



Evolution processes of trace metal speciation in leachates with different ages from Laogang Refuse Landfill, Shanghai

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ABSTRACT

Trace metals in leachates are forced to be removed with the implement of the latest Chinese standards of landfill pollution control (GB-16889, 2008), and understanding the speciation distribution processes is important for the development of the leachate treatment process. In this work, evolution processes of trace metal speciation were investigated according to the size fractions, hydrophobic/hydrophilic fractions, and geochemical species with the disposal time. The main trace metals, that is, Pb, Ni, Cr Cu, and Zn, were around 0.04–0.52 mg Pb/L, 0.24–1.22 mg Ni/L, 0.12–0.86 mg Cr/L, 0–0.28 mg Cu/L, and 0.58–1.98 mg Zn/L in leachates with the disposal ages of 2–12 years. Pb was found to predominate in suspended solids, with the percentage of 46–100%, and the dominant fraction of Ni and Zn was in dissolved fractions, with the percentage of 28–83 and 34–67%, respectively, while Cr and Cu scattered between suspended solid and dissolved fractions. The predominated geochemical species of Pb, Ni, Zn, and Cu was Me dissolved organic matter and Me CO₃, while Cr was dominated as hydroxide complexations. For the hydrophobic distribution in leachates, the main part of Pb, Ni, Cr, and Cu was presented as hydrophobic fractions, with the percentage of >50%, while Zn predominated in the hydrophilic fractions. The evolution processes of trace metals are dependent on both the metals' nature and the landfill conditions greatly, and the corresponding treatment process should be changed with the landfill ages.

Keywords: Trace metal speciation distribution; Evolution process; Landfill leachate; Different ages

1. Introduction

Landfilling is the last resort for the waste management option, and the by-product of leachate is a headache problem for landfill managers due to the complex components, including the high concentration

of organic matter [1–3], and variations of trace metals. With the implementation of the latest Standard for Pollution Control on the Landfill Site of Municipal Solid Waste (MSW) (GB16889-2008) by China Environmental Agency [4], the leachate discharge standards are stricter, and trace metals in leachate should be considered simultaneously for the leachate treatment processes.

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Trace metals are strongly associated with the organic matters in leachates [4–8], and the corresponding speciation changed greatly when the chemical conditions in landfill, such as redox condition, pH, varied as the disposal time extended. It should be pointed out that the species distributions have important implications on the leachate treatment options and the potential mobility in the surrounding environment [9–13]. Understanding the speciation distribution process would be important for the landfill managers.

Some works have been reported on the trace metal speciation in landfill leachate and its plume [7,8,13], while limited works were available on the variation of trace metal speciation with different ages, especially in East Asia, where the distinct MSW contains more than 60% putrescible waste, such as food waste. Besides, most of works reported were a simulation through the laboratory-scale tests rather than the representative of reality in a real-working landfill, while it is a big task to simulate a laboratory-scale landfill lysimeter with more than 10 years (the minimal operation life for a working landfill in practice). In particular, laboratory test values were found not to correlate well with field values because of a lack of sampling representativeness and the difference of the landfill operation conditions, being of one to two orders of magnitude less than field measurements sometimes [14,15].

To gain insight into the trace metal speciation distribution in landfill leachates, a working landfill with special design in Shanghai (details described in Section 2.1), where refuses have been disposed separately in the cell compartments, was chosen for the field investigation of trace metal speciation. Leachates with different ages were collected and analyzed. The results acquired enable a better understanding of the fate of trace metals in leachates and provide the useful information for the selection of leachate treatment facilities.

2. Materials and methods

2.1. Laogang Refuse Landfill site

Shanghai Laogang Refuse Landfill, with an approximate surface footprint of 6 km², was constructed in 1989 along the shore of the East China Sea and commissioned at the end of 1991. The refuse disposed was collected from the downtown area of Shanghai, with around 7,600 tons of refuse (75% of the total refuses generated in Shanghai) disposed per day. This landfill was divided into 57 cell compartments in the first III phases, where each compartment comprises between 5 and 10 ha area and 4 m filling depth. There is no proper leachate collection system, which leads to

the leachate accumulation beneath old cell compartments. Around 343-ha landfill cell compartments have been closed, with approximately 30 million tons of refuses disposed since 1991.

