Mobilities and leachabilities of heavy metals in sludge with humus soil

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Abstract

Chemical forms of Zn, Ni, Cu, and Pb in municipal sewage sludge were investigated by adding humus soil to sludge and by performing sequential extraction procedures. In the final sludge mixtures, Zn and Ni were mainly found in Fe/Mn oxide-bound (F3) and organic matter/sulfide-bound (F4) forms. For Zn, exchangeable (F1), carbonate-bound (F2), and F3 forms were transformed to F4 and residual forms (F5). For Ni, F1 and F2 forms were transformed to F1, F2, and F3 forms. Both Cu and Pb were strongly associated with the stable forms F4 and F5. For Cu, F2 and F3 forms were major contributors, while for Pb, F3 and F4 forms were major contributors to F5. Humus soil dosage and pH conditions in the sludge were strongly correlated with the forms of heavy metals. Five forms were used to evaluate metal mobilities in the initial and final sludge mixtures. The mobilities of the four heavy metals studied decreased after 28 days. The metal mobilities in the final sludge mixtures were ranked in the following order: Ni > Zn > Cu = Pb. Leaching tests showed that the mobilities of Zn and Ni in lower pH conditions (pH 4) were higher than those in higher pH conditions (pH 8).

Key words: sludge; heavy metal; humus soil; mobility; leachability

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Introduction

Global sewage sludge production is increasing rapidly every year. In China, five million metric tons of dry sewage sludge is produced annually (Tang and Zhao, 2005). Sewage sludge contains a large amount of organic matter and substantial N and P concentrations; hence, it is used preferentially as a fertilizer in agriculture and as a regenerator of soil (Hemández et al., 1991; Granato and Page, 1992). Organic pollutants and heavy metals limit agricultural application of sewage sludge, however, as they tend to accumulate along the food chain and bring potential risks to animals and humans (Chang and Smith, 1996). Therefore, reducing heavy metals in sewage sludge is a major concern for its application on land. Recent studies have found that heavy metal bioavailability in the environment depends not only on their total concentration but also on their speciation (Legret, 1993; Zufiaurre et al., 1998; Su and Wong, 2003; Fuentes et al., 2004). Therefore, heavy metals were divided into five forms: exchangeable, acid extractable (carbonate bound), reducible (Fe-Mn oxides bound), oxidizable (organic bound), and residual (Tessier et al., 1979). Different forms of heavy metals have different mobilities (Shrivastava and Banerjee, 1998). The exchangeable and acid-extractable forms are mobile fractions and easily available. The oxidizable and reducible

forms are leached out only under extreme conditions, while the residual form is almost inert.

Application of different additives to sewage sludge to induce various processes, such as adsorption to mineral surfaces, surface precipitation, and ion exchange, is one of the most common ways to transform heavy metal forms and immobilize heavy metals (Su and Wong, 2003; Wong and Selvam, 2006; Wang et al., 2008; Zorpas et al., 2008). The possibilities for environmentally sound co-application strategies, such as organic matter amendment, may be explored to minimize the potential negative impact of sewage sludge application on land (Singh and Agrawal, 2008).

