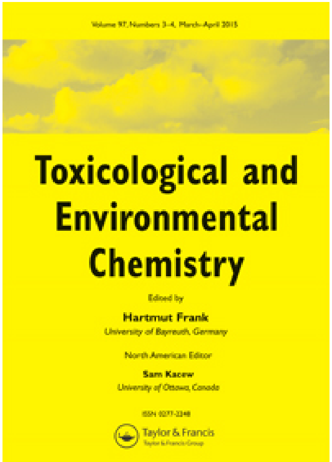

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Land utilization of sewage sludge has become one of the major issues in environmental mitigation in China. This is particularly important in the Loess Plateau of Northwest China. Sludge enriched with organic matter and nutrients might effectively help to resolve the problems of silty loess soils as evidenced by porosity defect, structural alterations and absence of fertility. After sludge flows into the loess, irrigation water contains heavy metals that leach and migrate into sludge and consequently related human health risks may occur that raise concerns; and this situation needs to be rectified. The characteristics of the vertical migration and composition form change of Cu, Zn, and Cd from composted sludge to loess, and corresponding influencing factors, were investigated by performing a soil column simulation test under the leaching treatment of a one-year irrigation water capacity. Results demonstrated that: (1) composted sludge significantly improved loess fertility, and irrigation leaching transported only a small quantity of organic matter in sludge in the plough layer; (2) although some of the Cu, Zn, and Cd in composted sludge migrated to and concentrated in the middle and upper layers of the soil column during leaching, these metals were mostly retained in the plough layer; and (3) after the leaching treatment of the one-year irrigation water capacity, the compositions of Cu, Zn, and Cd (particularly Cu and Zn) in both plough layer and loess began to stabilize with low concentrations in the exudate. These findings confirmed the applicability of composted sludge in loess regions. This study provided a new insight into the sludge reuse in alkaline soils in arid and semi-arid region.

Keywords: composted sludge; form transformation; heavy metal; leaching; loess; migration

Introduction

China's current treatment rate of domestic sewage has reached 72% and is predicted to reach 85% by 2015 (State Council 2011). Annual sludge quantity would soon exceed 30 million tonnes because of continuous increase in urbanization and sewage treatment capacity (PRC Ministry of Housing and Urban–Rural Development 2011). Sludge poses the China's second major solid waste problem, next to municipal solid waste (Zeng et al. 2012). Therefore, the scientific and reasonable disposal of sludge has become an urgent ecological and environmental issue (Singh and Agrawal 2007).

Among the existing sludge disposal methods, such as land application, landfill, incineration, and building materials (Lowman et al. 2013; Yu et al. 2005; Donatello and Cheeseman 2013; Yang et al. 2013), land application has been considered to have the

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highest development potential (Stabnikova et al. 2005). In view of the high content of organic matter, nitrogen (N), phosphorus (P), potassium, and other various microelements in sludge, its reuse on farmland might improve soil structure and increase soil fertility, thereby benefiting plant growth and yield increase. More than 40% of sludge has been reused on farmlands in Europe and the United States (Corrêa et al. 2006; Werle and Wilk 2010; Fytili and Zabaniotou 2008). Sludge land application is also one of the recommended disposal methods in China (PRC Ministry of Housing and Urban–Rural Development 2011; Ministry of Environmental Protection 2010). In particular, sludge reuse on loess areas with poor fertility might improve and offset inadequate water stability aggregates as well as poor water retention and nutrient preservation of loess soils which are produced by loose powder structure of loess soils. In this sense, sludge application can be very promising and valuable in the Loess Plateau of Northwest China. However, since sludge contains various harmful heavy metals, long-term application might induce enrichment and migration of heavy metals into the soil (Wu and Chen 2005), which creates a major constraint for sludge land application. A number of studies reported enrichment and migration of heavy metals in sludge into the soil. Sludge treatment was examined in a desert in Yunlin City (Shanxi Province, China), the sandy soil in Nigeria, the silty loam in the United States, the farming clay in Sweden, and the forest soil in the Mediterranean where enrichment and migration of heavy metals in sludge into the soil was noted (Huang et al. 2008; Mbila et al. 2001; Baveye et al. 1999; Bergkvist et al. 2003; Toribio and Romanyà 2006). However, only a few studies investigated leaching-induced migration and ionic change of heavy metals from sludge to loess.

The aim of this study was to simulate conditions in sludge land application in loess plateaus in China. The characteristics of the vertical migration and compositional form change from sludge to loess of three heavy metals (Cu, Zn, and Cd) were analyzed by performing an indoor soil column simulation test under irrigation leaching. The study was designated to provide scientific insight into heavy metal control and establishment of a technical evaluation system for risk assessment during sludge application in loess regions.

