendments.

Compost addition to soil resulted in a greater than twofold increase in organic matter content and humic substances compared to non-amended soil (Table 2). The content of HAs and DP values in individual series depended on the compost maturation time. The highest content of humic acids and the highest DP index were obtained in series 4, at which the most mature compost was used. In all mixtures, there was a slight decrease in total Cu and Zn concentration, compared to non-amended soil. This is because metal concentrations in composts were significantly lower than in soil.

Metal distribution in non-amended and amended soils

In Figs. 1and 2, metal distribution (as percentage content of individual fractions) determined according to sequential extraction procedure of BCR, is shown.

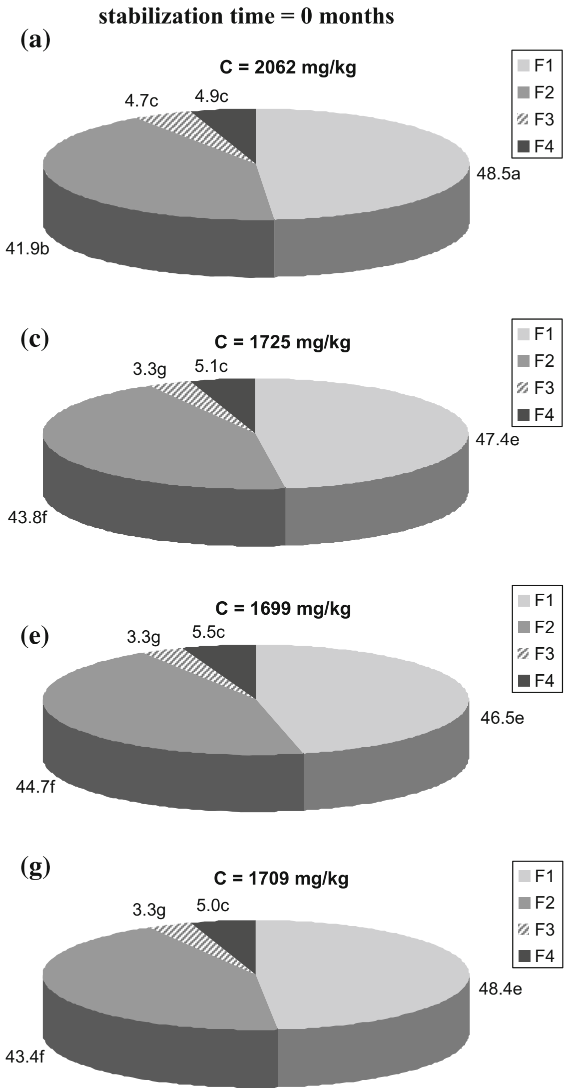

Fig. 2 Percentage content of Zn in individual fractions (*F*1–*F*4) in non-amended and compost-amended soils at the beginning and the end of stabilization: **a**, **b** non-amended soil (control, series 1); **c**, **d** soil with compost of 3-month maturation (series 2); **e**, **f** soil with compost of 6-month maturation (series 3); **g**, **h** soil with compost of 12-month maturation (series 4). Mean values ($n = 3$) between the same fractions at $t = 0$ and 12 months in individual series followed by the same letters do not differ significantly at $p < 0.05$

Copper

In non-amended (control) soil, Cu prevailed in reducible (*F*2) fraction (52 %, 520.3 mg/kg), followed by exchangeable and acid-soluble (*F*1) fraction (19 %, 189.4 mg/kg). The contents of Cu in oxidizable (*F*3) and residual (*F*4) fractions were lower, but comparable (on average 14 %, 141.3 mg/kg) (Fig. 1a). This distribution pattern is in agreement with results obtained by Lu et al. (2009). Although the authors used loamy soil with higher content of silt fraction and Cu concentration was two times lower than in the present study, the metal prevailed in *F*1 and *F*2 fractions, 54 and 26 %. In addition, Cu hardly entered the residual fraction in artificially contaminated soil during 56 days incubation. Similarly, Hong et al. (2002) found that Cu at concentrations 1521–2181 mg/kg in clay loam, loam and sandy clay loam contaminated by more than 2 years was predominantly found in Fe–Mn oxide fraction followed by carbonate fraction. In the present study, Cu content in oxidizable (*F*3) fraction was markedly higher than Zn. This confirms higher Cu affinity for organic matter and that Cu easily forms highly stable complexes with organic matter (Li et al. 2001).