Leachate samples were collected from the middle of the cell compartments selected based on the random sampling procedure. A crawler excavator (WY160; the max rotary speed is 9 rmp, and the max digging depth and the max radius are 4.5 and 7.38 m, respectively) was applied to remove the cover soil and refuse in the upper layers for 1.8 m (leachate level was about 2 m below the surface), and a stainless steel tube, which had a coarse 10-cm screen at the bottom end to prevent clogging, was then inserted 2 m into the cell compartments approximately. Leachate samples were collected from the tube using a peristaltic pump with a velocity < 100 mL/min, after the turbidity and conductivity values were stable.

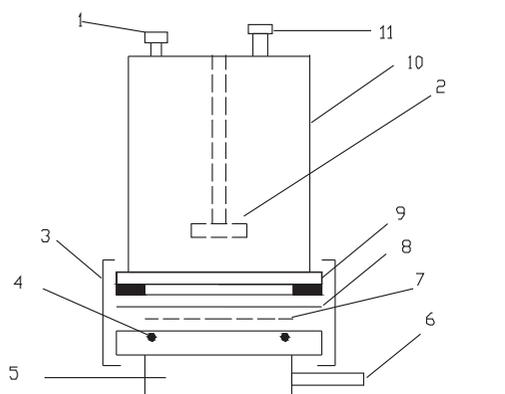
The samples collected were stored in 10-L PE containers, which were washed with 2 m nitric acid and filled with 1% (v/v) suprapur nitric acid in deionized water until use. Leachate samples were stored separately: one kept in 4°C for the analysis of the total solid, alkalinity and so on, and another one was acidified with the HNO₃ additions (10 mL HNO₃ in 1 L leachate samples) for trace metal analysis.

2.2. Size fractions distribution process

Leachate samples were separated into size fractions using a 0.45- μ m membrane and subsequently 1 k-Da MW membrane, which were made of mixed cellulose and polyether sulfone, respectively. Three fractions, that is, suspended solid fraction (the fraction > 0.45 μ m), colloid fraction (1 k-Da MW < the fraction < 0.45 μ m), and dissolved fraction (<1 k-Da MW), were obtained subsequently. The ultrafiltration apparatus used consists of a methyl-methacrylate glass holder with a volume of 300 mL, as shown in Fig. 1. Samples were pressed by a N₂ pressure, with an operation pressure of 0.1 and 0.25 MPa. Before each filtration step, the filtration units were washed with acid and cleaned by flushing with 2% (v/v) HNO₃ and deionized water. The filter was replaced when the flow rate decreased significantly (indicating clogging of the filter membrane).

2.3. Hydrophobic and hydrophilic distribution process

A resin equilibration method described by Leenheer was used to determine the chemical fraction according to its hydrophilic characters [16], and the separation



1, admitting pipe; 2, stirrer; 3, fixator; 4, O-ring seal packing; 5, seat; 6, outlet; 7, flow deflector; 8, Ultra-membrane; 9, sealing plate; 10, ultrafilter cup; 11, feed inlet

Fig. 1. Schematic diagram of SCM type of ultrafilter.

process of hydrophobic/hydrophilic fractions is shown in Fig. 2. Leachate samples were pretreated by the filtration of 0.45- μm membranes. The exchange resin was cleaned with nitric acid and sodium hydroxide and followed with deionized water. Finally, it was adjusted to pH 5 using an acetate buffer prior to use. The fractions of hydrophobic base fraction (Ho-base), hydrophobic acid fraction (Ho-acid), hydrophilic base fraction (Hi-base), hydrophilic acid fraction (Hi-acid), hydrophilic neutral fraction (Hi-neutral), and hydrophobic neutral fraction (Ho-neutral) were obtained in series.