Materials and methods

Test materials

The test material consisted of a stable mixture of dewatered sludge collected from the Qilihe Sewage Treatment Plant in Lanzhou City and wheat-straw collected from farmlands (dry weight, 7:1) after artificial turning and composting under static aerobiotic conditions for two months. The test loess was Malan loess (Q_3^{2eol}, pH = 8.49) collected from a 30 cm deep layer at the foot of Cuiying Mountain in Yuzhong Campus of Lanzhou University. Loess is mainly composed of particles ranging from 0.005 to 0.075 mm in diameter. To elucidate major heavy metals present in and compositional form changes from composted sludge to loess, the collected composted sludge was evenly sprayed with a nitrate solution of Cu, Zn, and Cd to maintain total heavy metal content at approximately A-level sludge concentration, as suggested in *Sludge Disposal of Municipal Wastewater Treatment Plant: Agricultural-purpose Sludge* (CJT 309-2009). Sludge was stored under natural conditions for a half month to facilitate stabilization to be used in a follow-up test. The pot experiment on sludge-modified loess revealed that the optimal dry weight ratio of sludge and loess for plant growth ranged from 0.045 to 0.091. This test determined this ratio as 0.07. Loess mixed with test sludge was termed composite soil. Tap water (pH = 6.92) containing 0.0942 mg L^{-1} Cu, 0.5613 mg L^{-1} Zn, and 0.0001 mg L^{-1} Cd was used

Table 1. Distribution of the chemical forms of Cu, Zn, and Cd in raw materials.

			EF		CBF		FMOF		OBF		RF	
Heave metal	Material	TA (mg kg^{-1})	mg kg^{-1}	%	mg kg^{-1}	%	mg kg^{-1}	%	mg kg^{-1}	%	mg kg^{-1}	%
Cu	Composted sludge	72.9785	2.6950	3.69	1.2100	1.66	0.6750	0.92	45.4150	62.23	22.9835	31.49
	Test sludge	507.9460	27.5262	5.42	27.7811	5.47	3.6932	0.73	302.1689	59.49	146.7766	28.90
	Composite soil	49.76	1.2195	2.45	0.5248	1.05	0.5948	1.20	6.7123	13.49	40.7037	81.81
	Loess	20.0190	0.7046	3.52	0.0550	0.27	0.1599	0.80	0.1749	0.87	18.9245	94.53
Zn	Composted sludge	297.8611	15.1800	5.10	4.7250	1.59	130.1750	43.70	72.1350	24.22	75.6461	25.40
	Test sludge	1452.8486	79.2604	5.46	125.9670	8.67	658.2159	45.31	237.0665	16.32	352.3388	24.25
	Composite soil	186.1369	8.1267	4.37	8.2817	4.45	65.4588	35.17	22.6060	12.14	81.6637	43.87
	Loess	90.0230	3.8831	4.31	1.5892	1.77	3.6282	4.03	0.0600	0.07	80.8626	89.82
Cd	Composted sludge	2.0590	0.9200	44.68	0.1390	6.75	0.1745	8.48	0.1486	7.22	0.6768	32.87
	Test sludge	5.3773	2.6949	50.12	0.5396	10.04	0.1241	2.31	1.4408	26.79	0.5778	0.75
	Composite soil	0.6322	0.2784	44.04	0.0464	7.34	0.0414	6.55	0.0367	5.81	0.2292	36.26
	Loess	0.1854	0.0251	13.53	0.0050	2.69	0.0351	18.90	0.0036	1.96	0.1167	62.91

Note: EF = exchangeable fraction, CBF = carbonate-bound fraction, FMOF = Fe−Mn oxide fraction, OBF = organic-bound fraction, RF = residual fraction, TA = total amount.

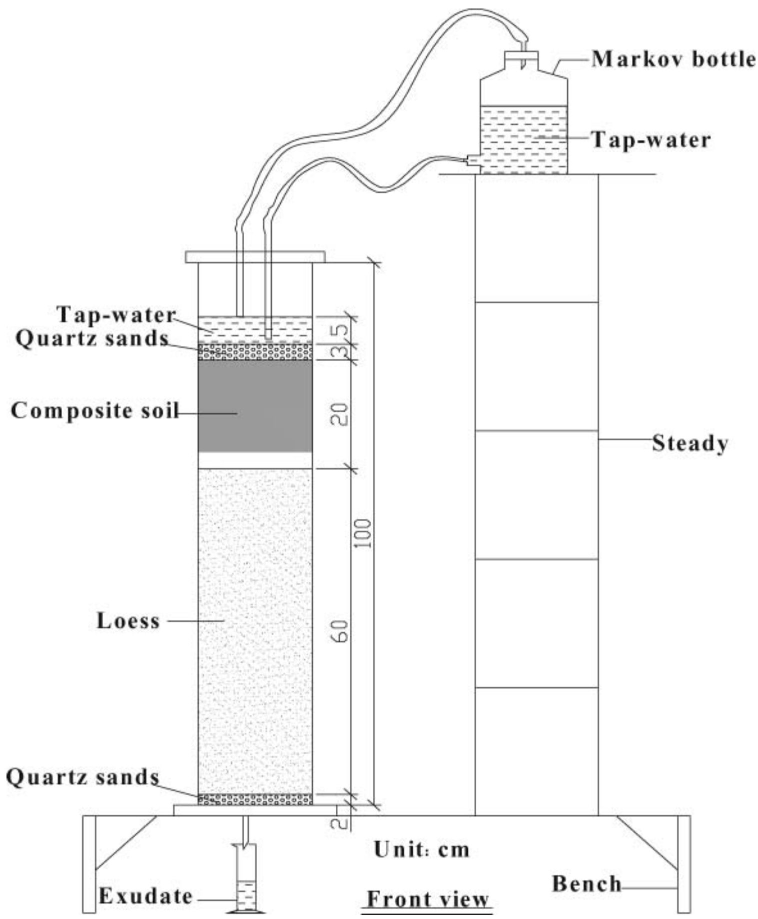


Figure 1. Simulation test apparatus.

for leaching. The tested total concentrations and form concentrations of Cu, Zn, and Cd in the test raw materials are listed in Table 1.

