



“Gheorghe Asachi” Technical University of Iasi, Romania



## FRACTIONATION OF ZINC IN MUNICIPAL SOLID WASTE LANDFILL LEACHATE: EFFECT OF LEACHATE RECIRCULATION

Jun Yao<sup>1,2</sup>, Wenbing Li<sup>3</sup>, Qingna Kong<sup>1</sup>, Huayue Zhu<sup>1</sup>, Yuyang Long<sup>4</sup>, Dongsheng Shen<sup>4\*</sup>

<sup>1</sup>College of Life Science, Taizhou University, Linhai 317000, China

<sup>2</sup>Department of Environmental Engineering, Zhejiang University, Hangzhou 310029, China

<sup>3</sup>College of Life and Environmental Sciences, Hangzhou Normal University, Hangzhou 310036, China

<sup>4</sup>Zhejiang Provincial Key Laboratory of Solid Waste Treatment and Recycling, College of Environmental Science and Engineering, Zhejiang Gongshang University, Hangzhou, 310018

### Abstract

Two sets of simulated landfill reactors, namely conventional landfill (CL) and leachate recirculated landfill (RL) were operated to investigate the effect of leachate recirculation on the fractionation of Zn in municipal solid waste (MSW) landfill leachate. Zn in landfill leachate was fractionated into three fractionations: particulate and colloidal matter >0.45  $\mu\text{m}$ , non-labile complexes <0.45  $\mu\text{m}$  and free cations/labile complexes <0.45  $\mu\text{m}$ . The result showed that the recirculation could decrease the total concentration of Zn in the leachate. The leached Zn was mainly present as free cations/labile complex and non-labile complexes at the beginning of the study. However, it was mainly present as particulate and colloidal matter >0.45  $\mu\text{m}$  and non-labile complexes at the end of the study, for both of landfills. The recirculation of leachate could facilitate the transformation of the fractionation of Zn. Moreover, large amounts of Zn mainly present as free cations/labile complexes and non-labile complexes were leached out from CL, while only small amounts of Zn mainly present as was particulate and colloidal matter >0.45  $\mu\text{m}$  were leached out from RL all through the study. It suggested the leachate recirculation not only reduced total leaching amounts of Zn, but also decreased the bioavailability and toxicity of the leached Zn.

*Key words:* fractionation, landfill, leachate recirculation, Zn

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### 1. Introduction

Landfill is still the most widely used technology for the disposal of municipal solid waste (MSW), especially in developing countries. In China, more than 90% of MSW is disposed by landfill (Long et al., 2010b; Qi et al., 2013). However, landfill is connected with a risk of pollution (Aliakari-Beidokhti et al., 2017). Several case studies have revealed the underground water and soil pollution by the landfill leachate surrounding the landfill sites (Butt et al., 2014; Kasassi et al., 2008; Rusu et al., 2017; Zawierucha et al., 2013). Heavy metals, due to their persistence and biotoxicity, are of particular concern among the various pollutants in the leachate (Fauziah

et al., 2013; Salem et al., 2014). Some studies argued that the risk of heavy metals discharge by the leachate is questionable due to the high retention of metals by the landfill, which resulted in the relatively low migration of heavy metals (Øygaard et al., 2004). However, a lot of case studies proved the heavy metal pollution by the leachate (Jensen et al., 1999; Prechthai et al., 2008). The difference might result from the variance of characteristics of the disposed MSW, as well as the conformation and operation modes of the landfill. The heavy metal issue is especially serious in China as a large portion of MSW is the kitchen food waste, which contains high level of organic matter. The organic matter will be degraded to the organic acid during the landfill operation and form

\* Author to whom all correspondence should be addressed: e-mail: yizaghi@126.com; Phone:+86 57186971156; Fax: +86 57186971156

the relatively acidic condition. In some case studies, the pH of landfill leachate can be lower than 5.0. Such an acidic condition can greatly facilitate the mobility of heavy metals and increase the risk of pollution. Among the heavy metals in the leachate, Zn is of particular interest. Firstly, the content of Zn is relatively high in MSW compared to other heavy metals (Yao et al., 2013; Prechthai et al., 2008). Secondly, it has a high leaching potential according to the previous studies (Long et al., 2010a, 2010b). Furthermore, Zn has a certain biotoxicity to the surrounding ecosystem (Wang et al., 2010; Zhang et al., 2017). It was thus selected for the discussion herein.