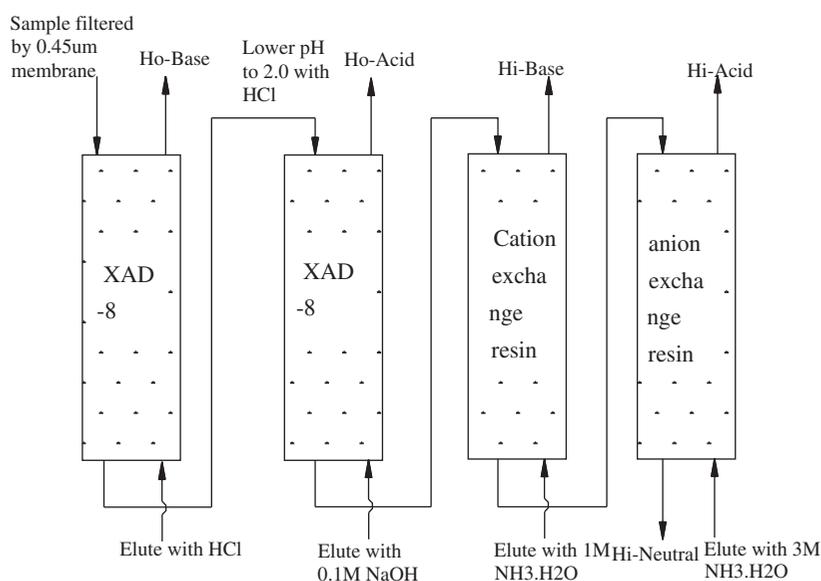


Fig. 2. The separation process of hydrophobic/hydrophilic fractions in leachate sample.

2.4. MINTEQA2

Geochemical speciation models of MINTEQA2 (version 3.0) were used to evaluate the mineralogical fractions of trace metals in leachate (samples filtered by 0.45- μm membrane) [7,17]. Gaussian distribution model was selected, with a defaulted data of GAUSSIAN.DBS for thermodynamic calculation process and ALK.DBS for materials formation. The molar site concentration of dissolved organic matter (DOM) was estimated according to total organic carbon content in leachate samples, with the value of 1,863, 1,267, 440, 303, 277, and 297 mg/L as the disposal time extended. The conductivity value is around 41,500, 40,500, 17,870, 15,030, 10,630, 12,270, 12,080, and 6,380 $\mu\text{S}/\text{cm}$. A temperature of 20°C was used for all the model speciation, since this was the approximate temperature for the experiment.

2.5. Measurement methods

Trace metals were determined by ICP-AES (Inductively Coupled Plasma Atomic Emission Spectrometer, Optima 2100DV, PerkinElmer, USA), after digested by microwave-assisted acid digestion with a combined acid ($\text{HNO}_3(65\%):\text{H}_2\text{O}_2(30\%): \text{leachate} = 7:1:50 \text{ mL}$) using Microwave Extraction/Digestion System ETHOSE (Micesione, s.r.l, Italy) due to the high content of total solid. The other parameters, such as chemical oxygen demands (COD), ammonia nitrogen ($\text{NH}_4^+\text{-N}$), total nitrogen (TN), and total phosphate (TP), were determined by the standard methods

recommended by USEPA, that is, COD was determined by the method #: 410.3.

3. Results

3.1. Fundamental characterization of leachates with different ages

Leachates properties with different ages were determined in terms of COD, TN, TP, pH, and DOM, etc. [1]. COD in leachate decreased from 7,200 mg/L in the 2nd year to 695 mg/L in the 12th year. TN and $\text{NH}_4^+\text{-N}$ were 4,368 and 4,251 mg/L in leachate with 2 years, and the corresponding TP and orthophosphate were 34.9 and 34.3 mg/L. All of them decreased considerably as the disposal time extended. For alkalinity, the concentration decreased significantly in the beginning, from 18,162 to 1,714 mg/L, and then kept about 1,700 mg/L after 12 years, suggesting that the source of bicarbonate (e.g. CO_2) was not depleted in leachate after disposal for 12 years. pH values showed a nearly constant trend during the 12 year monitoring period, with a variation of 7.34–8.15, meaning that all leachate samples were in the methanogenic period.

The trace metal contents are shown in Table 1. Pb content in leachates with different ages was in a range of 0.04–0.52 mg/L. Ni fluctuated between 0.24 and 1.22 mg/L as the disposal time extended, and Zn was in 0.58–1.98 mg/L. Cr and Cu were around 0.12–0.86 and 0.00–0.28 mg/L, respectively. Pb, Cr, and Cu in leachates decreased greatly as the disposal time extended, and Cd in all leachate samples was under the determination limits.

3.2. Geochemical speciation of trace metals

Geochemical speciation is related to the form of the trace metal speciation such as the free ions, neutral, and charged complexes. The speciation of Cr, Cu, Pb, Cd, and Zn is shown in Fig. 3.