Natural humus soil is formed from decaying leaf litter that contains humic substances (Fig. 1) such as organic macromolecule humic acids, which possess multiple properties and high structural complexity (Chaturvedi et al., 2006). Recent research has reported that humus soil or humus soil with hydroxyapatite are appropriate amendment materials for decreasing mobility of heavy metals (Misra and Pandey, 2005; Chaturvedi et al., 2006). To our knowledge, however, most heavy metal studies have concentrated on sorption, desorption, and leaching ability, with little focus on the distribution of heavy metals forms in humus-soil-amended sludge. In China, sewage sludge contains relatively high concentrations of Zn, Ni, Cu, and Pb compared to other heavy metals (Yin and Tan, 2004; Wang et al., 2008). We determined the forms of Zn, Ni,
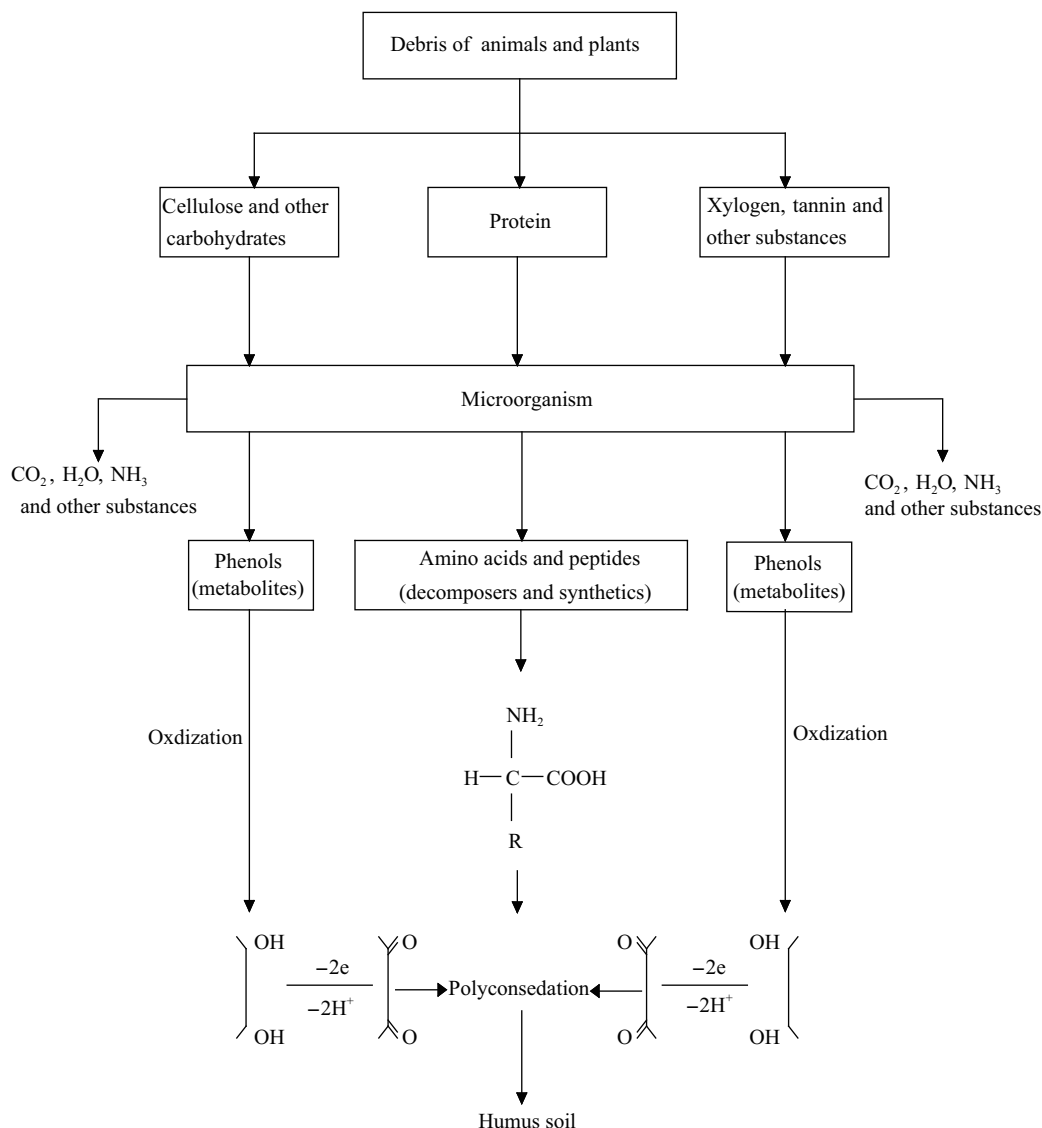
Fig. 1   Formation of natural humus soil.

Cu, and Pb in humus-soil-amended sewage sludge to better understand the mobility of heavy metals. To understand heavy metal reduction in humus-soil-amended sludge, sequential extraction procedures were used to identify changes associated with heavy metal forms. In addition, heavy metal leachability in humus-soil-amended sludge was investigated at different pH values (pH 4 and pH 8).

1 Materials and methods

1.1 Sewage sludge and humus soil

Sewage sludge (SS) was collected from the secondary sedimentation tank of Quyang Municipal Wastewater Treatment Plant in Shanghai, China. Humus soil (HS) was obtained from the WTM Company of Japan. Through physico-chemical processes, natural humus soil and silicate were converted into HS (Fig. 1). Fourier transform infra-red spectroscopy (FT-IR) of HS is shown in Fig. 2. According to Abouelwafa et al. (2005), each peak represents a functional group (Table 1).

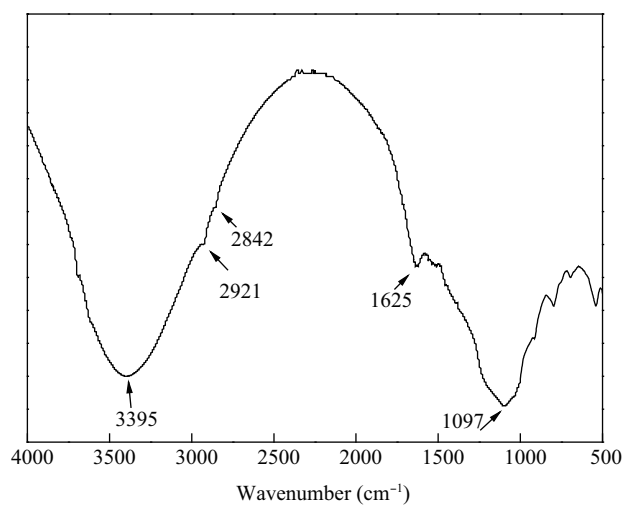Moisture content of the SS and HS was determined