Test design

The test apparatus consisted of a homemade simulation soil column, steel support bench, Markov bottle, and the collection of exudate cylinder (Figure 1). The simulation soil column is a cylinder (length: 100 cm; inner diameter: 18.6 cm) made of transparent organic glass. Before sample filling, the column was cleaned with tap water, rinsed with distilled water three times, and then dried. A piece of nylon coarse mesh was placed inside the bottom of the soil column and covered by 2 cm thick clean quartz sand. A piece of nylon fine-mesh filter filled with 60 cm thick loess was placed on top of the quartz sand. Sample filling was prepared through the following steps: (1) the column was filled with loess in a layer-based manner with weighing (2 cm thick for each layer), and filling density was controlled at the natural dry density of loess (1.36 g cm^{-3}); (2) a piece of stick (diameter: 5 cm) and another smaller stick were simultaneously used for artificial natural compaction and for in-wall surrounding compaction, respectively, in order to prevent from in-wall effect; (3) a 20 cm thick layer of composite soil served as the plough layer; and (4) a piece of nylon coarse mesh covered by 3 cm clean quartz sand (liquid buffer) was paved above the composite soil. In this test, a Markov bottle was used for liquid storage. Liquid was injected into the soil column from the top, keeping the top liquid layer stable at 5 cm high ensuring the stable piston discharge of leachate under the effect of gravity. The exudate was stored in a 1L cylinder below the soil column.

To simulate the migration behavior and composition form changes of heavy metals from composted sludge to loess in China's loess plateau regions under irrigations, a leaching experiment was conducted with the use of tap water. In accordance with the annual irrigation norm, which ranged from 300 to 400 m^3/mu/year (this study considered 350 m^3/mu/year) for dry crops in northwest China (*Main Crop water requirement and irrigation of China*) (Chen et al. 1995), 14 L of tap water was continuously injected into the soil column during the simulation test. The test lasted over 12 days from May 30 to June 10.

Soil column sampling and analysis method

During the simulation test, experimental phenomena and exudate data were observed and recorded daily at 10:00 am. The exudates collected every day were stored in water bottles cleaned with deionized water and labeled with the corresponding date. The simulation test was ended after no exudate was collected for three successive days. The heavy metal concentrations in the exudates were tested by using the flame method and graphite furnace method of SOLLAR AA M6 atomic absorption spectrometer (Thermo Fisher, the United States).

After the simulation test was conducted, composite soil was lowered for approximately 1 cm, whereas loess remained unchanged. Sampling from the soil column was prepared through the following steps: (1) remove the top quartz sand and the composite soil successively; (2) collect samples with a cutting ring (5 cm depth) every 10 cm depth; (3) place the collected samples from each layer on ceramic plates for natural drying; (4) grind the samples and screen them with a 100-mesh screen; and (5) store the samples in sealed bags for subsequent analysis.

Organic matter in the soil samples were tested in a potassium dichromate-sulfuric acid oil bath (NYT 1121.6-2006). The heavy metals in the soil samples were successively digested with nitric acid, hydrofluoric acid, and perchloric acid. Thus, total heavy metal content was assessed by the flame method of SOLLAR AA M6 atomic absorption spectrometer. Five forms of heavy metals (EF, CBF, FMOF, OBF, and RF) in the exudates samples were collected through Tessier five-step continuous extraction method (Tessier et al. 1979). Afterward, the form changes of these five heavy metals were tested by the flame method or graphite furnace method of SOLLAR AA M6 atomic absorption spectrometer.

Results and analysis

Effect of leaching on organic matters in sludge

Organic matter is major nutrient source for soil. A major purpose of composted sludge reuse in loess is to increase organic matter content in loess and consequently improve its fertility. Some organic matter in the composite soil is transported by leachate. Moreover, the migration and composition form of heavy metals are also affected by leaching treatment. In the present study, organic matter content in the test sludge was found to be