Cr(OH)^{2+} content increased from 48.8 to 83.9% as the disposal time increased from the 2nd year to the 12th year, while that of $\text{Cr(OH)}_3(\text{aq})$ decreased from 17.4 to 5.7%, and Cr(OH)^{2+} is the main species in leachate, assuming that Cr existed as Cr(III) in the reduced environment of landfill [13]. Cu in leachate was presented as the complexation of $\text{Cu(CO}_3)_2^{2-}$, Cu DOM, and $\text{CuCO}_3(\text{aq})$. $\text{Cu(CO}_3)_2^{2-}$ was the predominated form, while it dropped from 89.8% in the initial stages to 23.1% latter. Pb predominated as the forms of $\text{Pb(CO}_3)_2^{2-}$, Pb DOM and $\text{PbCO}_3(\text{aq})$, and $\text{Pb(CO}_3)_2^{2-}$ decreased from 81.8 to 5.8%, while $\text{PbCO}_3(\text{aq})$ increased from 4.9 to 37.5% as the disposal time extended. Carbonates were suggested to be the key phase for binding trace metals and metalloids in leachates either by adsorption or by incorporation [13,18,19].

A diverse pattern of Zn speciation, that is, Zn^{2+} , ZnHCO_3^+ , $\text{ZnCO}_3(\text{aq})$, and Zn DOM, was observed in all leachate samples. Zn DOM decreased from 70.3 to 27.8% as the disposal time extended, while the percentage of Zn^{2+} and ZnHCO_3^+ increased from 1.1 to 19.4% and 3.1 to 17.9%, respectively, due to the decrease of carbonate and DOM content in leachate. $\text{ZnCO}_3(\text{aq})$ maintained around 30% in the entire test period. The content of NiHCO_3 increased from 10.1 to 50.3%, while that of Ni DOM decreased from 36.9 to 6.3% as the disposal time extended, meaning that Ni prefers to complex with DOM when more DOM presented in the leachate samples in the initial period, and the dominant speciation changed from Ni DOM in the initial phase to NiHCO_3 after 5-year disposal.

3.3. Trace metal distribution in different molecular weight range

Trace metal distributions are shown in Fig. 4. It was found that Pb predominated in suspended solids with the percentage range of 46.3–100%, similar results were found by Øygard et al. [18], meaning that Pb

Table 1

Relationship between trace metal contents in leachates and the different ages (Units: mg/L) (mean \pm S.E.)

Disposal ages	Pb	Cd	Cr	Cu	Zn	Ni
2	0.14 \pm 0.01	B.D. ^a	0.86 \pm 0.07	0.16 \pm 0.03	1.60 \pm 0.10	0.70 \pm 0.05
3	0.52 \pm 0.01	B.D. ^a	0.54 \pm 0.04	0.22 \pm 0.02	2.40 \pm 0.15	0.82 \pm 0.05
4	0.48 \pm 0.01	B.D. ^a	0.16 \pm 0.04	0.14 \pm 0.03	1.08 \pm 0.05	0.48 \pm 0.01
6	0.08 \pm 0.02	B.D. ^a	0.22 \pm 0.01	0.10 \pm 0.04	1.28 \pm 0.05	0.40 \pm 0.01
7	0.04 \pm 0.02	B.D. ^a	0.28 \pm 0.03	0.16 \pm 0.03	1.10 \pm 0.1	0.30 \pm 0.03
9	0.08 \pm 0.01	B.D. ^a	0.12 \pm 0.03	0.28 \pm 0.03	0.58 \pm 0.03	0.24 \pm 0.03
11	0.06 \pm 0.05	B.D. ^a	0.26 \pm 0.02	B.D.	1.90 \pm 0.05	0.34 \pm 0.02
12	0.05 \pm 0.03	B.D. ^a	0.22 \pm 0.02	B.D.	1.98 \pm 0.07	1.22 \pm 0.02
Discharge standards	0.1	0.01	0.1			

^aB.D. means below the detection limit, the same to the below.