Fig. 2   FT-IR spectra of humus soil.

by oven-drying at 105°C for 24 hr. Carbon, hydrogen, and nitrogen content was determined by an automated elemental analyzer (Vario EL spectrometer, Elementar Co., Germany). Chemical and physical properties of SS and HS

Table 1 Functional groups of humus soil

Wavenumber (cm^{-1})	Functional group
3400	H–OH (phenols, alcohols and carboxylic groups), N–H (amides and amines)
2920	C–H stretching vibration of aliphatic structures (fatty acids, waxes and various other aliphatics)
2840	Asymmetrical stretching vibration of C–H in –CH$_2$ and –CH$_3$ (fatty acids and alkanes)
1620–1660	Stretching vibration of COO and C=O in amides I, stretching vibration of C=O in ketones, carbolylicacids and/or quinones, stretching vibration of aromatic C=C
1420–1460	Stretching vibration of C–H–, CH$_2$– and –CH$_3$ radicals in aliphatic structures
1170–1080	C–O–C vibration of carbohydrates and vibration of aromatic ethers, Si–O vibration

Table 2 Properties of sewage sludge (SS) and humus soil (HS)

Item	SS	HS
pH	7.2[a]	5.1
Moisture (%)	99.8	10
Total C (g/kg)	333.2	148.6
H (g/kg)	51.2	29.5
Total N (g/kg)	54.1	9.4

[a] Values are means ($n = 3$).

are listed in Table 2.

The SS was mixed with HS at 0%, 7%, 14%, 28%, 41%, and 68% (W/W, dry weight) and shaken at 100 r/min for 30 days in polyethylene flasks at (27 ± 1)°C. The mixture was sampled at the end of the 30 day period. Samples were air-dried, thoroughly mixed, ground to pass through a 1-mm sieve, and stored in polyethylene flasks at room temperature for further analysis.

1.2 Sequential extraction

The conventional method of Tessier et al. (1979) was used with modification for sequential extraction. The extraction was carried out in 50 mL polypropylene centrifuge tubes with an initial mass of 0.5 g samples. The five heavy metal forms were divided in the following steps.

Step 1: exchangeable form (F1): the solid sample (0.5 g) was extracted by 1 mol/L MgCl$_2$ at initial pH 7 (10 mL), and was shaken at 250 r/min for 1 hr at room temperature; Step 2: carbonate-bound form (F2): the residue from step 1 was extracted by 1 mol/L CH$_3$COONa (15 mL), buffered to pH 5 with CH$_3$COOH, and shaken at 250 r/min for 6 hr at room temperature; Step 3: Fe/Mn oxide-bound form (F3): the residue from step 2 was extracted by 0.04 mol/L NH$_2$OH·HCl (15 mL) at initial pH 2 with CH$_3$COOH (25%, V/V), and was shaken at 250 r/min for 5 hr at 96°C; Step 4: organic matter/sulfide-bound form (F4): the residue from step 3 was extracted by 0.02 mol/L HNO$_3$ (5 mL) and H$_2$O$_2$ (5 mL, 30%, V/V) at initial pH 2, and was shaken at 250 r/min for 3 hr at 85°C, then 5 mL of 30% H$_2$O$_2$ was added into the same tube with continuous shaking for 2 hr, finally, 5 mL of 3.2 mol/L CH$_3$COONH$_4$ in HNO$_3$ (20%, V/V) was added into the tube, and it was then shaken at 250 r/min for 0.5 hr at room temperature; Step 5: residual form (F5): the residue from step 4 was digested with HNO$_3$, HF, and HCl according to the microwave digestion method

(Method 3502, US EPA, 1996).

After each successive extraction, separation was accomplished by centrifugation at 4000 r/min for 20 min, while the residue was washed with 10 mL of deionized water and then centrifuged at 4000 r/min for 20 min. The supernatant from the washing step was mixed with the supernatant obtained initially, filtered through a 0.45-μm membrane filter, and analyzed for the heavy metals using ICP-OES (Optima 2100 DV, PerkinElmer, USA).

1.3 Leaching test

To assess the leachabilities of the heavy metals in the final mixtures, 1 g of sample was mixed with 20 mL of deionized water adjusted to pH 4 and pH 8 with H$_2$SO$_4$ or NaOH solutions and shaken for 4 hr (An et al., 2003). The mixtures were centrifuged at 4000 r/min for 20 min, and the supernatant was then filtered through a 0.45-μm membrane filter and analyzed for the heavy metals of interest.