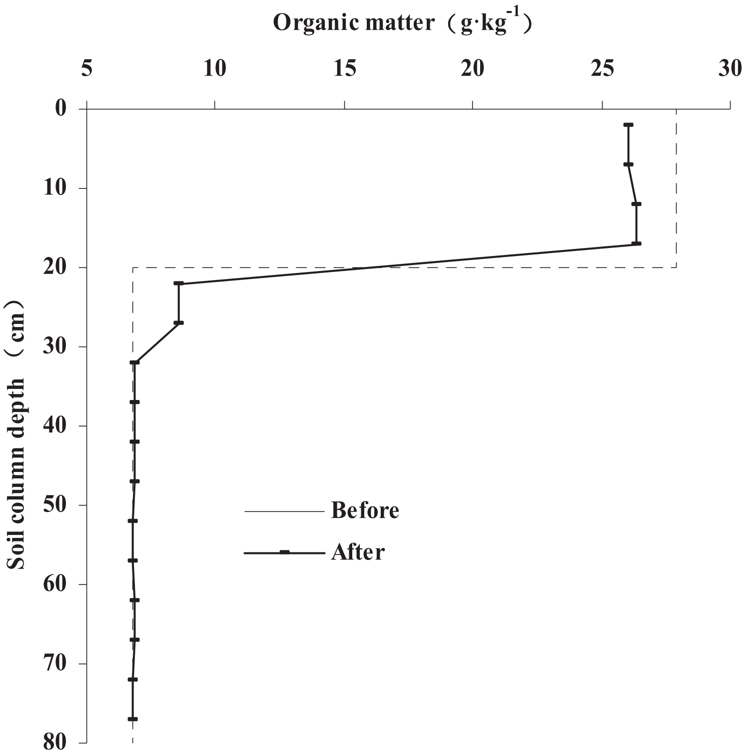


Figure 2. Distribution of organic matters in soil column before and after leaching treatment.

337.32 g kg^{-1}, which was higher than the demanded level for agricultural application of sludge proposed by CJ-T 309-2009. Organic matter content was found to be 27.87 and 6.81 g kg^{-1} in composite soil and loess, respectively, indicating a 309.25% increase in organic matter in soil. The vertical distribution and variation in organic matter in the soil column before and after leaching treatment are shown in Figure 2.

Figure 2 illustrates that after the leaching treatment of the one-year irrigation water capacity, most organic matter in sludge was still retained in the composite soil (at a depth ranging from 0 to 20 cm), suggesting a small average loss (5.97%). Organic matter exhibited an increase at a depth ranging from 20 to 32 cm (quicker at upper position) but these were kept similar with those in loess at a depth below 32 cm. These findings indicate that irrigation only slightly affects migration and loss of organic matter in sludge in the plough layer. Barber (1962) postulated that the three forms of nutrients migration (intercepted, mass-flow, and diffusion) are essential for migration of soil nutrients. Previously, Liang et al. (2009) noted that after the sludge flows into the alkaline silty loam, most of soil nutrients were still retained in the plough layer after the leaching treatment, and results indicate this was related to the interception effect of soil nutrients. Therefore, the migration of organic matter in loess is related to the interception effect of loess on organic matter.

Leaching-induced migration of heavy metals in loess

A total of 6470 ml exudates were collected in the simulation test.

Copper, Zn, and Cd concentrations in the exudate fluctuated as illustrated in Figure 3. However, the concentrations were still higher than those detected in tap water, indicating

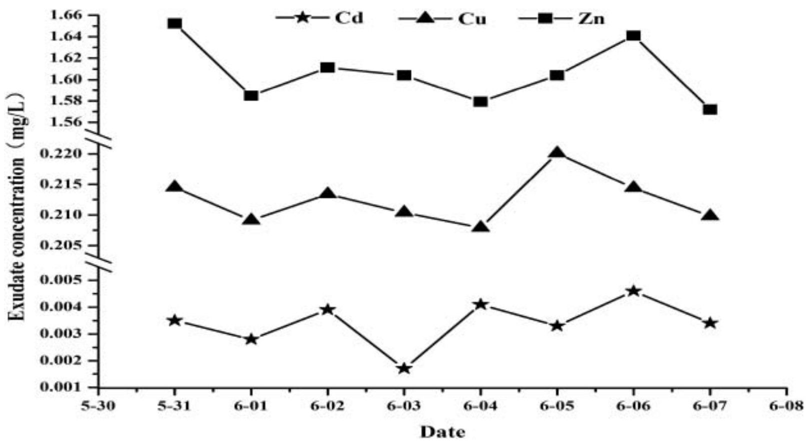


Figure 3. Variation of exudate concentration.

that Cu, Zn, and Cd in the soil column migrated with the leachate. A total of 0.7658 mg Cu, 6.7609 mg Zn, and 0.0210 mg Cd excluding the corresponding contents in tap water were filtered from the soil column, accounting for 0.11%, 0.23%, and 0.28% of their concentrations before leaching, respectively. This finding indicates that most heavy metals in composted sludge were still retained in the soil column after leaching with the one-year irrigation water capacity. This observation was in agreement with data of Alloway and Jackson (1991), who postulated that metals applied with sewage sludge may be retained in the soil as a result of (1) adsorption on hydrous oxides, clays, and organic matter; (2) formation of insoluble salts; or (3) presence of residual sewage sludge particles.