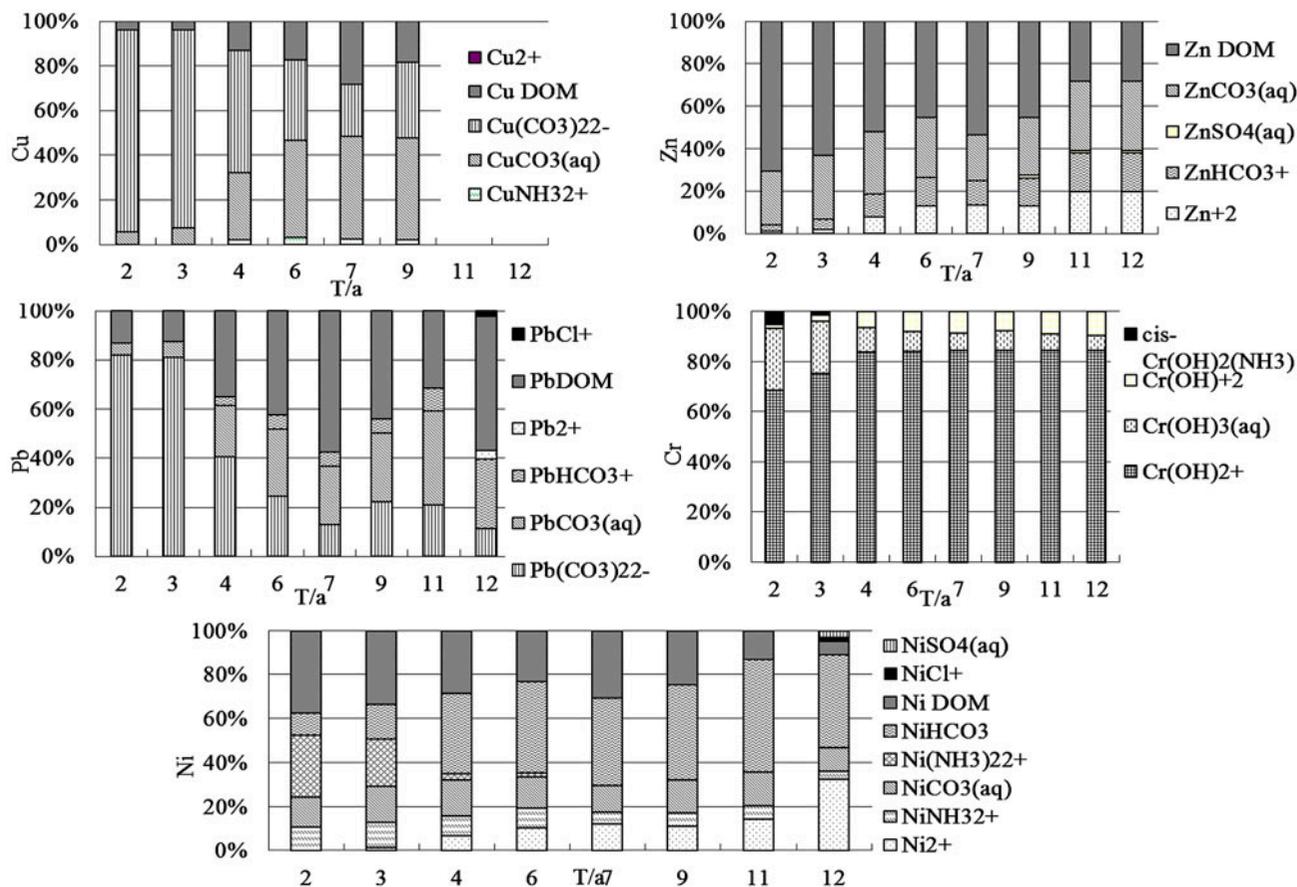


Fig. 3. The speciation distribution process of trace metals in the leachate with different ages.

prefers to combine with the large size particle in leachate. Cu in suspended solid increased from 12.5 to 85.7% as the disposal time extended and that in dissolved fractions was in the range of 14.2–50.0%, while only 5–30% of Cu was found in the fraction $<0.45 \mu\text{m}$ according to the result by Øygaard et al. [18].

Ni in the dissolved fraction dropped from 82.9 to 27.8%, while that in suspended solid increased from 7.3 to 65.5% as the disposal time extended from 2 to 12 years. The size distribution of Cr varied greatly, and about 30–50% of Cr was found in the suspended fractions, while less than 30% of Cr was in the colloid fractions. Zn in the dissolved fractions was around 4–68% and that in the colloid fraction and suspended solid was dispersed in 20.7–34.5% and 9.1–42.5%, respectively. The results are something different from the reports available [7]. The redox conditions and the aged refuse property in Laogang Refuse Landfill might lead to the different distribution process, since higher percentage of organic matter (especially for the food waste) presented in this landfill, compared to those in EU and USA.