1.4 Calculations and statistical analysis

All results were presented as the average of three replicates. Correlation (Pearson correlation) analysis was performed by Statistical Program for Social Sciences 13.0 for Windows.

2 Results and discussion

2.1 pH and total heavy metals in mixtures

The pH level and heavy metal concentrations of the initial mixtures are given in Table 3. The addition of HS significantly decreased pH level initially from 7.2 to 6.2. The tests also clearly show that with an increasing amount of HS, the concentrations of all heavy metals in the samples decreased. The most abundant heavy metal in the initial mixture was Zn, followed by Cu, Pb, and Ni. The total contents of heavy metals in the initial mixture were lower than the corresponding limits required for municipal wastewater treatment plant sludge (GB18918-2002) (Table 4). However, Zn exceeded the control standards of pollutants in sludge for agricultural use (GB4284-84) (Table 4). Similar ranges of heavy metal concentrations have been reported in previous studies (Wang et al., 2005; Alonso et al., 2006, 2009; Wang et al., 2008). After 28 days, pH decreased with different HS dosages of 0%, 7%, 14%, 28%, 41%, and 68% as follows: 6.9, 6.4,

Table 3 pH and heavy metal concentrations of the initial mixtures

Treatment	pH	Zn (mg/kg)	Ni (mg/kg)	Cu (mg/kg)	Pb (mg/kg)
SS + 0% HS[a]	7.2	1803	31	228	58
SS + 7% HS	7.0	1717	28	223	58
SS + 14% HS	6.8	1582	24	199	48
SS + 28% HS	6.5	1447	24	176	46
SS + 41% HS	6.4	1304	23	169	40
SS + 68% HS	6.2	1182	20	137	34

[a] HS amendment rates for sewage sludge were 0%, 7%, 14%, 28%, 41% and 68% (W/W).

* Values are means ($n = 3$).

Table 4 Chinese legal limits for heavy metals

Standard	pH	Zn (mg/kg)	Ni (mg/kg)	Cu (mg/kg)	Pb (mg/kg)
GB18918-2002[a]	< 6.5	2000	100	800	300
	⩾ 6.5	3000	200	1500	1000
GB4284-84[b]	< 6.5	500	100	250	300
	⩾ 6.5	1000	200	500	1000

[a] Discharge standard of pollutants for municipal wastewater treatment plant.
[b] Control standards for pollutants in sludge for agricultural use.

6.2, 6.0, 6.0, and 5.8, respectively. The pH changes may relate to acid released from HS. Due to evaporation and decomposition, the mixed liquid suspended solids did not change obviously; hence, the total content of Zn, Ni, Cu, and Pb also did not change in the final mixtures.

2.2 Chemical speciation of heavy metal in sludge

2.2.1 Initial sludge

Different chemical forms of heavy metals represented as sum concentrations (%) in the initial sludge are shown in Fig. 3. Results show that the dominant speciation of Zn in the sludge was in F1 (15.6%) and F3 (46%) forms. Similar to Zn, a high percentage of Ni was found in the F1 (30%) and F3 (30%) forms. Most Pb existed in the F5 form (66.2%), with F1 and F2 forms not detected. The most part of Cu appeared in the F4 form (70%) and not in the F1 form. Most Zn and Ni were associated with mobile F1–F3 forms (> 70%), and most Cu and Pb were distributed in the stable F4 and F5 forms (> 80%). In the initial mixture, the addition of HS did not affect the percentage of each form, due to HS containing significant amounts of SiO_2 and very low amounts of heavy metals.

2.2.2 Changes of metal distribution

Table 5 shows the results obtained after sequential extraction. For Zn, adding HS to sludge in F4 form increased Zn content from 284 to 528 mg/kg after 28 days, and decreased in other forms. Table 5 also presents the changes in the percentages of each Zn form; in particular, the percentage of Zn in F4 form increased from 16.2% to 44.7%. Conversely, the percentage of Zn in F1, F2, and
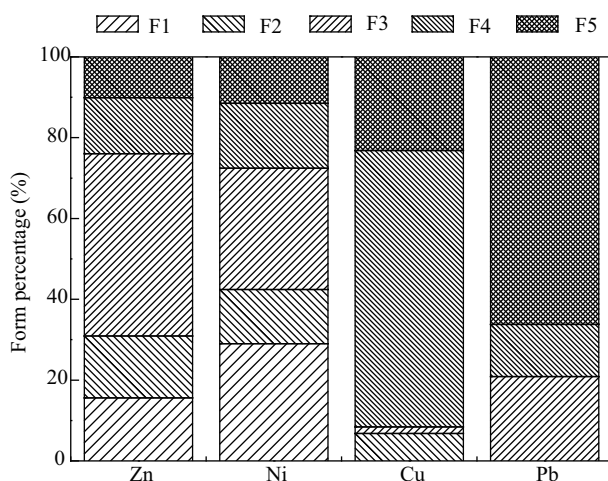

Fig. 3 Percentage of each form of Zn, Ni, Cu, Pb in the initial sludge.