After compost addition ($t = 0$ months), Cu distribution in soil changed compared to soil without amendments. In all treatments with composts, Cu content in *F*1 and *F*2 fractions decreased, while it increased in *F*3 and *F*4 fractions (Fig. 1c, e, g). The Cu concentration in oxidizable fraction (calculated in relation to % content of this fraction in non-amended soil at $t = 0$ months) increased by 70 % (69 mg/kg) in series 2, by 72 % (76 mg/kg) in series 3 and by 68 % (62 mg/kg) in series 4. The Cu content increased in the residual fraction by 6–18 % (25–52 mg/kg). Although total Cu concentration in composts was lower than in soil, the distribution pattern was different than in contaminated soil (on average): 1.0 % ($F1$ fraction), 1.5 % ($F2$ fraction), 79.3 % ($F3$ fraction) and 18.2 % ($F4$ fraction). This can be attributed to a Cu increase in the most stable fractions in amended soil already at the beginning of the stabilization process.

In soil without organic amendment, long-term stabilization ($t = 12$ months) did not cause considerable changes in Cu fractionation (Fig. 1b). There was only a slight drop in the concentration of $F1$ fraction and an increase in the concentration of the remaining fractions ($p > 0.05$). In contrast, long-term metal stabilization in soil with composts had a positive effect on Cu transformations (Fig. 1d, f, h). Regardless of compost maturation time, Cu was redistributed mainly from exchangeable and acid-soluble fraction ($F1$) into oxidizable form ($F3$). In series 2, 3 and 4, the content of $F1$ diminished by 43 % (calculated in relation to % content of this fraction in amended soil at $t = 0$ months), whereas in oxidizable fraction it increased by 19, 24 and 21 %, respectively (Fig. 1). The addition of organic matter to soil in the form of compost increases Cu sorption due to the high affinity of this metal for organic compounds (Bolan et al. 2003). However, the changes in Cu distribution among composts with a different maturation time were not high, which could be connected with slight differences between the content of organic matter and humic substances in all composts. In compost from sewage sludge, the formation of humic substances occurs most intensively during the first 3–4 months of maturation, then the rate of humification decreases. However, during long maturation time polymerization of FA to HA takes place, which was also shown in the present study (the increase of DP index from 0.83 to 1.13 with maturation time from 3 to 12 months, respectively). However, unexpectedly, it did not affect the Cu content in the oxidizable ($F3$) fraction ($p > 0.05$). This indicates that prolonging the compost maturation time from 3 to 12 months does not affect Cu distribution in soil. Moreover, due to the fact that in amended soil, proportion between soil and composts was high (80 %:20 % w/w, respectively), the differences between content of organic matter and humic substances in amended soil were small.

Available literature data do not define a clear relationship between the degree of humification of organic matter and metal distribution. Pérez-Esteban et al. (2012) showed that Cu immobilization in soil depended on compost type. The manure compost reduced more Cu and Zn bioavailability than pine bark compost. The authors emphasized that, in that case, metal redistribution was associated not only with the degree of organic matter humification, but also with pH and organic matter content in both composts.

In the present study, the differences in Cu redistribution for three composts were less visible because they were made from the same substrates, had comparable pH, organic matter content and, despite differences in DP index, their addition to soil caused no statistically significant ($p > 0.05$) differences in Cu distribution for composts with different maturation times (3, 6 and 12 months).

