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Migration of Cu, Zn and Cr through municipal solid waste incinerator bottom ash layer in the simulated landfill

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ABSTRACT

Municipal solid waste incinerator (MSWI) bottom ash is used as the cover, intermediate cover and liner in the landfill. In this study, simulated landfills were operated for 507 days to investigate the effect of MSWI bottom ash layer on the migration of Cu, Zn and Cr. All through the study, Cu was greatly released from the MSWI bottom ash layer. Zn was captured by the MSWI bottom ash layer at the initial stage of the landfill. After then, the release of Zn from the MSWI bottom ash layer was observed, corresponding to the decline of the leachate pH. No trapping or release was found for Cr at the initial stage of the landfill. Then, a slight release of Cr from the MSWI bottom ash layer was observed. Over the study, a totally 0.93% of Cu, 0.12% of Zn, and 0.01% of Cr in the MSWI bottom ash layer were released. The released metals were not discharged to the surrounding environment. They were immobilized by the sub-MSW layer. It suggested that the MSWI bottom ash layer could not increase the metal discharge when it was used as the cover or intermediate cover in the landfill.

1. Introduction

Incineration has become more and more important for the treatment of municipal solid waste (MSW) due to the shortage of the urban land as well as the advantages of hygienic control, volume and mass reduction and energy recovery. For instance, in the past ten years, the number of municipal solid waste incinerator (MSWI) facilities increased sharply from 47 to 166 in China, with the treatment capacity rising from 15,000,000 to 158,488,000 kg/d (National Bureau of Statistics of China, 2015). In the incineration process, about 20% of the incinerated MSW was transformed to the solid residue. Among the residue, bottom ash is the main stream, which accounts for approximately 80% (Chimenos et al., 1999). MSWI bottom ash is allowed to be disposed in the MSW landfill sites in several countries and areas, such as China, Japan and Taiwan (Inanc et al., 2007; Lo and Liao, 2007; Youcai et al., 2002). The compacted ash can have a hydraulic conductivity similar to that of clay, which means that it could meet the functional requirements as the cover and liner in the landfill (Chandler et al., 1997; Muhunthan et al., 2004). Therefore, it has been used as the cover, intermediate cover and liner instead of natural clay minerals in the landfill (Su et al., 2013). For example, Travar et al. (2009) used the MSWI ash as the substitute for natural materials in landfill cover construction. Li et al. (2014) used the MSWI bottom ash as the intermediate layer. These studies suggested that the MSWI bottom ash layer could provide several advantages for the landfill construction and operation. For example, it could save the natural mineral material source, fasten the stabilization of the landfill and improve the leachate quality.

MSWI bottom ash is a mineral assemblage, containing a high level of alkaline minerals, adsorption medium and heavy metals (Yao et al., 2010; Yao et al., 2015). The alkaline minerals in the ash result in the high acid neutralization capacity (ANC), which can increase the pH of the leachate vertically flowing through the MSWI bottom ash layer. As the mobility of the heavy metals is dependent on the pH, the migration of them can be changed. The adsorptive medium, including Friedel's salt, kaolin and iron (hydr)-oxides, were reported to have adsorption capacity for the metals. For instance, Dai et al. (2009) pointed out that Friedel's salt could remove 99% of CrO_4^{2-} from the aqueous solution. Adebowale et al. (2005) suggested that kaolin showed a strong affinity for Cu, Zn, Pb and Cd. The ANC and adsorptive medium can help the MSWI bottom ash layer capture the heavy metals from the leachate. However, on the other hand, the high content of heavy metals in the MSWI bottom ash can lead to the heavy metal release, which may aggravate

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the heavy metal pollution (Yao et al., 2010). Several studies have been done to investigate the effect of the disposal of MSWI bottom ash on the heavy metal leaching from the landfill (Inanc et al., 2007; Lo and Liao, 2007; Su et al., 2013). In these studies, MSWI bottom ash was blended with the MSW. The effect of the co-disposal on the final discharge of heavy metals from the landfill was discussed. However, the effect of MSWI bottom ash layer on the migration of heavy metals in the landfill is still poorly investigated. This theme is important as it can provide scientific reference for the control of heavy metal pollution when the MSWI bottom ash layer is used as the cover, intermediate liner and liner in the landfill.