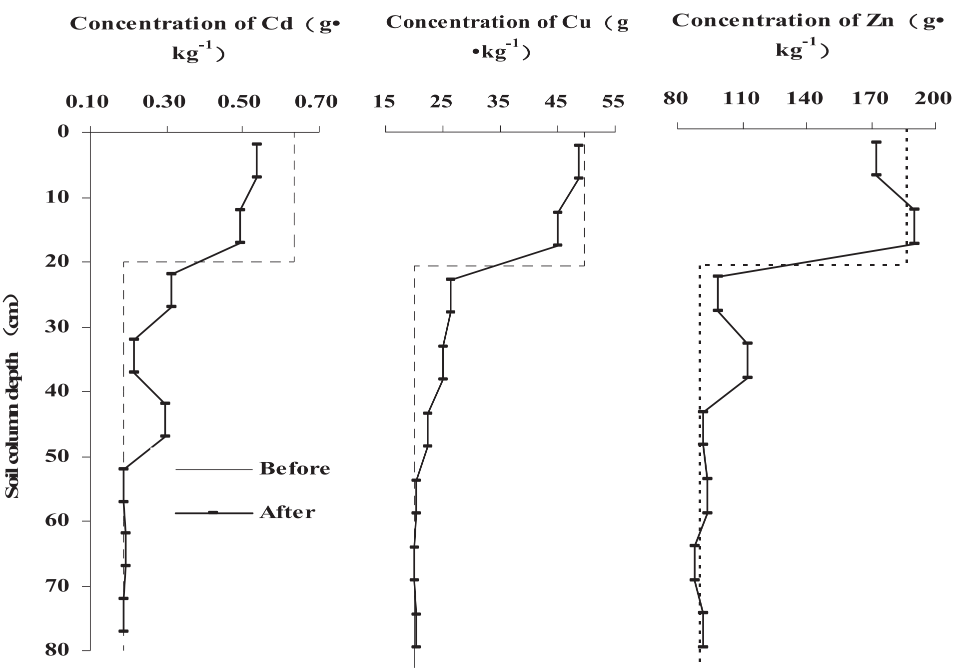


Figure 4. Concentration variations of Cd, Cu, and Zn before and after the leaching treatment.

Figure 4 shows that after the leaching treatment of the one-year irrigation water capacity, the concentrations of Cd, Cu, and Zn in the composite soil at a depth extending from 0 to 20 cm decreased to a certain level while Zn accumulated at the depth ranging from 12 to 17 cm. In general, the maximum concentrations of heavy metals were still present at the top soil column. Cd and Cu in composted sludge migrated downward for approximately 30 cm with the leachate, as evidenced by significant concentration growth at the depth from 20 to 50 cm and the apparent enrichment of Cd at the depth ranging from 42 to 47 cm. These levels were similar to concentrations levels in loess below 50 cm. Zn in composted sludge migrated downward for approximately 20 cm with the leachate, as manifested by significant concentration growth at the depth from 20 to 40 cm and enrichment at the depth of 32−37 cm with similar levels as loess below 40 cm. These findings demonstrate that Zn, Cd, and Cu in composted sludge are readily transported by leachate. However, most of the metals are still retained in the plough layer. These data are in agreement with Loganathan and Hedley (1997), Al-Wabel et al. (1998), and Barbarick et al. (1998).

Distribution characteristics of heavy metal forms in loess

In view of the differences of heavy metal compositions, the organic matter content, and the alkalinity between loess and composted sludge, it is conceivable that the compositional forms and bio-availability of heavy metals in composted sludge may change when metals migrate downward with the leachate. Thus, understanding the distribution characteristics of heavy metal forms in loess is essential for sludge land application and environmental safety.

Cu forms

As an essential microelement for healthy growth of animals and plants, Cu is vital to normal physiological actions and metabolism. However, excessive Cu produces adverse effects and may be lethal to animals and plants. Table 1 shows that before the leaching treatment, Cu in composite soil (0−20 cm) existed mainly in biological stability form, including 81.81% RF and 13.49% OBF. Cu below 20 cm was predominantly RF (94.53%). Data indicate that before the leaching treatment, Cu in loess was more stable than those found in composite soil.

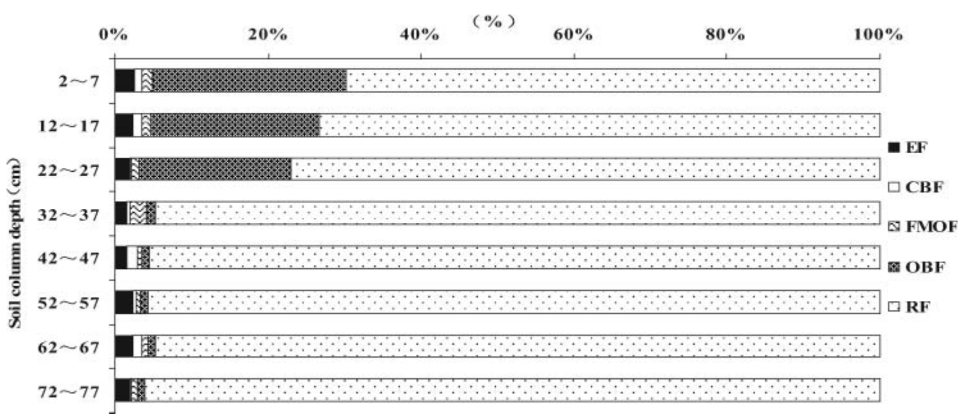


Figure 5. Distributions of Cu forms in soil column after the leaching treatment.