3.4. Trace metals in hydrophobic and hydrophilic distribution

Ion-exchange resin technique was applied to determine the chemical speciation, and trace metal distribution process is shown in Fig. 5. It was found that Zn was mainly in the hydrophilic phase, and the distribution of Zn in the Hi-acid, Hi-neutral, and Hi-base occupied around 20–55%, <20%, and 10–48%, respectively. Zn in Ho-acid, Ho-base and Ho-neutral accounted for about 2–21.7%, 2–23.8%, and 5–15%, respectively. Pb was dominated in Ho-base fraction, with the percentage of 53–100%, and that in the Ho-acid was in the range of 12–29.5%.

Cr in hydrophobic fractions was higher than that in hydrophilic fractions, and the percentage of Cr in the Ho-acid, Ho-base, and Ho-neutral was in the range of 9–30%, around 20%, and 6.9–36%, respectively, while that in Hi-acid, Hi-base, and Hi-neutral was about 10–15%, 10–15%, and 3.8–30%, respectively. Cu content was mainly presented in the hydrophobic fractions, where Ho-acid and Ho-base fractions were 44.4–55.4% and 20–40%, respectively. Cu in Ho-neutral

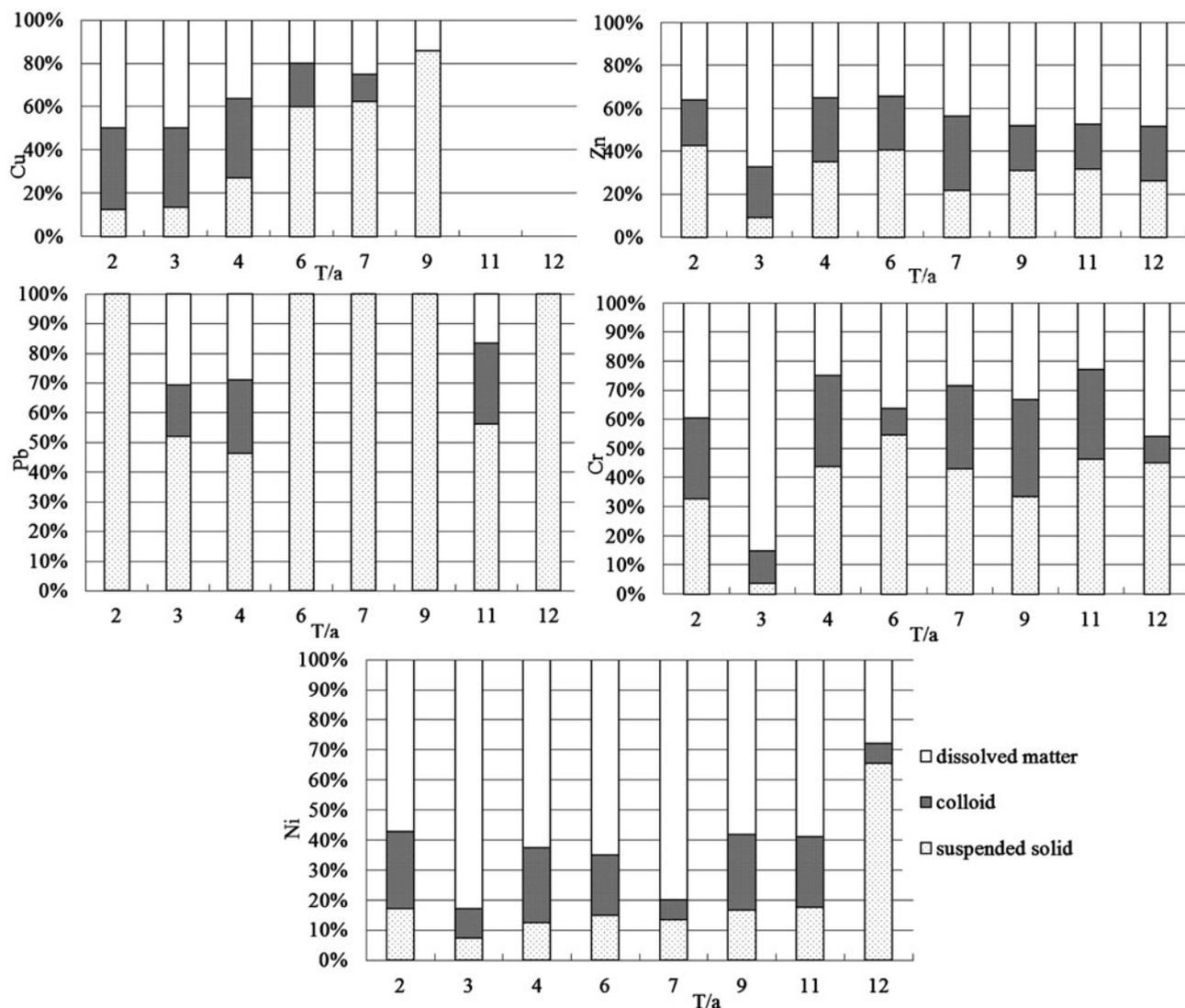


Fig. 4. The distribution of trace metals in the different fractions in leachate with different ages.

decreased from 30% to below the determination limits as the disposal time extended. More than 80% of Ni presented in the hydrophobic fraction, and only 0.3–79% and 6–60% Ni complexed in Ho-acid and Ho-base fraction. The percentage of Ni in Ho-neutral was kept around 20% in all leachate samples and that in Hi-neutral fraction was below 10%.

4. Discussion

Trace metals in MSW were mitigated into leachate through the leaching process and biodegradation processes during the landfill stabilization process. There are two reasons caused the decrease of trace metals in leachates as the disposal time extended. The first one is

the decrease of metals contents in MSW due to the update of the cooking burning system from coal to the coal gas and natural gas in 1990s in Shanghai. More food wastes were included in MSW with the improvement of the living conditions simultaneously. The leaching process was another cause, since the cover system in the old cell compartments was poor, and more rain water entered landfill body in such a rain-rich area. However, it was found that some trace metals were still presented in leachates after disposal for 12 years, since parts of trace metals in MSW were leaching out in a slow rate, especially for those presented in the construction and demolition wastes, with the occupied percentage of 5% in Laogang Refuse Landfill [1,19]. Sorption by soil in landfill and precipitation as sulfide complex were