Zinc

In the present study, almost 90 % of total Zn in non-amended soil was associated with exchangeable and acid-soluble ($F1$) and reducible ($F2$) fractions. The content of oxidizable and residual forms was about 10 % (Fig. 2a). Therefore, Zn may offer higher environmental risk compared to Cu since more mobile metals are immediately bioavailable. In general, in artificially contaminated soils, since metals enter less into the crystal lattice of layer silicates, they are therefore relatively mobile compared to unpolluted soils. Al Chami et al. (2013) found that a considerably higher proportion of Zn added artificially to soil occurred in the acid-soluble fraction and in the reducible fraction (39.1 and 30.5 % of total Zn, respectively). Only 5 % of total Zn occurred in an oxidizable form. According to the authors, this was due to the low content of organic matter in soil (around 2 %) or non-selectivity of acetic acid used for extraction of the first fraction in the BCR procedure. This reagent is able to release some metal from all soil fractions, even those which are organically bonded. Although, in the present study, the content of organic matter in non-amended soil (10.1 %) was five times higher than in soil used by Al Chami et al. (2013), Zn also prevailed in $F1$ and $F2$ fractions. In contrast to Cu, at the beginning of stabilization process the distribution pattern of Zn in amended soil was similar to that in non-amended soil (Fig. 2c, e, g), despite 2 times higher content of organic matter in amended than in non-amended soils. This means that in the case of Zn, organic matter does not affect its redistribution in soil. Analogous results, but for other types of soil and other organic materials, were evidenced in literature. Al Chami et al. (2013) found that compost from green waste and manure added to silt clay loam did not affect Zn distribution. Pérez-Esteban et al. (2012) showed that the application of organic materials to soil did not increase the amount of Zn associated with the organic fraction. Similarly, Narwal and Singh (1998) reported that organic matter provided by cow and pig manure amendments did not influence the Zn fractionation in soil.

After 12 months of stabilization in series with composts, the reducible fraction ($F2$) prevailed over exchangeable and acid-soluble form ($F1$) (Fig. 2d, f, h). In non-amended soil, the proportions between these two fractions were comparable (Fig. 2b). Changes in oxidizable and residual fractions were rather negligible ($p > 0.05$) compared to the beginning of the process. During stabilization, in all series the highest increase of Zn was in the reducible fraction (calculated in relation to the % content of this fraction at $t = 0$ months), by 6 % (35 mg/kg) in non-amended soil; by 7 % (68 mg/kg) in series 2, by 6 % (43 mg/kg) in series 3 and by 8 % (57 mg/kg) in series 4.

In the case of Zn, which reacts weakly with organic matter (Stevenson 1994), the main mechanism responsible for its redistribution might be associated with pH changes in soil, especially pH increase. The increase in the soil pH may be attributed to the mineralization of organic carbon and subsequent production of OH^- ions by ligand exchange (Mkhabela and Warman 2005). However, in our study, the efficiency of organic matter (OM) mineralization was rather low and amounted to 1.5 % (series 1), 10.0 % (series 2), 8.5 % (series 3) and 6.5 % (series 4). This is probably because a high metal concentration affects soil microorganisms or their activity and, in turn, the degradation of organic matter (Clemente and Bernal 2006). Therefore, it may be assumed, that in this study other factors were responsible for pH increase. Firstly, the maintenance of soil moisture around 60 % of their field capacity over stabilization time by periodical adding of distilled water at neutral pH might affect pH increase. Secondly, as compost is a source of organic nitrogen, transformations of this form of N (ammonification) could influence soil pH. Ammonification of organic nitrogen and microbial decarboxylation due to proton consumption led to pH increase in soil (Yan et al. 1996). In contrast, nitrification releases protons and decreases soil pH (Devries and Breeuwsma 1987). In the present study, the highest pH increase (from 6.1 to 6.7, on average) in compost-amended soil was observed during the first 3 months of stabilization then it remained unchanged up to the end of stabilization (data not shown). This might indicate that in metal-contaminated soil, ammonification of nitrogen from compost proceeded relatively low, what could be related to inhibitory effect of heavy metals on ammonifying bacteria in soil (Baath 1989). In contrast, Huang and Chen (2009) found that in uncontaminated agriculture soils amended with sewage sludge compost, soil pH increased relatively fast and reached its maximum after 21 days of incubation, then it started to slowly decrease after 42 days of incubation due to nitrification process.