Among the heavy metals, Cu and Zn are presented in high concentration in MSWI bottom ash and MSW (Long et al., 2010; Yao et al., 2014). Cr, as a valence variable metal, is of particular concern as its high toxicity to the surrounding ecosystem (Paquin et al., 2000). They were thus selected for the discussion herein.

The main aim of this study is to reveal the effect of the MSWI bottom ash layer on the migration of heavy metals in the landfill. Two parallel leachate recirculated landfill bioreactors were established and operated for 507 days. MSWI bottom ash was disposed as the landfill layer. The variation of pH, chemical oxygen demand (COD), Cu, Zn and Cr concentrations in the leachates, which were sampled above the MSWI bottom ash layer (L1), beneath the MSWI bottom ash layer (L2) and at the bottom of the landfill (L3), was monitored.

2. Material and methods

2.1. Experimental setup

Two parallel simulated landfill bioreactors with leachate recirculation were set up. The reactor was 287 mm in diameter and 1430 mm in height, with a total working volume of 92 L. Five ports were equipped with the reactor: the two ports at the top were used for gas exporting and leachate recirculation; the two ports at the side were used for sampling the leachate above (L1) and beneath (L2) the MSWI bottom ash layer; the port at the bottom was used for leachate drainage and sampling (L3). A 100 mm thick layer of gravel was placed at the bottom of the landfill to simulate a leachate collection system and to prevent clogging of the leachate withdrawal outlets. The MSWI bottom ash layer was placed between the MSW layers. Another 50 mm thick layer of sand was placed at the top of each landfill to simulate the cover and top drainage layer. The schematic of the simulated landfill system was shown in Fig. 1.

2.2. MSWI bottom ash and MSW

MSWI bottom ash was sampled from the Green Energy MSWI plant in Zhejiang province, East China. The plant consists of two parallel stoker incinerators with an MSW treatment capacity of 650,000 kg/d. The MSWI bottom ash sample had undergone water quenching and magnetic separation before being sampled. A part of the MSWI bottom ash sample was mingled, air-dried and ground into

lesTm(incinerators)TjET/GS1gsBT/F21Tf.021061gsBT/F21Tf.000200-.0002258.248193.675c07.2ST/962eBT008.1057.97c297.8386193.6751Tm(gr

Table 1
Components of the MSW

Components	Food waste	Plastic	Paper	Textile	Dust	Ceramic	Metal	Timber	Residue
W/W, %	44.3	8.2	7.5	0.3	6.2	5.1	0.1	1.7	27.6

Table 2
Physicochemical properties and bulk composition of the MSWI bottom ash sample

Properties or elements	Value
Physi-chemical properties	
Moisture content (%)	1.61
Bulk density (kg/m^3)	1277.6
Loss on ignition (LOI) (%)	2.2
pH	11.2
Acid neutralization capacity ($ANC_{7.5}$)	1.0 H^+ mmol g^{-1}
Element composition (mg kg^{-1})	
Al	40920 ± 1600
Si	223600 ± 4657
Na	9040 ± 178
K	15792 ± 167
Mg	5997 ± 115
Ca	69413 ± 2613
Fe	26008 ± 28
Mn	1246 ± 231
Zn	1922 ± 33
Cu	315 ± 22
Cr	252 ± 42
Cd	7 ± 1
Mo	7 ± 2
As	138 ± 42
Co	21 ± 2
Ni	48 ± 24

Table 3
Fractionation distribution of Cu, Zn and Cr in MSWI bottom ash (mg kg^{-1})

	F1	F2	F3	F4	F5
Cu	3.73	74.61	7.86	141.60	86.79
Zn	0.96	452.55	485.95	79.01	903.53
Cr	3.41	8.67	25.44	4.76	209.42

3. Result and discussion

3.1. Main characteristic of MSWI bottom ash

The result of physicochemical properties and bulk composition analysis is shown in Table 2. The alkali metals, including Na, K, Al, Ca and Mg, were abundant in the MSWI bottom ash sample, which contributed to the high pH (11.2) and acid neutralization capacity (1.0 H^+ mmol g^{-1}). Cu, Zn and Cr were the major trace elements. Their contents in the MSWI bottom ash were 4.1, 14.9 and 25.9 times higher than those in the soils (National Environmental Monitoring Centre of China, 1990), indicating a high potential environmental risk.