F3 forms decreased substantially. The percentage of Zn in F5 form was similar for all samples at 11%. Changes in the relative percentages of each form of Zn are given in Fig. 4. With the addition of HS, the relative percentage of Zn in F1, F2, and F3 forms decreased gradually, while the relative percentage of Zn in F4 form increased. These results indicate that Zn of F1, F2, and F3 forms were transformed into F4 form.

As shown in Table 5, Ni content of F1, F2, F3 and F4 forms decreased by HS addition, but F5 form was always 4 mg/kg. The percentage of Ni in F1 and F2 forms decreased from 26.4% to 16.3% and from 10.7% to 7.3%, respectively. The percentage in F3, F4, and F5 forms increased from 32.3% to 34.7%, from 17.3% to 21.7%, and from 13.3% to 20.0%, respectively. The relative percentage of Ni of F1 and F2 forms decreased, from –2.6% to –12.7% and from –2.8% to –6.1%, respectively. The relative percentage of Ni of F3 form increased slightly; Ni in F4 and F5 form increased about 4.4% and 6.5%, respectively, in the final sludge mixture (SS+68%HS) (Fig. 4). These results demonstrate that the two former forms of Ni were transformed into F4 and F5 forms.

In general, Zn and Ni in F4 and F5 forms were dominant in the final mixture, suggesting that Zn and Ni were more stable than in the initial sludge. The HS dosage had a negative correlation with the percentage of Zn in F1, F2, and F3 forms and Ni in F1 and F2 forms, and

Table 5 Heavy metals concentrations in the final mixtures for the different forms (unit: mg/kg)

Metal	F1	F2	F3	F4	F5
SS+0%HS					
Zn	116[a](6.6[b])	280 (16.0)	878 (50.2)	284 (16.2)	193 (11.0)
Ni	8 (26.4)	3 (10.7)	10 (32.3)	5 (17.3)	4 (13.3)
Cu	0 (0)	12 (5.3)	0 (0)	160 (73.6)	46 (21.1)
Pb	0 (0)	0 (0)	3 (5.9)	5 (9.1)	49 (85.0)
SS+7%HS					
Zn	75 (4.4)	212 (12.5)	800 (47.1)	423 (24.9)	190 (11.1)
Ni	7 (23.7)	3 (10.0)	10 (33.5)	6 (18.7)	4 (14.1)
Cu	0 (0)	7 (3.2)	0 (0)	156 (74.0)	48 (22.8)
Pb	0 (0)	0 (0)	0 (0)	2 (5.1)	45 (94.9)
SS+14%HS					
Zn	57 (3.7)	182 (11.6)	700 (44.7)	442 (28.3)	183 (11.7)
Ni	6 (21.8)	2 (8.5)	9 (34.2)	5 (19.3)	4 (16.2)
Cu	0 (0)	2 (1.2)	0 (0)	145 (74.0)	49 (24.8)
Pb	0 (0)	0 (0)	0 (0)	1 (3.0)	45 (97.0)
SS+28%HS					
Zn	48 (3.4)	155 (10.7)	594 (41.0)	480 (33.2)	170 (11.7)
Ni	5 (19.8)	2 (8.5)	9 (35.3)	5 (19.1)	4 (17.3)
Cu	0 (0)	0 (0)	0 (0)	141 (73.9)	50 (26.1)
Pb	0 (0)	0 (0)	0 (0)	1 (2.1)	45 (97.9)
SS+41%HS					
Zn	32 (2.5)	110 (8.4)	498 (38.1)	512 (39.1)	156 (11.9)
Ni	4 (18.6)	2 (7.8)	8 (34.9)	5 (20.1)	4 (18.6)
Cu	0 (0)	0 (0)	0 (0)	122 (72.3)	47 (27.7)
Pb	0 (0)	0 (0)	0 (0)	0 (0)	40 (100)
SS+68%HS					
Zn	28 (2.4)	86 (7.3)	399 (33.8)	528 (44.7)	140 (11.8)
Ni	3 (16.3)	1 (7.3)	7 (34.7)	4 (21.7)	4 (20.0)
Cu	0 (0)	0 (0)	0 (0)	96 (69.0)	43 (31.0)
Pb	0 (0)	0 (0)	0 (0)	0 (0)	33 (100.0)