Figure 5 illustrates that after the leaching treatment, the sum of the EF and CBF of Cu in composite soil decreased by approximately 0.5%. RF was dominant for Cu in the whole soil column. OBF ranged from 19.8% to 25.27% at the depth of from 0−27 cm because of the leaching effect of heavy metals and organic matter in composted sludge. This behavior was mainly related with the potent binding of Cu with organic matter (Campbell 1989). Moreover, OBF variation law conforms to the distribution of organic matter in the soil column (Figure 2). At the depth of 32−80 cm, CF reached an amount higher than 94% and equal to that in loess. Comparative analysis based on Figure 4 suggested that the form composition of Cu at the depth extending from 32 to 52 cm was similar to that found in loess, indicating that compositional forms of Cu changed during the leaching-induced migration. In essence, loess inactivated Cu and decreased its bio-availability. In general, heavy metal composition forms are directly influenced by the physical composition and chemical properties of soil, especially pH and organic matter. It is well known that loess contains a small amount of organic matter because of its alkalinity.

Zn forms

Zn is an essential micronutrient for animals and plants which might increase crop yield and improve crop quality. However, excessive Zn may accumulate in plants and pose risks to human health when these plants enter biologic or food chain. Chen et al. (2003) reported that Zn was the heavy metal with the highest average content in China's municipal sludge. Therefore, increased attention needs to be given to variation of its EF with high bio-availability and CBF during sludge land applications.

Table 1 illustrates that EF with high bio-availability and CBF in composite soil and loess were 8.82% and 6.08%, respectively, indicating low bio-availability of Zn in loess. The sum of FMOF, OBF, and RF in composite soil was 91.18%, which was lower than that in loess (93.92%). In addition, RF accounted for 89.82% Zn in loess, which is more than twice that in composite soil (43.87%). Data indicate that loess has higher Zn stability than composite soil before leaching.

Figure 6 illustrates that after the leaching treatment, the sum of EF and CBF in composite soil at the depth of 0−20 cm decreased by approximately 1%. As a result of leaching, Zn increased slightly at the depth extending from 22 to 27 cm (Figure 4) and had higher CBF, FMOF, and OBF compared to loess. This observation indicates that Zn in

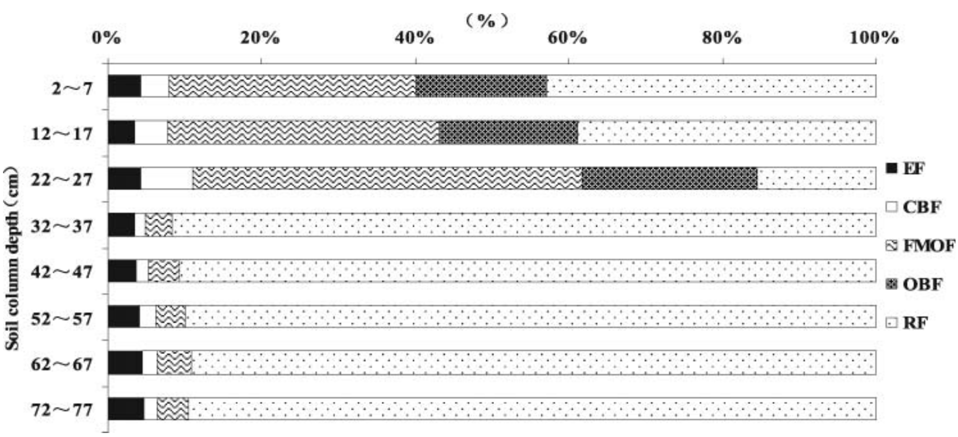


Figure 6. Distributions of Zn forms in soil column after the leaching treatment.

loess mainly exists in FMOF and OBF, followed by RF and CBF. The reason is that the rich Fe and Mn as well as alkaline environment (pH = 8.49) in loess are beneficial to the production of CBF and FMOF (Singh et al. 1999; Li et al. 2004). Further, organic matter that migrated from surface sludge can increase OBF of Zn (Fang et al. 2009). Although Zn was enriched at the depth of 32−37 cm (Figure 4)), Zn predominantly existed in RF (89.10%−91.72%), leaving only 8.28% to 10.9% for the rest of the four forms in the soil column below 32 cm. Such form composition was similar to that in loess. OBF only accounted for a small amount, which is difficult to present in Figure 6. This small amount was produced by the lower content of organic matter in loess and slight migration of organic matter from sludge. Overall, loess stabilized the composition form change and decreased bio-availability of Zn during the leaching-induced migration.

Cd forms

As one of the heavy metals with the most potent toxicity, Cd tends to be bio-accumulated in biotic environment. (Prokop et al. 2003).

From Table 1, before the leaching treatment, Cd in composite soil was composed of 44.04% EF with high bio-availability, 36.26% most stable RF, 7.34% CBF, 6.55% FMOF, and 5.81% OBF. Cd in loess included 62.91% RF, 13.53% EF, and 18.90% FMOF. Therefore, loess has higher Cd stability than composite soil.