The increase of soil pH favors metal sorption on the oxides. In general, amorphous Fe and Mn oxides have a great affinity for Zn. This could explain the Zn increase in the reducible fraction during stabilization. Our findings are in agreement with the results obtained by other authors using compost made from completely different waste products than in our study. Pérez-Esteban et al. (2012) found that Zn distribution in soil amended with manure and pine bark composts was more affected by pH-dependent sorption processes than by complexing with organic amendments. Wong et al. (2007) demonstrated that soluble organic carbon did not affect Zn concentrations as for Cu because of the lower affinity of Zn for organic compounds and pH had a greater influence on Zn sorption. At higher pH in soil amended with spent mushroom compost, Zn was redistributed from the exchangeable fraction to the Fe–Mn oxide fraction in coarse-textured soil (Shuman 1999). The above results obtained for different soils indicate that the addition of organic amendments had little effect on Zn redistribution in oxidizable fraction in soil, whereas the opposite trend was for reducible fraction.

Changes in Cu and Zn mobility and stability during stabilization with sewage sludge composts

Mobility factor (M_F)

In the present study, changes in Cu and Zn mobility were analyzed based on mobility factor (M_F) affected by the content of $F1$ fraction. Metals in this fraction represent the highest potential risk to the environment.

The changes in Cu and Zn mobility versus stabilization time are shown in Figs. 3 and 4, respectively. In general, the mobility of Zn was significantly higher than Cu, both in non-amended and amended soils ($p < 0.05$).

As illustrated in Fig. 3, at the beginning of stabilization, Cu in non-amended soil showed medium mobility ($M_F = 19.1$ %). In soil amended with composts, the mobility was lower ($M_F = 11.4$ %, on average), but was still classified as a medium. A decrease in Cu mobility by about 8 % in the amended soil at the beginning of stabilization (series 2–4) was affected by a different, initial distribution pattern of Cu, both in soil and composts. In contaminated soil, 19 % of the total Cu concentration was in the $F1$ fraction, whereas in the composts it averaged 2.2 % (full data not shown). Therefore, mixing soil with composts caused changes in the proportions between individual fractions, especially in the $F1$ form, which affects metal mobility. Although Branzini and Zubillaga (2012) used biosolid compost at dosage six times lower than in the present study for metal stabilization in spiked sandy loam, they found that already after amendment there were some changes in metal distribution. For example, Cu content in soluble and exchangeable fraction increased by 45 %, in carbonate fraction by 0.7 %, in oxidizable fraction by 7.5 % and in residual fraction by 12 %.

In contrast, no significant decreases in M_F values for Zn were noted in soil without compost and in soil after mixing with amendments ($p > 0.05$). The average Zn mobility in