[a] Values are means (n = 3).
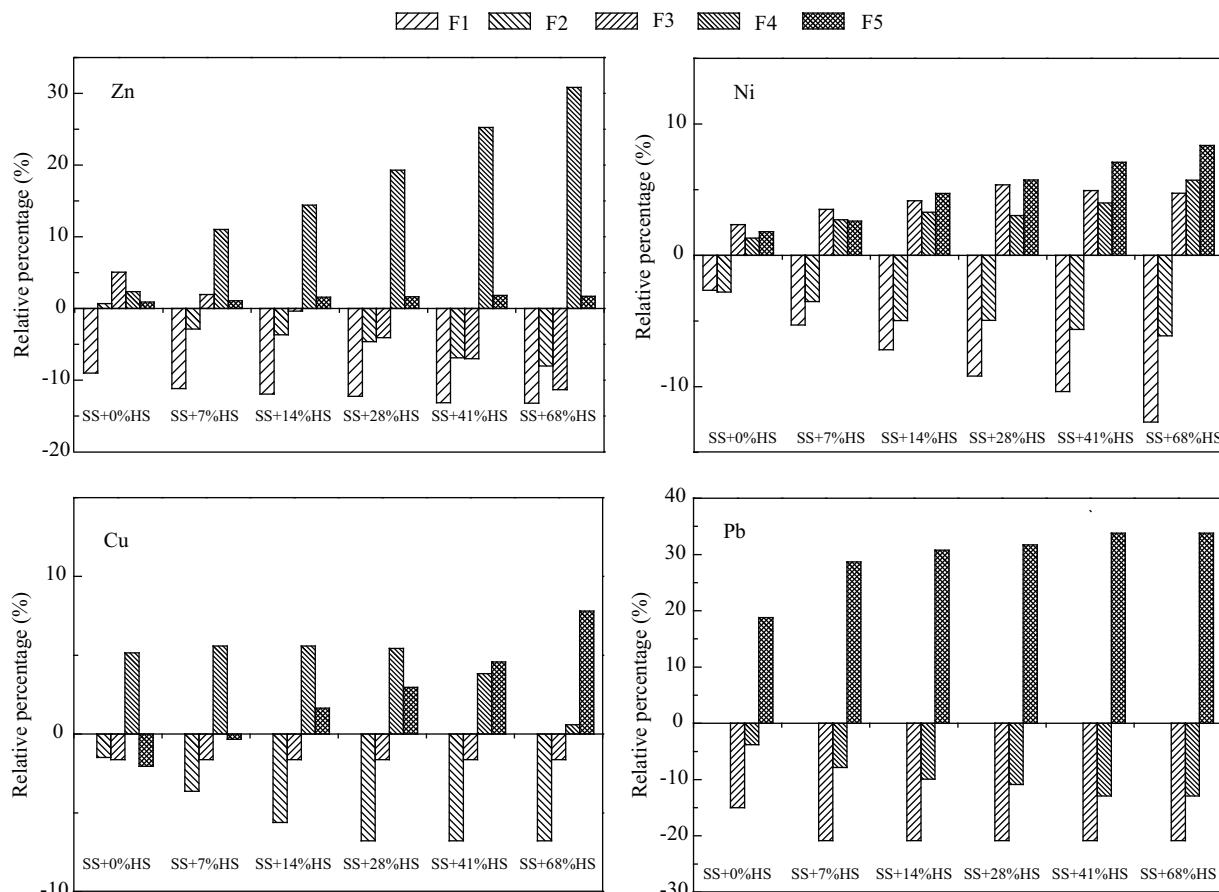[b] Percentage of each form of Zn, Ni, Cu, Pb in the final mixtures.

Fig. 4   Variations of relative percentage of each form of Zn, Ni, Cu, Pb in the final mixtures.
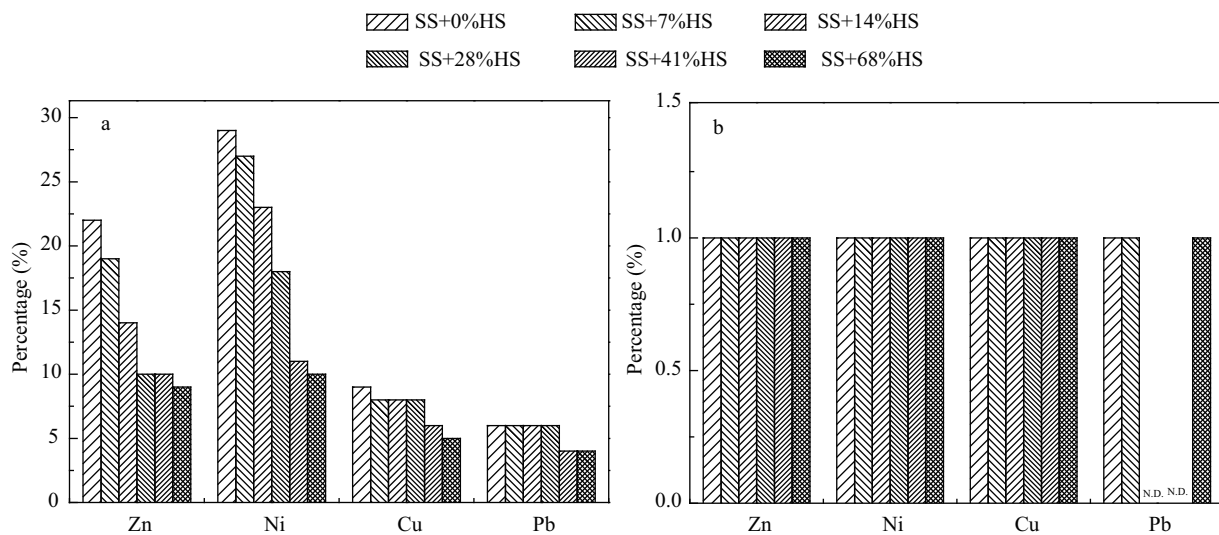