Nowadays, the concentration of Zn in the leachate is commonly determined for the surveillance purpose as the total content. This method quantifies the total discharge from the landfills, but does not give a real indication of the environmental load caused by Zn (Wu et al., 2011). The physico-chemical form of Zn is a critical factor influencing its mobility, bioavailability and toxicity. For example, Zn present as free ion or labile complexes (which can easily dissociate) is generally more harmful than Zn present as non-labile complexes, since Zn in free ion form is both more mobile and easily absorbed into the tissue of marine organisms (Øygaard et al., 2007). Therefore, it is crucial to specify the physico-chemical form of Zn in the leachate. Generally, there are two approaches for the speciation of metals in aquatic environment: physical method and chemical method. The physical method was related to the size/molecular weights of the species. The screen filtration is the most widely used physical method. Pore size of 0.45  $\mu\text{m}$  is often suggested in standard protocols for water samples as the cutoff diameter between dissolved and particulate matter (Baun and Christensen, 2004; Matura et al., 2010). The chemical fractionation is related to the chemical form of the species like free ions and complexes. The ion-exchange resin technique is the most used method to fractionate the dissolved species. It is often used to divide the metal complexes by their stability. In this study, the screen filtration technique and ion-exchange resin technique were combined used to fractionate Zn in the landfill leachate into three fractionations: particulate and colloidal matter >0.45  $\mu\text{m}$ , non-labile complexes <0.45  $\mu\text{m}$  and free cations/labile complexes <0.45  $\mu\text{m}$ .

The conventional landfill usually needs a long-term maintenance due to the slow rates of decomposition. Bioreactor landfill can enhance the decomposition process with the help of leachate recirculation (Sun et al., 2014; Kong et al., 2016). The leachate recirculation has become a popular operation mode for landfills. Due to the recirculation, the leachate characteristics, such as pH, the concentration of volatile fatty acid (VFA), humic substance and sulfide etc. are changed. Therefore, the fractionation of Zn in the leachate can be altered. To assess the environmental impact of Zn discharge by the leachate from the bioreactor landfill, it is necessary to investigate the effect of leachate recirculation on the

fractionation of Zn in the leachate. Several studies have been done to characterize the fractionation of Zn in the leachate from practical landfill sites (Baun and Christensen, 2004; Jensen and Christensen, 1999; Li et al., 2009). However, to our best knowledge, the effect of leachate recirculation on the transformation of the fractionation of Zn in the leachate was rarely reported.

In this study, two landfills namely conventional landfill (CL) and leachate recirculated landfill (RL) were set up. The effect of the leachate recirculation on the transformation of fractionation of Zn in the leachate was presented. The results acquired enable a better understanding of the bioavailability and environmental impact of Zn discharge from landfills with different operation modes.

## 2. Materials and methods

### 2.1. Experimental set-up

Two sets of simulated landfill reactors were set up. One was the conventional landfill with leachate single leaching, while another was bioreactor landfill with leachate recirculation. Each of the reactors was 0.3 m in diameter and 1.0 m in height, with a total work volume of 65 L. Each landfill reactor was equipped with two outlet ports. The outlet at the top lid was used for the gas exporting, while the outlet at the bottom was used for leachate drainage and sampling. Besides, an inlet ports was equipped at the top lid of RL, which was used for the leachate recirculation. A 50 mm thick layer of gravel was placed at the bottom of each landfill to simulate a leachate collection system, and to prevent clogging of the leachate withdrawal outlets. MSW was loaded and compacted using the shovel and sledgehammer. The second gravel layer was placed on the top of the MSW to simulate intermediate cover and top drainage layer, and further to provide even distribution of the recirculated leachate. The nominal size of the gravel used for both filter layers range from 10 to 40 mm. Finally, the two landfills were sealed with gasket and silicone. Fig. 1 shows the schematic of the simulated landfill systems.