The chemical speciation of Cu, Zn and Cr is shown in Table 3. Most of Cu, Zn and Cr were occupied as the organic matter bound fraction (F4) and residual fraction (F5), which were considered as the stable fractions. However, there were still certain amounts of Cu, Zn and Cr occupied as the exchangeable fraction (F1), carbonate bound fraction (F2) and Fe-Mn oxide bound fraction (F3), which were thought to be unstable with the potential leachability. The total amounts of the unstable Cu, Zn and Cr in the MSWI bottom ash were 86.21, 939.46 and 37.52 mg kg^{-1}, respectively. The unstable Cr was less than the unstable Cu and Zn.

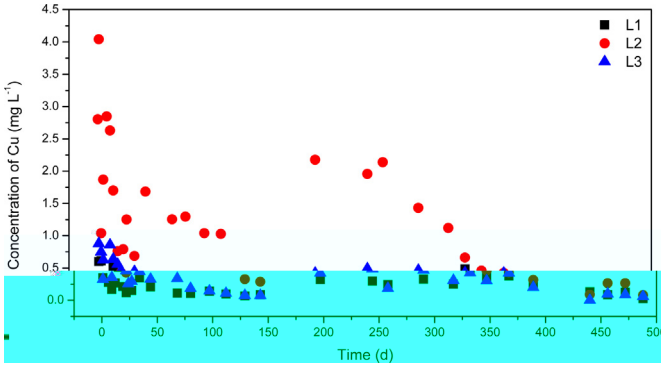

Fig. 2. Variation of Cu concentrations of L1, L2 and L3.

3.2. Migration of Cu, Zn and Cr through the MSWI bottom ash layer

3.2.1. Cu

Generally, the concentrations of Cu in L1, L2 and L3 showed a decreasing trend (Fig. 2), which was consistent with the previous report (Long et al., 2009). The concentration of Cu in the leachate was relatively high at the beginning of the study, which was due to the intensive release of the unstable Cu from the MSW and MSWI bottom ash. Our previous study had showed that the sulfate in the landfill was reduced to sulfide with the time extension, leading to the increase of sulfide level (Long et al., 2010). S^{2-} could react with Cu^{2+} to form CuS (Eq. (1)), a compound of copper with a low solubility. Therefore, the Cu concentration of the leachate was attenuated.

$$Cu^{2+} + S^{2-} \rightarrow CuS \downarrow \quad (1)$$

The Cu concentration of L2 was higher than that of L1 all through the study. The average Cu concentration of L2 was 1.22 mg L^{-1}, while the average Cu concentration of L1 was 0.25 mg/L. It meant that Cu was largely released from the MSWI bottom ash layer. Due to the high acid neutralization capacity (ANC) of MSWI bottom ash (Table 2), the pH of the leachate increased after flowing through the MSWI bottom ash layer (Fig. 3(a)). Notably, the pH of L1 ranged from 4.27 to 6.13 in the first 42 days, while the pH of L2 ranged from 9.07 to 11.76. The increase of pH was thought to be able to immobilize the metal and decrease the Cu concentration of the leachate. However, the Cu concentration of the leachate increased after it flowed through the MSWI bottom ash layer. Cu is reported to have a good affinity with the organic matter. Several researches have showed that the leaching of Cu from MSWI bottom ash could be greatly enhanced by the organic matter (Meima et al., 1999; Olsson et al., 2007; van Zomeren and Comans, 2004). Landfill leachate contained a high level of organic matter. The COD of the leachate exceeded 10000 mg/L all through the study (Fig. 3(b)). The release of Cu from the MSWI bottom ash layer was probably facilitated by the organic matter in the leachate. To approve the assumption, the distribution pattern of Cu in L2 was analyzed by the model Visual MINTEQ. It showed that Cu in L2 was almost 100% bound with the organic matter (Table 4). This result verified the role of organic matter on the Cu release from the MSWI bottom ash layer. It should be noted that the difference of Cu concentrations in L2 and L1 declined with the extension of time. After day 335, the Cu concentration of L1 became close to that of L2, suggesting the release of Cu receded. According