Fig. 5   Percentage of leaching heavy metals in the final mixtures at pH 4 (a) and pH 8 (b).

had a positive correlation with the percentage of Zn in F4 form and Ni in F4 and F5 forms (Table 6). The pH value of the final mixture also had a positive correlation with the three former forms of Zn and the two former forms of Ni, and had a negative correlation with the rest (Table 6). These results indicate that HS and pH are important factors influencing Zn and Ni forms. After 28 days, the pH in the mixture decreased substantially, which accelerated the mobilization of Zn and Ni, especially those in mobile forms (Yoo and James, 2002; Simpson

et al., 2002). Conversely, the sludge had a large amount of organic matter, such as protein and polysaccharides, which were easily degraded. Therefore, the Zn and Ni that were bound with this organic matter were easily released into the environment (Merring et al., 2003). In addition, humic substrate of aromatization increased as HS dosage increased. This organic matter contained large amounts of aromatic carbon, carboxyl, alkyl and other organic functional groups, and were easily complexes with Zn and Ni; therefore, Zn and Ni in F4 forms increased.

Amir et al. (2005) and Chirenje et al. (1999) showed that humic substrate promotes the mobilization of heavy metals transformed into stable forms, while Zheng et al. (2007) reported that pH influenced the evolution of heavy metals forms during composting. Amir et al. (2005) did not, however, find any relationship between pH and transformation of heavy metals in composting. These different results might depend on the raw materials used, different durations of compost, or different experiment durations. In our study, we found pH and heavy metal forms had strong relationships, but we did not reach the same conclusion as Zheng et al. (2007). He et al. (2009), however, reasoned the contradictory results about the relationships between pH and heavy metal forms were due to the same reasons as posited by us.

With the addition of 28% HS in the sludge, Cu was converted to F4 and F5 forms as 141 and 50 mg/kg, respectively, and the percentage of the two forms was 73.9% and 26.1% (Table 5), respectively. Figure 4 shows that without HS, Cu in F2, F3, and F5 forms were transformed to F4 form, while transformation changed with HS addition. Adding 68% HS to the sludge, Cu in F2 and F3 form were mainly converted to F5.

After 28 days, the Pb content of F5 form in the final mixtures increased from 33 to 49 mg/kg. The percentage was over 90.0% expect without HS. Figure 4 shows that Pb in F3 and F4 forms were transformed to the last fraction with the addition of 41% HS to the sludge.

Cu in F4 and F5 form and Pb in F5 form were dominant. As Cu has a high affinity for organic matter and due to the acidic environment, Cu in F2 form was easily released and ions were bound to one or more organic functional groups, mainly carboxylic, carbonyl, and phenolic groups (Nomeda et al., 2008). Previous studies (Brown et al., 2000; Torri and Lavado, 2008) have reported that Cu is preferentially associated with soil organic matter and can form stable complexes with organic matter at low pH. Amir et al. (2003) also reported that the formation of humic substrate transforms Cu to organic form during composting. The formation of Pb in F5 form was due to Pb strongly binding in the crystal structure (Virendra et al., 2009). Furthermore, HS content was negatively correlated with Cu and Pb in F4 form, while pH was negatively

correlated with Cu and Pb in F5 form (Table 6). The results proved that in an acidic environment, F1, F2, and F3 forms were easily released into water, and Cu and Pb form inner-sphere metal surface complexes with SiO_2 and Fe oxide from HS. In addition, Cu in F2 form showed a strong positive correlation with pH (Table 6), which was also observed in sludge composting performed by Cai et al. (2007).