### 2.2. MSW

MSW used in this experiment was collected from the Kaixuan transport station of Hangzhou, Zhejiang, East China. In order to obtain a representative sample of solid waste that normally goes to the landfill, MSW was collected continuously at different time of the day. The moisture content of MSW was 64% (wet basis). The average wet density of the MSW compacted in the bioreactor landfills was 600  $\text{kg}/\text{m}^3$ . Larger particles of the collected MSW were all shredded into 10mm by 30 mm approximately. MSW were thoroughly mixed prior to the loading of bioreactor landfills. The main components and Zn contents of MSW utilized in this study is presented in Table 1.

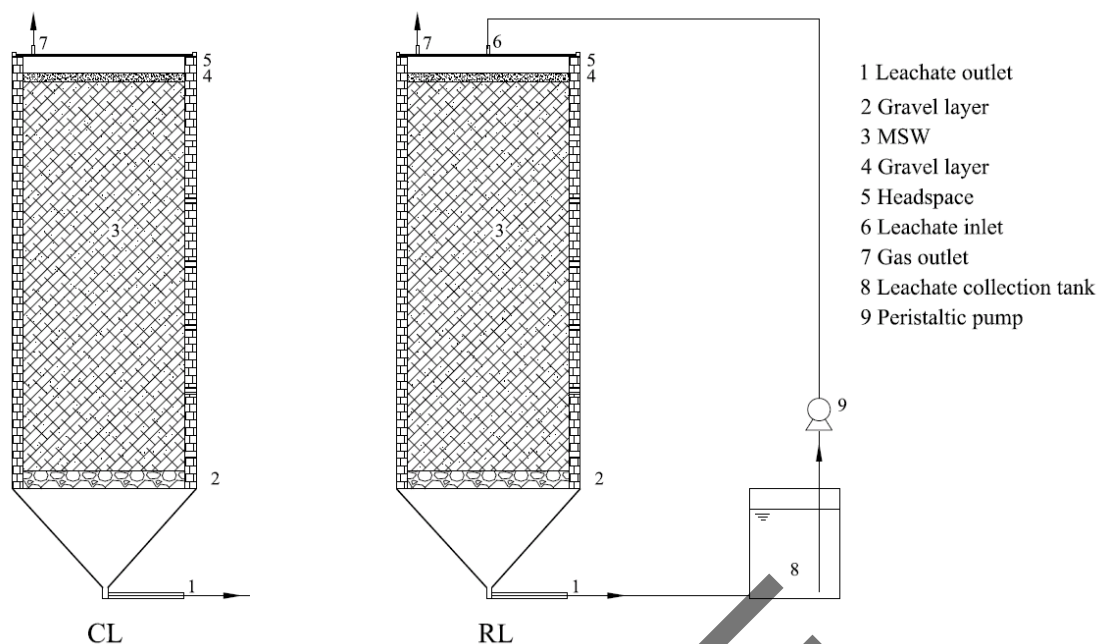


Fig. 1. Schematic of simulated landfill systems

Table 1. Components and Zn contents of experimental MSW

Components	Food waste	Plastic	Paper	Textile	Dust	Ceramic	Metal	Timber	residue
W/W, %	45.5	8.5	9.5	0.1	5.2	5.8	0.1	0.7	24.5
Content (mg/kg)	1540.5	774.5	929.1	169.4	707.6	761.7	NA	834.0	846.3

NA, not available

It showed that the food waste, the largest fraction of MSW, had the highest Zn content, which exceeded 1500 mg/kg. Besides, the content of Zn in the plastic, paper, dust, ceramic, timber and residue fraction exceeded 700 mg/kg. The high content of Zn in the MSW could result in the high concentration of Zn in the landfill leachate.

### 2.3. Operation of simulated landfills

Firstly, MSW was loaded and compacted using the shovel and sledgehammer. Then, the moisture content of refuse was adjusted to 75% (Benson et al., 2007) by adding tap water to the simulated landfill after loading. Leachate was collected and stored in the leachate collection tank. The leachate of CL was discarded without further treatment, while the leachate of RL was continuously recirculated using peristaltic pumps with a flow rate of 0.01 L/min every day.

### 2.4. Analyses

Leachate was sampled every week from the leachate outlet ports (~100 mL). In order to keep the volume equilibrium of leachate from RL before recirculation, a same volume of tap water was added back into the leachate after sampling.