In Figure 7, composition form of Cd ranged from 0 to 52 cm after leaching differed significantly to that prior to leaching treatment. As a result of leaching, EF in composite soil (0−20 cm) decreased by 17.06% and increased markedly from 22 to 52 cm depth. This rise was attributed to the potent migration capacity of EF (Sun et al. 2003). Comparison of Figures 4 and 7 indicates that leaching produced Cd accumulation in partitions, a significant elevation in OBF from 22 to 27 cm depth, and a significant increase in FMOF from 32 to 52 cm depth. These changes are attributable to leaching-induced migration of Cd from composite soil, organic matter from sludge, and loess properties such as alkalinity as well as richness in Fe and Mn compounds. Naidu et al. (1997) and Zhang and Li (2007) showed that the alkaline soil is good for the production of and FMOF. In addition, Childs and Leslie (1977) indicated that Cd content of CBF in loess was positively correlated with Fe and Mn in loess.

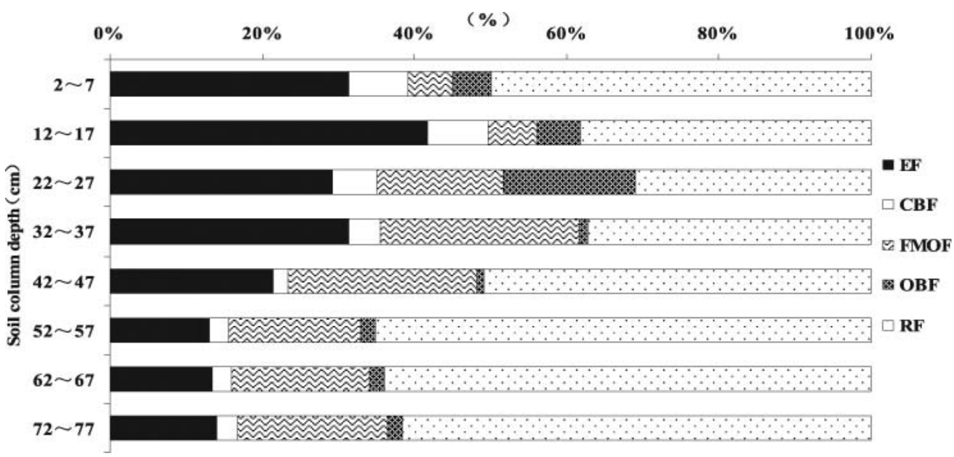


Figure 7. Distributions of Cd forms in soil column after the leaching treatment.

In summary, after continuous leaching treatment of the one-year irrigation water capacity, the composition form of Cd in composite soil (0−20 cm) began to stabilize. The composition form of Cd in the accumulation layer was more active than in loess below 20 cm, but less active than that in composite soil. Data indicate that loess retains Cd and stabilize its composition form to a certain extent. Unlike Cu and Zn, Cd was predominant in EF, resulting in its high bio-availability and toxicity. As a result, attention needs to be given to Cd during the sludge land application.

Leaching mechanism

In leaching experiments, water-karst leaching, precipitation, and adsorption affected the migration and transformation of heavy metals in soil column. The former generated exudate was alkaline, and resulted in heavy metals precipitation or complex with gel.

Water−karst filtration

Zhang et al. (2011) and Zeng et al. (2008) noted that, because minerals such as quartz, feldspar, and calcium carbonate were rich in loess, the tap water which contained dissolved CO_2 from the air and the biodegradation process of sludge organic matter in the plough layer might affect the leaching of feldspar minerals and dissolution of calcium carbonate in leaching experiments. This may be described by following reactions:

$$Na_2Al_2Si_6O_{16}(\text{sodaclase}) + 2CO_2 + 3H_2O \rightarrow 2HCO_3^- + 2Na^+ + H_4Al_2Si_2O_9(\text{secondary mineral}) + 4SiO \quad (1)$$

$$CaO \cdot 2Al_2O_3 \cdot 4SiO_2(\text{anorthite}) + 2CO_2 + 5H_2O \rightarrow 2HCO_3^- + Ca^{2+} + 2H_4Al_2Si_2O_9(\text{secondary mineral}) \quad (2)$$

$$CaCO_3 + H_2O + CO_2 \rightarrow 2HCO_3^- + Ca^{2+} \quad (3)$$

These reactions result in increasing alkalinity ion HCO^{3-}, alkali, and alkaline earth metal ions Na^+ and Ca^{2+} in the exudate, with pH elevated from 6.92 in the tap water to 7.2−7.8 in exudate. Thus, the exudate was alkaline. In addition, the secondary clay minerals were increased in loess. The two reactions differed from each other. The process of non-congruent dissolution of feldspar which can also be termed as leaching (Shen et al. 1993), is a kind of thermodynamically irreversible process during the whole experimenting period. On the other hand, the reaction of calcium carbonate was a congruent dissolution which was reversible and dissolution occurred in the early stage of the experiment. In the mid-to-late stage, because of increasing Ca^{2+} and alkalinity produced by the leaching of feldspar, calcium carbonate became secondary precipitation. Hence, the main sources of leaching are aluminosilicate minerals of loess, such as feldspar.