2.2.3 Assessment of heavy metal mobility

Heavy metals in exchangeable (F1), carbonate-bound (F2), and Fe/Mn oxide-bound (F3) forms are considered more mobile and bioavailable. The organic matter/sulfide-bound (F4) and residual (F5) metals are stable and non-bioavailable (Zufiaurre et al., 1998). In our study, the ratio of the sum of heavy metals bound in mobile forms (F1–F3) to that in stable forms (F4 and F5) was used to evaluate the potential mobilities of heavy metals in the initial sludge and the final mixtures (Perin et al., 1997). The mobility of heavy metals in the mixtures after 28 days is shown in Table 7. The heavy metals with greater mobility in the initial sludge and the final mixtures were Zn and Ni; however, Cu and Pb content were higher than Ni (Table 3). Heavy metal mobilities in the initial sludge were in the following order: Zn > Ni > Pb > Cu. Thus, heavy metal mobility depended not only on total amounts, but also on their forms in the samples. With the addition of HS to the sludge, the mobility of heavy metals decreased substantially. With the addition of 41% HS, the mobilities of Zn, Cu, and Ni were relatively low (<1), but the mobility of Ni was still high. Heavy metal mobility in the final mixtures was, therefore, in the order of Ni > Zn > Cu = Pb.

2.2.4 Leaching test

Heavy metals display different mobilities at different pH values; hence, the ratio of leaching heavy metals at pH 4 and pH 8 to total heavy metals was used to investigate heavy metals leachability (Fig. 5). As shown in Fig. 6a, leaching Zn and Ni were higher at pH 4. The addition of HS effectively prevented heavy metals leaching from the samples (Zn from 9% to 22%, Ni from 10% to 29%, Cu from 5% to 9%, and Pb from 6% to 4%). These results indicate that the mobilities of Ni and Zn in low pH were higher, while Cu and Pb were relatively stable. In Fig. 6b, the leachabilities of the four heavy metals were very low (<1%) at pH 8. Under acidic conditions, Zn and Ni are released and thus pollute the environment.

Table 6　Pearson correlation between HS concentration or pH and the percentage of Zn, Ni, Cu and Pb form in the final mixtures ($n = 6$)

	F1	F2	F3	F4	F5
Correlation between HS concentration and percentage of metals					
Zn	−0.824*	−0.921**	−0.982**	0.958**	0.776
Ni	−0.952**	−0.893*	0.691	0.947**	0.955**
Cu	–	−0.773	–	−0.905*	0.984**
Pb	–	–	−0.506	−0.846*	0.738
Correlation between pH and percentage of metals					
Zn	0.980**	0.963**	0.925**	−0.955**	−0.926**
Ni	0.967**	0.954**	−0.936**	−0.895*	−0.931**
Cu	–	0.974**	–	0.571	−0.923**
Pb	–	–	0.854*	0.979**	−0.968**

* $P < 0.05$; ** $P < 0.01$.

Table 7　Mobility of heavy metals in the initial sludge and the final mixtures after 28 days

Sample	(F1 + F2 + F3)/(F4 + F5)			
	Zn	Ni	Cu	Pb
Initial sludge	3.17	2.63	0.09	0.26
SS + 0% HS	2.67	2.26	0.06	0.06
SS + 7% HS	1.76	2.04	0.03	0
SS + 14% HS	1.48	1.80	0.01	0
SS + 28% HS	1.18	1.73	0	0
SS + 41% HS	0.90	1.54	0	0
SS + 68% HS	0.68	1.31	0	0

3 Conclusions

Generally, total heavy metal content in sewage sludge from Quyang Municipal Wastewater Treatment Plant was lower than the standard listed in GB18918-2002. Only Zn exceeded the GB 4284-84 standard for agricultural use.

In the initial sludge, most Zn and Ni were associated with the mobile F1–F3 forms (>70%) and most Cu and Pb were found in stable F4 and F5 forms (>80%). In the final mixtures, Zn and Ni were mainly found in F3 and F4 forms. For Zn, F1, F2 and F3 forms were transformed to F4 and F5 forms, while for Ni, F1 and F2 forms were transformed to F1, F2, and F3 forms. Both Cu and Pb were strongly associated with the stable forms (F4 and F5). For Cu, F2 and F3 forms were major contributors, while for Pb, F3 and F4 forms were major contributors to F5 form. Heavy metal mobilities in the final mixtures were in the following order: Ni > Zn > Cu = Pb. With the addition of 41% HS to sludge, the mobilities of Zn, Cu, and Ni were relatively low (<1), but the mobility of Ni was still high. Humus soil dosage and pH had strong correlations with different forms of heavy metals. Leaching tests showed that the mobilities of Zn and Ni in low pH (pH 4) were higher than in high pH (pH 8); however, the mobilities of Cu and Pb did not change substantially.

Acknowledgments

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