Leachate samples were analyzed for volume (V), pH, dissolved organic carbon (DOC), volatile fatty acid (VFA), sulfide, sulfate, humic substance and Zn concentration. Humic substance was analyzed

according to He et al. (2006). Zn concentration was analyzed using atomic absorption spectrophotometer (Shimadzu A-650). Other analyses were performed in accordance with Standard Methods (APHA, 1999).

The fractionation of Zn in the leachate was determined according to the size charge fractionation (SCF) procedure suggested by Driscoll (1984). In the procedure, the leachate was firstly filtrated by a 0.45 µm screen filter, in which the fraction particulate and colloidal matter >0.45 µm was removed from the leachate. Then, the filtered leachate was passed through a sulphonic acid cation exchange resin, in which the Zn presented as free cations/labile complexes <0.45 µm was adsorbed on the cation exchange resin. Zn concentration in the raw leachate, filtered leachate and cation exchanged leachate was analyzed by the atomic absorption spectrophotometer. The fraction particulate and colloidal matter >0.45 µm was calculated by the difference between the Zn concentration in the raw and filtered leachate. The fraction free cations/labile complexes was calculated by the difference between the Zn concentration in the filtered leachate and cation exchanged leachate.

## 3. Results and discussions

### 3.1. Total concentration

The total concentration of Zn in the leachate from CL and RL is exhibited in Fig. 2. It showed a decreasing trend with the proceeding of the study for

both of landfills, which ranged from 1.9 to 21.0 mg/L for RL and 3.4 to 19.0 mg/L for CL. This level was at the moderate level of the range reported by the previous literatures (Baun and Christensen, 2004; Christensen et al., 2001; Kim et al., 2014). The initial concentration of Zn in RL leachate was high, which was 21.0 mg/L. It decreased sharply to 9.7 mg/L since the first time of leachate recirculation. It further decreased to 1.9-5.6 mg/L after day 15. For CL, the total concentration of Zn in the leachate decreased from 19.0 to 7.9 mg/L after 4 months. It kept at the level of 3.5-6.8 mg/L since then. The decreasing of Zn concentration in the leachate of RL and CL was generally corresponding with the variation of pH (Fig. 3). As is known, the solubility of Zn decreases with the increase of pH. The initial pH of the leachate was 4.99 for RL and 4.96 for CL, which resulted in the high level of Zn in the leachate.

With the proceeding of the study, the leachate pH increased. As a result, the concentration of Zn in the leachate was significantly decreased.

It should be noted that although the concentration of Zn decreased with the proceeding of the study, it still exceeded the limited standard issued in the ‘integrated waste water discharge standard’ (2.0 mg/L) (State Environmental Protection Administration of China, 1996) all through the study for both of landfills, indicating a great potential risk to the surrounding ecosystem. Compared to CL, the concentration of Zn in the leachate of RL was relatively low. For RL, the leached Zn was brought back to the landfill by the recirculation. Some of the Zn present as the particulate matter in the recirculated leachate could be retained by the refuse due to the action of filtration (Baun and Christensen, 2004; Wu et al., 2011).