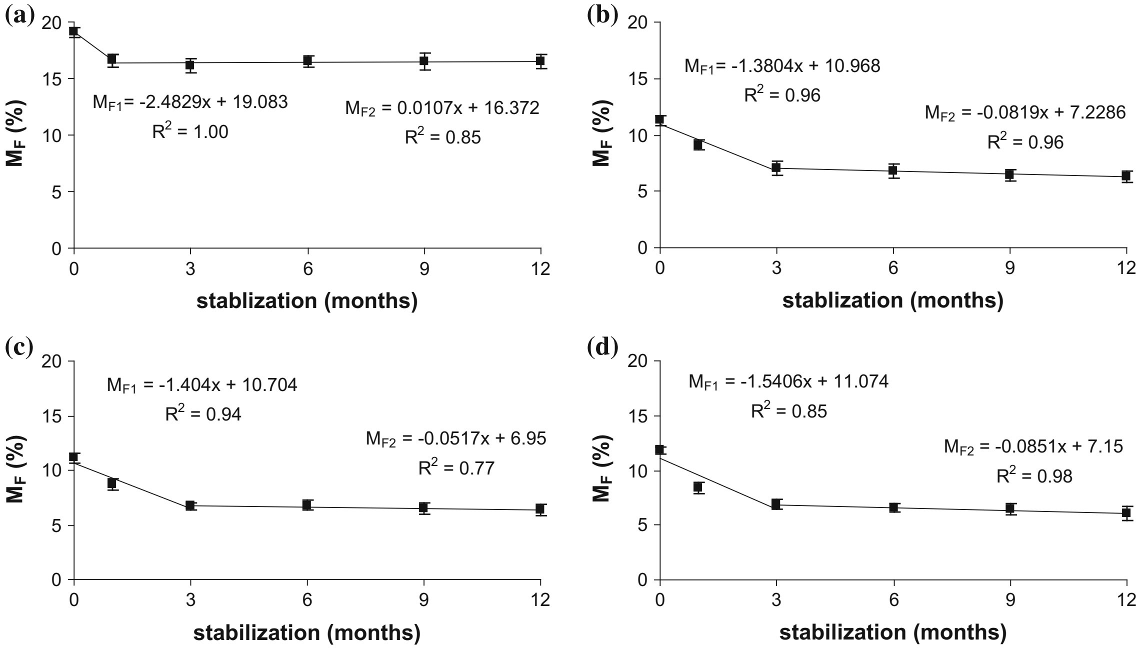

Fig. 3 Changes of mobility factor (M_F) for Cu as a function of stabilization time: **a** non-amended soil (control, series 1), **b** soil with compost of 3-month maturation (series 2), **c** soil with compost of 6-month maturation (series 3), **d** soil with compost of 12-month maturation (series 4) (bars refer to ±SD, $n = 3$)

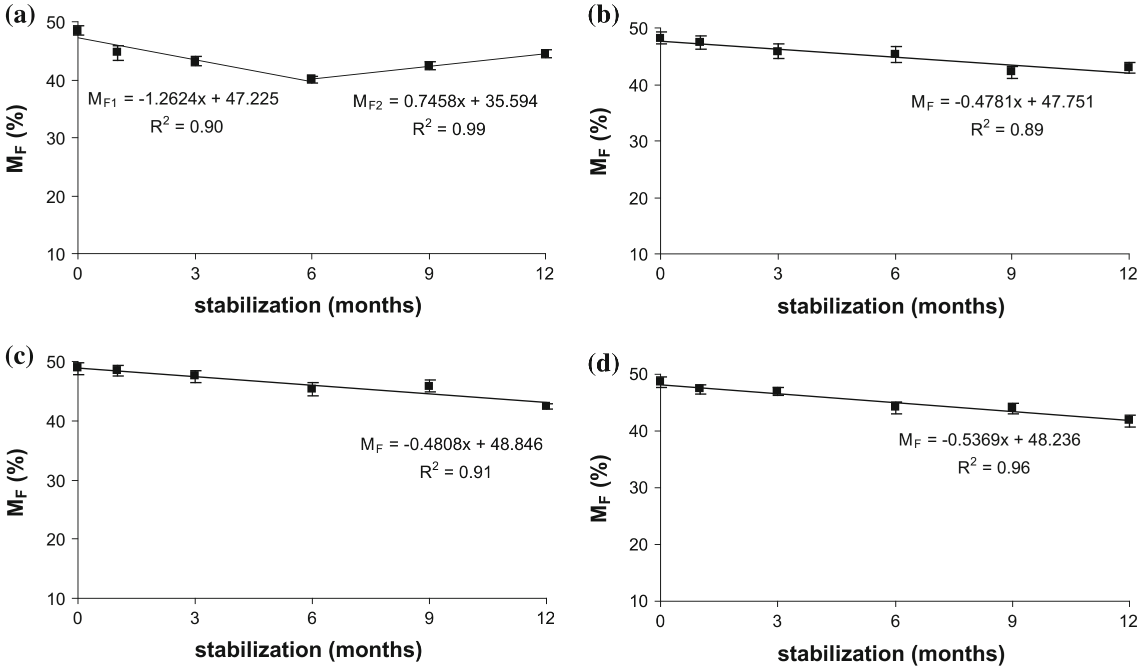

Fig. 4 Changes of mobility factor (M_F) for Zn as a function of stabilization time: **a** non-amended soil (control, series 1), **b** soil with compost of 3-month maturation (series 2), **c** soil with compost of 6-month maturation (series 3), **d** soil with compost of 12-month maturation (series 4) (bars refer to ±SD, $n = 3$)