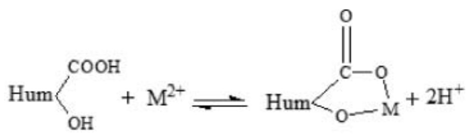
Precipitation

The major components of the alkalinity in the solution comprise the incremental HCO_3^- derived from the leaching and the OH^- and CO_3^{2-} in the solution. The relationship to pH was determined by carbonate-balanced system. HCO_3^- played a major role in the pH of leaching experiment, followed by OH^- and CO_3^{2-}. The hydroxides and carbonates of Cu, Zn, and Cd are insoluble salts, and their solubility product KS (2.2×10^{-20}~$5.27 \times$

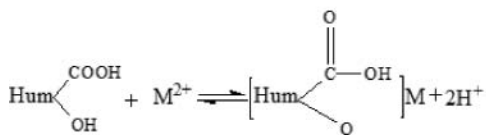
10^{-15}, 1.0×10^{-12}~1.40×10^{-10}) are smaller than KS of calcium carbonate (3.36×10^{-9}). Therefore, the alkalinity ions of OH^-, CO_3^{2-}, and heavy metals (Cu, Zn, and Cd) lead to precipitation reactions, producing Me(Cu, Zn, Cd)$(OH)_2$(s) and Me(Cu, Zn, Cd) CO_3(s). Moreover, humus is rich in composted sludge. According to previous studies, multivalent cation might bind separate humus molecules together to form a chain structure, leading to coagulation processes (Stevenson 1994; Chen et al. 2008). These two functions contributed to the stabilization of heavy metals during the leaching-induced migration in the present study and are attributed to be the basis of most of the heavy metals still being retained in the plough layer.

Adsorption

It was found that adsorption occurred in the whole section of the soil column. The clay mineral surface adsorbs heavy metals. In addition, the colloids humus of sludge or clay-humus particles surface also adsorb heavy metals. Clay minerals mainly originate from loess clay, the component of Malan loess clay of Lanzhou is about 10%−20% (Li et al. 2007), followed by clay minerals and hydroxide colloids of Mn, Al, Si, and Fe oxides produced by leaching (Ashworth and Alloway 2008). Heavy metals adsorption of humus mainly occurs in farming surface, followed by the topside of the soil column, namely, the distribution of organic matter or humus site. It was thought that Cu, Zn, and Cd adsorption of humic colloids were predominantly achieved by exchange interaction and ion chelation (Lin and Xu 2008; Chen and Sun 2002; Hao et al. 2010). Humus adsorption mechanism of heavy metal is described by following reactions:



(4)



(5)

From (4) and (5), the adsorption capacity of humus is related to the pH. The higher the pH in the reaction system, the more adsorption of humus occurs. On the other hand, since loess was alkaline, humus might influence the retention and stabilization of heavy metals in the leaching process. In this sense, the composition form of Cu, Zn, and Cd exhibited stability after one-year irrigation and leaching in the soil column.

In general, leaching of aluminosilicate minerals feldspar in loess was a reaction result of heavy metal precipitation and adsorption. Adsorption and precipitation were the main reasons that Cu, Zn, and Cd showed a steady-state trend in the soil column. These two processes also contributed primarily to the retention of most of heavy metals in the plough layer after the leaching treatment of one-year irrigation water capacity.

Conclusions

(1) The present study made efforts to elucidate sludge land application in China's loess plateaus. The results filled knowledge gaps in sludge reuse in alkaline soil

of arid and semi-arid region. Sludge reuse in loess might improve the fertility of loess significantly. It was found that irrigation leaching transported organic matter in sludge in the plough layer whereas organic matter presented poor migration in loess. Some of the heavy metals migrated and concentrated in the middle and upper layer of the soil column. Among these three heavy metals, Cu and Cd were more readily migrated with the leachate compared to Zn.

(2) After the leaching treatment of the one-year irrigation water capacity, the composite soil possessed less unstable heavy metal forms with high bio-availability (EF and CBF), but more CBF of Cu. This indicates that leaching treatment facilitated stabilization composition form of heavy metal in the plough layer. As a result of the irrigation leaching effect, some organic matter and heavy metals in composite soil might migrate to and concentrate in the middle and upper layers of the soil column, accompanied with form stabilization of Cu, Zn, and Cd. Composition form of Cu and Zn in their enrichment depth was equal to those in loess, indicating better stabilization effect and lower bio-availability in comparison with those of Cd. Change and stabilization in heavy metal forms were influenced by properties of heavy metals, loess alkalinity, Fe and Mn content, and organic matter levels.

(3) The leaching of aluminosilicate minerals feldspar in loess was the reaction result of heavy metal precipitation and adsorption. Adsorption and precipitation were the main contributors to the steady state of Cu, Zn, and Cd in the soil column. These two processes also contributed to the retention of heavy metals in the plough layer after the leaching treatment of one-year irrigation water capacity. Data indicated that sludge reuse on loess regions is feasible. However, migration and composition form change of heavy metals under leaching by long-term irrigation water capacity or application of composted sludge needs further studies.

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