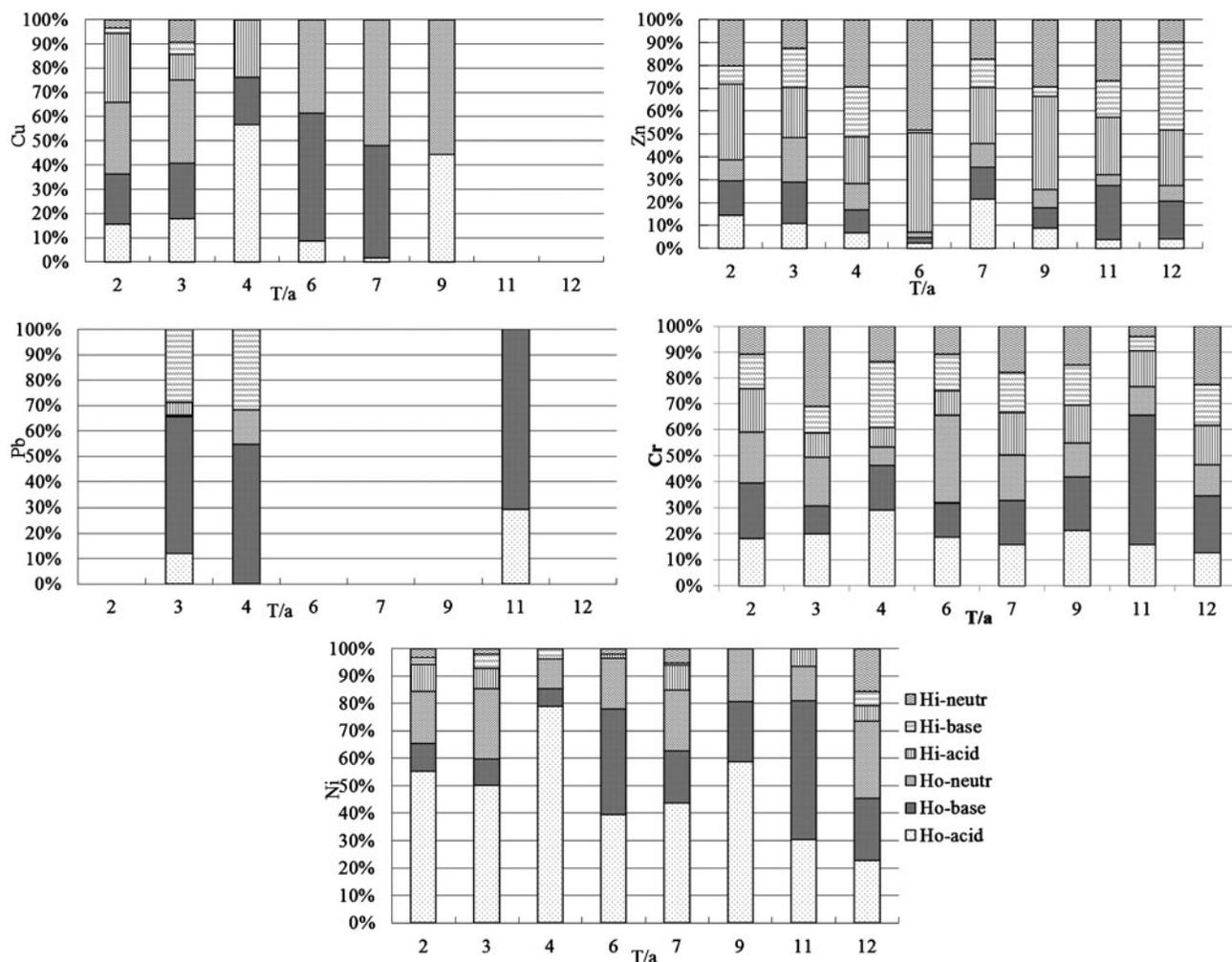


Fig.5. The fractionation distribution of trace metals in DOM of landfill with different ages according to the hydrophobic character.

believed to be the other two reasons for immobilization of trace metals and the subsequent low leachate concentrations in all leachate samples.

Generally, a gradual decline was expected as the dissolved fractions and available free trace metals were washed out from MSW, and the behaviors of trace metals were related to the complexed metal ions, that is, $Me\ DOM$, $Me(CO_3)_x$, and $Me(OH)_x$, which were also influenced by the leachate property greatly. Landfill leachate changed greatly as the disposal time extended, especially for pH and ORP, and thus, the binding forms of trace metals varied. Our results showed that the free ion compositions in leachate samples were less than 20% of the total metal concentration, which was in accordance with the results reported by Christensen et al. [20]. Cr was mainly in hydroxide complexes, with the value of 48.8–83.9%. 36.9% of Ni was complexed with DOM in the initial

time after disposal, and then, carbonate complexes predominated as $NiHCO_3^+$ after 10 years. Cu and Pb were predominated with carbonate complexes, with the value of 23.1–89.8% and 5.8–81.8%, respectively. 27.8–70.3% of Zn was complexed with DOM. Therefore, the distribution of trace metals between these different species would partly depend on the nature of each metal and different chemical parameter in landfill, such as pH, ORP, primary cations and anions concentration, and the type and amounts of organic and colloidal material in leachate.