all series at the beginning of the experiment was high ($M_F = 48.5$ %) (Fig. 4). The reason for this was because the proportions between individual fractions in composts were similar to those in non-amended soil, although soil and composts contained different total Zn concentrations. In composts, the average content of Zn in individual fractions was: 38 % (*F*1 fraction), 55.2 % (*F*2 fraction), 4.5 % (*F*3 fraction) and 2.3 % (*F*4 fraction).

During stabilization, the changes in Cu and Zn mobility proceeded in a different way. For Cu, its mobility changed in two stages, both in non-amended and amended soil. Initially, Cu mobility visibly decreased and almost remained unchanged when the stabilization time was prolonged (Fig. 3). Similar trends of decreasing the content of exchangeable fraction were observed by Lu et al. (2005) during a 56-day incubation of soil artificially contaminated with Cu at a concentration of 500 mg/kg. The changes proceeded for the first 42 days of experiment and after that remained unchanged. In our study, in all series transformations of $F1$ fraction for Cu concerned a relatively short time. The steady-state conditions in control soil were obtained by the end of the first month of stabilization, whereas in amended soils this occurred after the third month.

Based on changes of the M_F in time, rate constants of Cu transformations in the $F1$ fraction referring to metal mobility (Eq. 7) were determined. Although in all series Cu mobility changed in two phases (M_{F1} and M_{F2}), transformations at the first phase occurred at considerably higher rate compared to the second phase. This means that Cu redistribution from $F1$ fraction into more stable fraction, i.e., oxidizable (data not shown) proceeded for 3 months. Above this time Cu showed low environmental risk (based on M_F) in amended soil. In addition, despite the fact that the $F1$ transformation rate was higher in non-amended soil ($M_{F1} = 2.48$ %/month) than in amended soil ($M_{F1} = 1.44$ %/month, on average), it is worth noting that in non-amended soil the changes occurred mainly for the first month of stabilization, while in amended soils this occurred for 3 months.

In contrast, Zn mobility in the amended soils decreased continuously during the 12-month stabilization period and at the end of the process it was at the level of 42 % (on average), indicating still high metal mobility and environmental risk ($30\ \% < M_F \leq 50\ \%$). In all treatments with compost, the transformation rates of Zn mobility were comparable (0.49 %/month, on average) (Fig. 4b–d). In non-amended soil, Zn mobility initially decreased at a transformation rate of 1.41 %/month, but after the sixth month it started to slightly increase from 40 to 44 % at a transformation rate of 0.75 %/month (Fig. 4a).

The phenomenon of increasing metal mobility over time is observed in both contaminated and native soils without amendments. Fengxiang and Banin (1997) during a year of incubation of sandy and loessial soils containing 25.2 and 38.5 mg Zn/kg, respectively, observed that the metal generally tended to be transferred to the carbonate fraction, responsible for metal mobility.

On the whole, changes in Zn mobility proceeded on average threefold slower than Cu (Figs. 3, 4). These findings may be explained by the fact that Zn concentration in the $F1$ fraction was above 8 times higher than Cu. The high contents of exchangeable and acid-soluble fractions could cause slower Zn transformations into more stable fractions, despite long-term incubation. At such a high proportion of exchangeable and acid-soluble fraction ($F1$) for Zn, the pH increase in amended soil during stabilization turned out to be insufficient for effective metal redistribution, although it is known that high pH favors Zn sorption, mainly on Fe and Mn oxides. In addition, simultaneous adding more than one metal to soil could affect their transformations because of competition for sorption sites (Zhang et al. 2011). In the present study, the soil contained mixture of Cu and Zn.