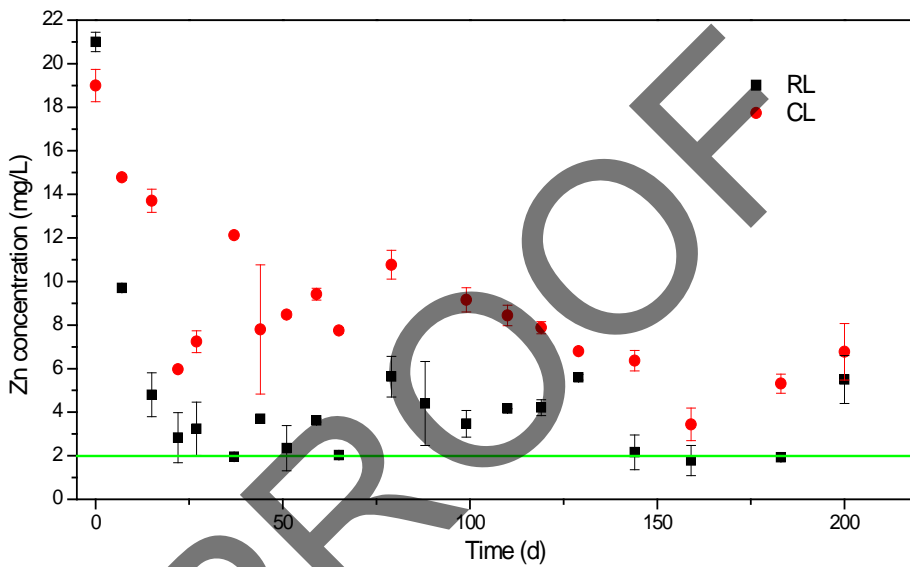


Fig. 2. Total concentration of Zn in leachate of simulated landfills (The linear line indicates the permitted concentration in the Chinese National Standards (State Environmental Protection Administration of China, 1996))

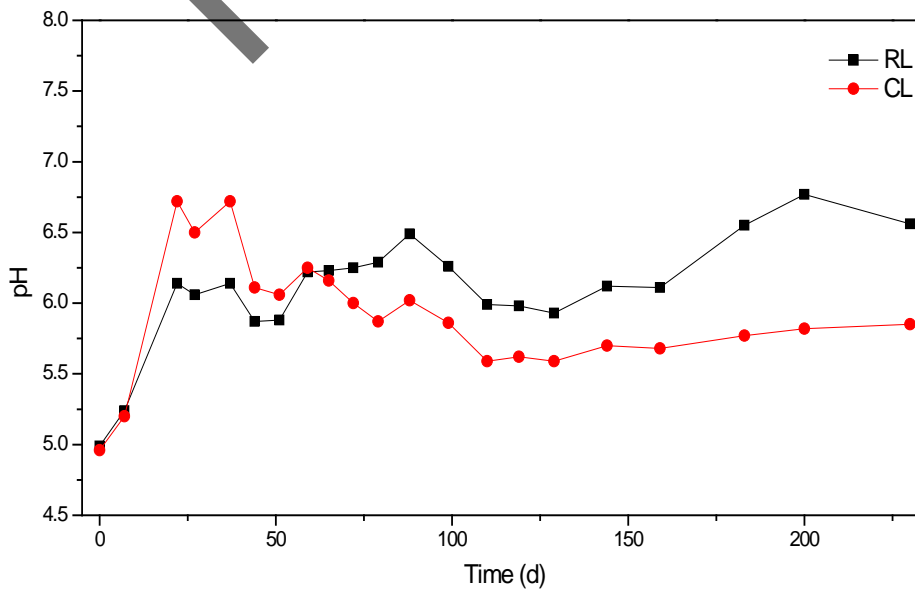
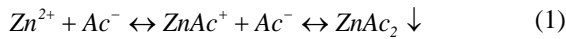


Fig. 3. Leachate pH of simulated landfills

The dissolved Zn in leachate could be immobilized by the VFA (Eq. 1), took Ac as example) (Long et al., 2009; Long et al., 2010a), S<sup>2-</sup> (Eq. 2), and humic substance (Eq. 3), took fulvic acid (FA) as example) etc. in the landfill. It was also possible that a part of Zn was adsorbed by the refuse (Suna et al., 2005). Besides, the recirculation of leachate could facilitate the degradation of organic acid and accelerate the stabilization process of the landfill. This phenomenon was reflected by the evolution of pH (Fig. 3). The pH of RL leachate reached 6.5 on day 230, while it was 5.9 for the leachate of CL. The high pH of the RL leachate might be another reason for its low Zn concentration (Yao et al., 2017).