Moreover, a major proportion of Cu and Pb concentration in leachates were bound to the suspended solid fraction, indicating that these metals were present in their less bio-toxicity and mobile form [18,21]. Ni and Zn were complexed in dissolved fractions, because the main speciation in leachate samples was $Me\ DOM$, and thus, these metals were considerably more mobile

than the metals bounded to suspended solids and colloidal matter in leachate, such as Cu and Pb. For the hydrophobic character distribution, Cu, Pb, and Ni were mainly presented as hydrophobic fractions, while Zn was predominated in hydrophilic fractions.

5. Conclusions

The distribution process of trace metal speciation was investigated in leachates with different ages from Laogang Refuse Landfill. Trace metal species varied and were dependent on the individual trace metal characteristics and the disposal ages greatly. Free metal ions were much less essential speciation. Pb predominated in the suspended solids, with the percentage of 46–100%, and the dominant fraction of Ni and Zn was in the dissolved fractions, while Cr and Cu scattered between suspended solid and dissolved fractions. More trace metals presented in suspended solid as the disposal time extended. For the hydrophobic distribution in leachates, the main fractions of Pb, Ni, Cr, and Cu, were presented as hydrophobic fraction, with the percentage of >50%, while Zn predominated in hydrophobic fraction.

Acknowledgments

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