The reduced partition index (I_R)

The reduced partition index (I_R) was introduced to quantitatively describe the relative binding intensity of metals in soils. Therefore, it enables comparison of the binding intensities of a given metal in different soils, or of different metals in the same soil (Han et al. 2003; Miretzky et al. 2011). In contrast to the M_F, the I_R includes metal transformations not only among labile fractions, but also stable ones. Therefore, this parameter seems to be suitable to assess metal redistribution and efficiency of soil amendments in metal immobilization.

For BCR sequential extraction, the I_R values can range between 0.06 and 1; high value indicates metal stability in soil resulting from its occurrence in oxidizable ($F3$) and residual ($F4$) fractions, low values represent a distribution pattern with a high proportion of exchangeable and acid-soluble fraction ($F1$). The intermediate value of I_R refers to the pattern in which a metal is distributed among all fractions. From a stabilization point of view, the higher I_R value in amended soil, the lower the environmental risk for a given metal.

The data given in Fig. 5 show that the I_R values for Cu and Zn changed over stabilization time as well as the redistribution rate constants for $F1$–$F4$ metal fractions, estimated from Eq. (8). In general, the intensity of metal bonding was higher for Cu than for Zn already at the beginning of stabilization in non-amended soil (Fig. 5a, b). Compost addition resulted in an increase in metal stability only for Cu. This was attributed to increase in Cu content in oxidizable and residual fractions. It is known that addition of organic matter to soil in the form of compost increases Cu sorption due to the high affinity of this metal for organic compounds, in particular humic substances have a large effect on its retention (Bolan et al. 2003). At the beginning of stabilization ($t = 0$ months), the I_R increased from 0.37 (series 1) to 0.46 (series 2–4). For comparison, the value of I_R for Zn was 0.21 in all series. The increase in I_R value, as for M_F, was affected by a different, initial distribution pattern of Cu in soil and composts.

The increase of incubation time led to changes in I_R values for individual metals due to their further redistribution. In non-amended soils, there were no significant