### 3.2. Size charge fractionation

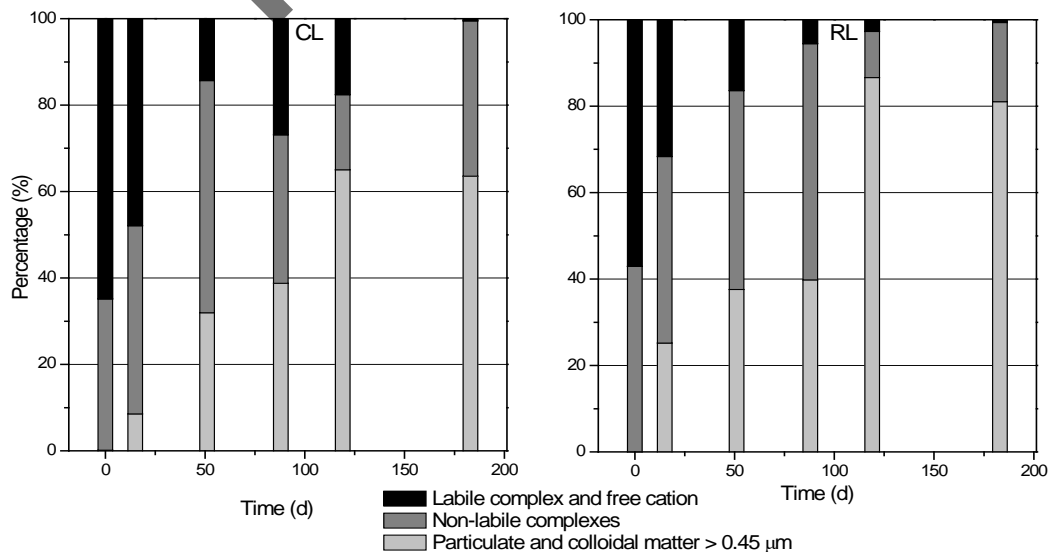
The relative deviation of the Zn concentration in the filtered leachate and cation exchanged leachate is shown in Table 2. The relative deviation was low, which indicated the reliability of the SCF procedure. With these source data, the variation of fractionation of Zn with the proceeding of the study is calculated, as shown in Fig. 4. Zn was mainly present as the labile complexes and free cations at the beginning of the study. It accounted for 57.2% and 64.9% in the leachate of RL and CL, respectively. However, it decreased consistently to 0.6% for RL and 0.4% for CL at the end of the study. It meant most of the Zn present as labile complexes and free cation was transformed to other fractions. Similar results were also found by Papini et al. (2001) and Ward et al. (2005), which suggested the binding capacity of

leachate increased with the proceeding of the study. The non-labile complexes was another major fraction at the beginning of the study, which accounted for 42.9% for RL and 35.0% for CL. Different from the fraction free cations/labile complexes < 0.45 μm, it increased to 54.7% on day 90 for RL, and 53.7% for CL on day 50. Afterwards, the percentage of the fraction non-labile complexes decreased to 18.3% for RL and 36.0% for CL. Interestingly, the concentration of DOC and VFA showed an increasing at the initial stage of landfills (Table 3). This result indicated that some of Zn present as the free cation was combined with the moderate or small organic molecule at the initial stage of the landfill, forming the fraction non-labile complexes. However, the fraction non-labile complexes was further transformed to the fraction particulate or colloidal matter > 0.45 μm with the proceeding of the study.

The fraction particulate and colloidal matter > 0.45 μm increased consistently all through the study for both RL and CL. It increased from 0 to 81.0% for RL and 0.1 to 63.5% for CL, suggesting Zn was mainly present as particulate and colloidal matter > 0.45 μm in the mature landfill leachate.

**Table 2.** Relative deviation of the Zn concentration in the filtered leachate and cation exchanged leachate (%)

Time (d)	Relative standard deviation (%)	
	Filtered leachate	Cation exchanged leachate
0	5.62	8.62
15	1.56	2.45
27	2.60	0.00
37	0.48	0.00
51	0.46	5.26
79	0.03	10.58
119	11.74	5.36
183	3.40	4.12
230	5.62	8.62



**Fig. 4.** Fractionation distribution of Zn in leachate from simulated landfills (Data were shown as percentage relative to total concentration)