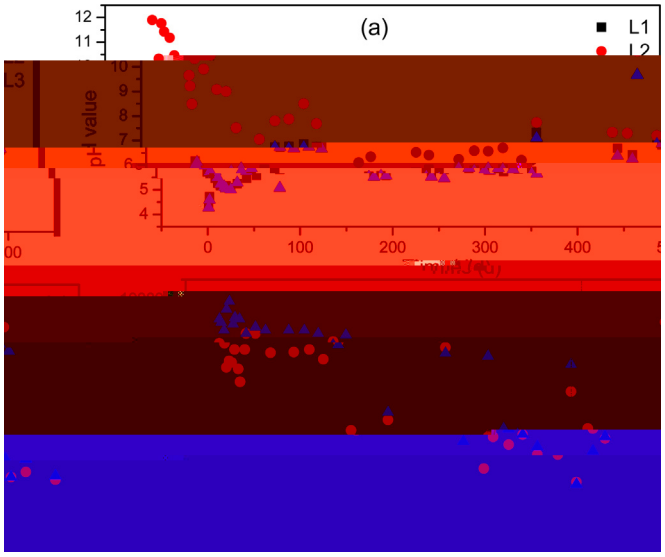

Fig. 3. Variation of pH (a) and COD (b) of L1, L2 and L3.

Table 4
Speciation of Cu in [illegible] calculated by Visual MINTEQ (mol L^{-1})

Speciation	Day 3	Day 193	Day 456
Bound to organic matter	1.64×10^{-5}	3.42×10^{-5}	4.15×10^{-6}
Bound to inorganic [illegible]	[illegible] $\times 10^{-18}$	1.07E $\times 10^{-15}$	[illegible]

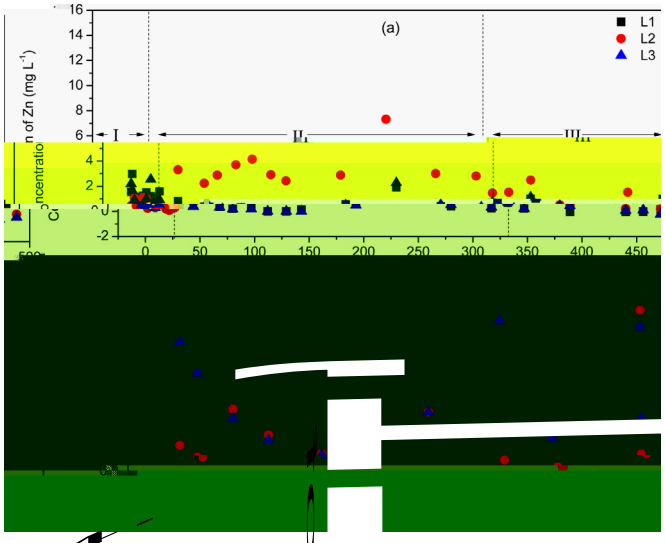

Fig. 4. Variation of Zn [illegible]

Table 5
Proportion of Cr(VI) of the total chromium in the leachates (%)

Time	L1	L2	L3
Day 9	111.22	108.91	114.40
Day 129	84.58	64.34	36.31
Day 244	17.53	18.42	19.12
Day 347	25.88	25.11	19.33
Day 488	20.36	23.82	17.05

ficial to the release of Cr. Therefore, the release of Cr exceeded the adsorption. After day 357, the Cr concentration of L2 was lower than that of L1. At that time, most of Cr was presented as Cr(III) under the anaerobic condition (Table 5). As the pH of L2 was higher than that of L1, Cr(III) was immobilized by the MSWI bottom ash layer with the formation of $Cr(OH)_3$. According to the leachate volume and the difference of Cr concentrations of L1 and L2, it was calculated that 0.78 mg of Cr was released from the MSWI bottom ash layer from day 28 to day 357, while 0.42 mg of Cr was captured by the MSWI bottom ash layer after day 357. Thus, as a whole, 0.36 mg of Cr was released from the MSWI bottom ash layer. The released Cr was lower than that of Cu and Zn, which could be due to its low unstable amounts in the MSWI bottom ash as well as its variable valence state in the landfill.

3.3. Fate of the released metals and the implication for the design of the landfill

Above results showed that 46.88 mg of Cu, 37.16 mg of Zn, and 0.36 mg of Cr were released from the MSWI bottom ash layer. However, the concentrations of Cu, Zn and Cr in L3 were generally at the same level with those of L1. For example, the average Cr concentration of L1 was 0.205 mg/L, while the average Cr concentration of L3 was 0.203 mg/L. It suggests that the MSWI bottom ash layer can not increase the final discharge of heavy metals from the landfill. The released Cu, Zn and Cr were probably adsorbed by the sub-MSW layer. Our previous study showed that the MSWI bottom ash layer increased the Cu and Zn contents of the MSW in the

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