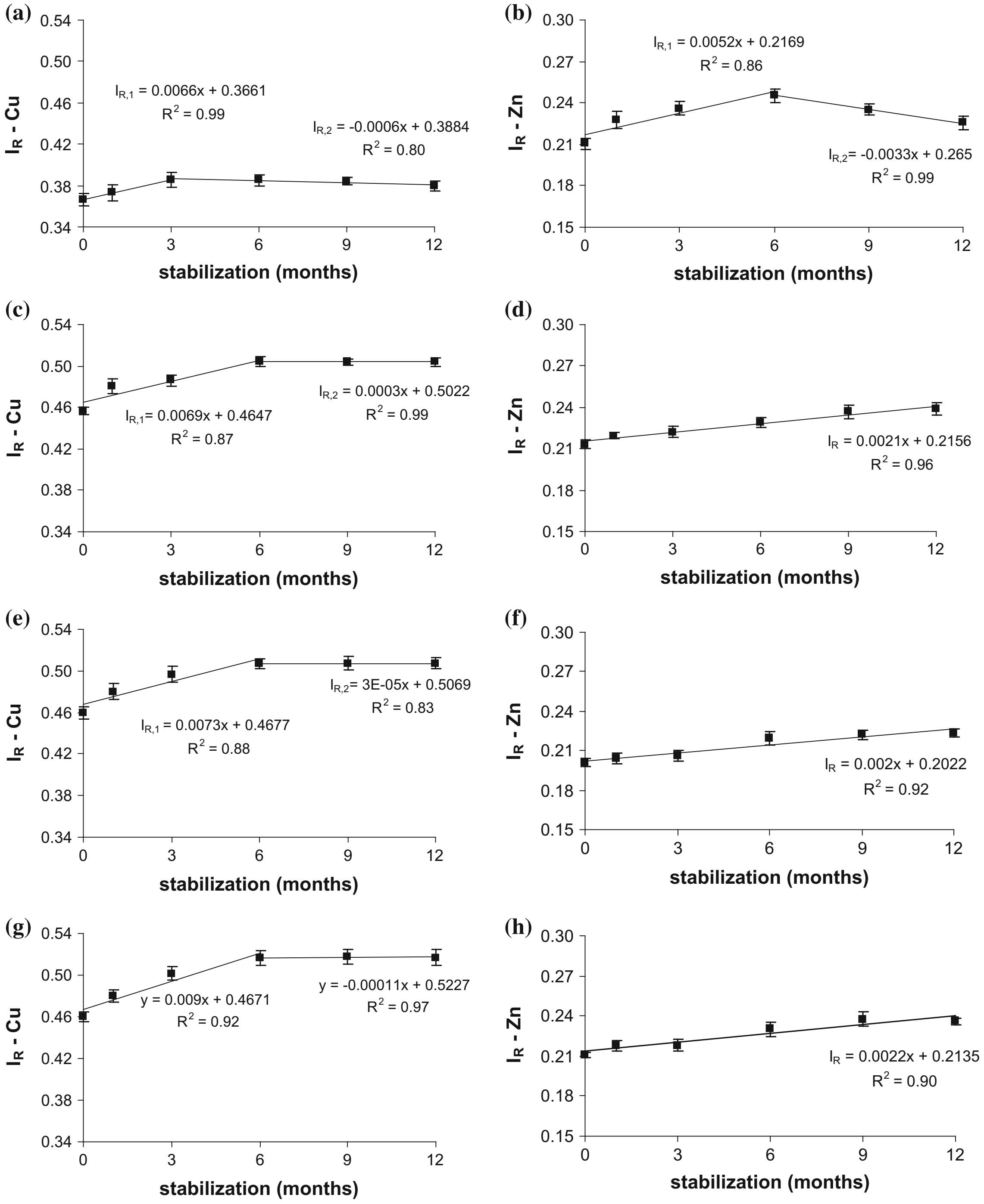

Fig. 5 Changes of reduced partition index (I_R) for Cu and Zn during stabilization process: **a**, **b** non-amended soil (control, series 1); **c**, **d** soil with compost of 3-month maturation (series 2); **e**, **f** soil with compost of 6-month maturation (series 3); **g**, **h** soil with compost of 12-month maturation (series 4) (bars refer to ±SD, $n = 3$)

differences in Cu stability (I_R with $p > 0.05$), because distribution patterns of Cu at the beginning and the end of stabilization were similar. In contrast, in compost-amended soils the I_R values increased to 0.51 after 6 months of stabilization and then remained almost constant. Zn stability in soil amended with composts increased continuously for 12 months, but just in a narrow range ($0.21 < I_R > 0.24$).

The rate constants of metal redistribution in *F*1–*F*4 fractions in amended soil ranged from 0.007 to 0.009/month for Cu (Fig. 5c, e, g) and 0.002/month for Zn (Fig. 5d, f, h). For both metals, the rate constants were an order of magnitude lower than rate constants of metal transformations in $F1$ fraction. This means that the metal redistribution preceded very slowly, which was also indicated by other authors (Han et al. 2003; Lu et al. 2005). However, to date there has been a lack of estimated rate constants of metal mobility and stability based on M_F and I_R indices in time.

In general, metal redistribution includes different reactions such as: complexation, adsorption, precipitation or diffusion into the mesopores and micropores of soil (Jalali and Khanlari 2008). In soils with recently added metal, their sorption is rapid during the first few hours or days, but then slows with time. At low concentration, metals are more redistributed among soil components than at high concentration (Han et al. 2003). In the present study, total Cu concentration was twofold lower than Zn, which might affect higher Cu stability in soil during stabilization.

Conclusions

The maturation time of sewage sludge compost (3, 6 and 12 months) and its related humification degree did not affect the Cu or Zn redistribution patterns. The application of compost allowed a decrease in Cu mobility in soil from "medium" to "low", in contrast to Zn, which in all treatments remained highly mobile. Cu stability in the amended soil was twofold higher than for Zn. The rate constants of Cu and Zn redistribution were an order of magnitude lower than the rate constants of metal transformations in the exchangeable and acid-soluble fractions. Metal transformations in the amended soil proceeded longer than in the non-amended soil.

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