**Table 3.** Concentrations of anions and organic matter in the leachate from simulated landfills (mg/L)

Time (d)	DOC		VFA		Humic substance		Sulfides		Sulfates	
	RL	CL	RL	CL	RL	CL	RL	CL	RL	CL
0	20433	20724	13432	14149	330	218	0.52	0.31	826	735
15	23426	18769	12196	12196	716	457	0.31	1.09	166	171
27	26985	23348	15056	16189	1640	1137	1.42	1.70	479	638
37	21462	18833	13599	15542	1837	1577	1.05	1.48	565	608
51	27668	25455	15757	20938	1413	1241	1.48	1.94	531	570
79	25469	35202	12088	16391	1757	3159	2.01	2.38	573	599
119	17733	26855	15717	23406	2314	3099	2.69	2.24	481	555
183	16740	25400	18584	26341	3054	4233	3.55	4.09	519	771
230	19216	32923	16391	26509	3345	4569	3.59	3.49	315	535

This result corresponded well with the work of Øygard et al. (2007) and Jensen and Christensen (1999), which showed 63-98% of Zn was present as particulate and colloidal matter  $>0.45\mu\text{m}$  in the leachate of the practical landfill sites. Correspondingly, the concentrations of the sulfide and humic substance increased consistently all through the study. ZnS was mainly present as insoluble particulate in the leachate. The humic substance bound Zn, which was mainly comprised by large molecules (He et al., 2006), was also probably present as the colloidal matter  $>0.45\mu\text{m}$ . It was thought Zn present as labile complexes, free cations and non-labile complexes were gradually bound with the  $\text{S}^{2-}$  or humic substance, and transformed to the fraction particulate and colloidal matter  $>0.45\mu\text{m}$ . This result was also proved by the simulation result of Visual MINTEQ model, which showed the proportion of ZnS and fulvic acid bound Zn increased from 0.1% to 47.4% for CL and 0.1% to 89.2% for RL.

Compared to CL, the fraction free cations/labile complexes  $<0.45\mu\text{m}$  decreased more sharply for the leachate of RL. The percentage of the fraction free cations/labile complexes  $<0.45\mu\text{m}$  decreased to 5.5% after day 83 for RL, while it took 119 days to decrease the fraction to 17.9% for CL. Besides, more Zn was present as the particulate and colloidal matter  $>0.45\mu\text{m}$  in the leachate of RL at the end of the study.

The recirculation of the leachate could provide a better opportunity for the combination of free cation with the various compounds, such as VFA,  $\text{S}^{2-}$ , and humic substance etc. Therefore, the reduction of the fraction free cations/labile complexes  $<0.45\mu\text{m}$  was enhanced. In addition, the pH of RL leachate was relatively high compared to CL, Zn was more likely to form the insoluble inorganic particulate and colloidal matter when the pH was high (Christensen and Christensen, 2000).

### 3.3. Environmental impact implication

To assess the environmental impact of Zn leaching from landfills with different modes, the accumulated amounts of fractionated Zn were evaluated. The leachate of CL was directly discharged without recirculation. The accumulated amounts of Zn leached from CL can be calculated by Eq. (4).

$$M_{Accu} = \sum_i^n V_i \times C_i \quad (4)$$

RL was the simulated bioreactor landfill with leachate recirculation. The accumulated amounts of fractionated Zn leached could be calculated by Eq. (5).

$$M_{Accu} = V_i \times C_i \quad (5)$$

where  $V_i$  is the volume of leachate on day  $i$ , and  $C_i$  was the concentration of Zn of each fraction on day  $i$ .

As shown in Fig. 5, the accumulated amounts of the fractionated Zn increased all along for CL, while it showed a decreasing trend for RL. For CL, the total leached amounts of Zn was 118.0 mg, in which 60.0 mg was present as labile complexes and free cations, 40.0 mg was present as non-labile complexes and 18.0 mg was present as particulate and colloidal matter  $>0.45\mu\text{m}$ . For RL, only 12.9 mg of Zn was leached out, in which 2.7 mg was present as non-labile complexes and 10.2 mg was present as particulate and colloidal matter  $>0.45\mu\text{m}$ . Almost no Zn present as labile complexes and free cations was leached out from RL. It proved that leachate recirculation could immobilize the Zn in the landfill and reduce the total leaching amounts of Zn. The fraction free cations/labile complexes  $<0.45\mu\text{m}$  was of high bioavailability and toxicity, while the fraction non-labile complexes, particulate and colloidal matter  $>0.45\mu\text{m}$  was of low bioavailability and toxicity (Driscoll, 1984). In the term of the environmental protection, the recirculation of leachate not only reduced the total leaching amounts of Zn, but also decreased the toxicity of the leached Zn. From this point of view, the recirculation of leachate could be a positive way to mitigate the environmental load of Zn leached from landfill.

Compared to RL, the Zn leaching from CL should be particularly concerned, especially at the initial stage of the landfill. As at that stage, both the high concentration and mobility of the leached Zn were observed.

## 4. Conclusions

The operation of bioreactor landfill with leachate recirculation could decrease the total concentration of Zn in leachate and reduce leaching of Zn from landfill.

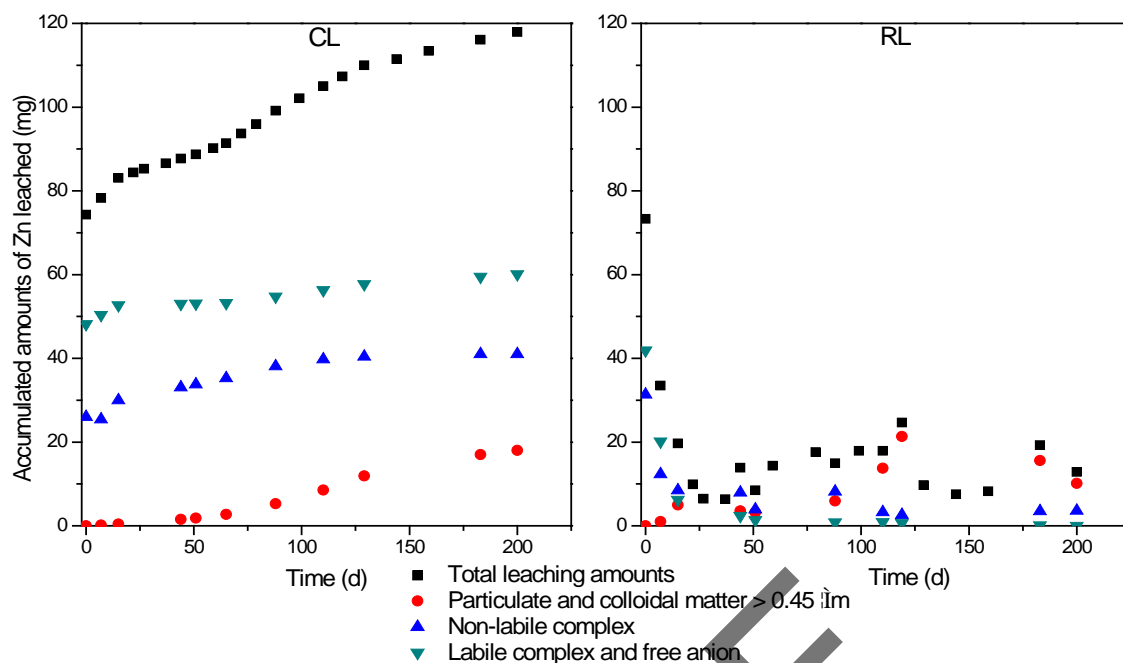


Fig. 5. Fractionation analysis of the leached Zn from simulated landfills

The leached Zn was mainly present as free cations/labile complexes <math><0.45 \mu\text{m}</math> and non-labile complexes at the initial stage of the study, but mainly present as particulate and colloidal matter >math>0.45 \mu\text{m}</math> at the end of the study, for both of CL and RL. The leachate recirculation could facilitate the transformation of the Zn fraction from free cations/labile complexes <math><0.45 \mu\text{m}</math> and non-labile complexes to particulate and colloidal matter >math>0.45 \mu\text{m}</math>. The results showed the operation of bioreactor landfill with leachate recirculation not only reduced total leaching amounts of Zn, but also decreased the bioavailability and toxicity of the leached Zn.

Therefore, it could be a positive way to mitigate the environmental load of the Zn leaching from the landfill. It should be noted that the result of present study mainly concerned about the initial and acidic stage of the landfill. Further study regarding the methanogenic and mature stage is required to fully understand the effect of leachate recirculation on the variation of fractionation of Zn. Besides, except for Zn, other metals which could be found in the leachate should also be investigated.

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