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Effect of leachate recirculation on the migration of copper and zinc in municipal solid waste and municipal solid waste incineration bottom ash co-disposed landfill

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Abstract Municipal solid waste incinerator (MSWI) bottom ash was allowed to be disposed of with municipal solid waste (MSW) in landfill sites in the recently enacted standard of China. In this study, three sets of simulated landfill reactors, namely, conventional MSW landfill (CL), conventional MSWI bottom ash and MSW co-disposed landfill (CCL), and leachate recirculated MSWI bottom ash and MSW co-disposed landfill (RCL), were operated to investigate the environmental impact of the co-disposal. The effect of leachate recirculation on the migration of Cu and Zn in the co-disposed landfill was also presented. The results showed that the co-disposal of MSWI bottom ash with MSW would not enhance the leaching of Cu and Zn from landfill. However, the co-disposal increased the Cu and Zn contents of the refuse in the bottom layer of the landfill from 56.7 to 65.3 mg/kg and from 210 to 236 mg/kg, respectively. The recirculation of the leachate could further increase the Cu and Zn contents of the refuse in the bottom layer of the landfill to 72.9 and 441 mg/kg,

respectively. Besides these observations, the results also showed that the co-disposed landfill with leachate recirculation could facilitate the stabilization of the landfill.

Keywords MSWI bottom ash · Co-disposal · Leachate recirculation · Cu · Zn

Introduction

Incineration, due to the primary advantages of hygienic control, volume reduction, mass reduction, and energy recovery, has become an attractive method for municipal solid waste (MSW) treatment in China [1]. Till 2009, there was a total of 93 MSW incinerators (MSWI), with a treatment capacity of 71,253 t/d. However, incineration is not a final waste treatment. Large volumes of MSWI bottom ash and fly ash are produced as a result of the incineration [2]. Unlike fly ash, MSWI bottom ash is not considered a hazardous material according to the Chinese Waste Catalogue. In 2008, the Chinese government enacted ‘Standard for Pollution Control on the Landfill Site of Municipal Solid Waste’ (GB 16889-2008) [3], which allowed MSWI bottom ash to co-dispose with MSW in landfill sites without any pretreatment.

However, MSWI bottom ash contains high levels of heavy metals [4]. The co-disposal would increase the total amounts of heavy metals in landfill, which might aggravate the heavy metal pollution of the landfill. Several studies have been carried out to investigate the effect of the co-disposal on the heavy metal leaching behavior from landfill. Lo and Liao [5] reported that the co-disposal of MSWI bottom ash with MSW had not increased the metal released during their 1-year observation. Inanc et al. [6] also argued that the metal leaching was not enhanced by

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the co-disposal when the mass proportion of the MSWI bottom ash was about 50 %. This might be attributed to the high acid neutralizing capacity (ANC) of the MSWI bottom ash. The ANC of the alkaline bottom ash can neutralize the acid substance and increase the pH of the landfill, which could reduce the mobility of the heavy metals. However, the case can be different in China, where the MSW has not undergone classification before landfilling. A large proportion of MSW transferred to the landfill is composed of kitchen waste, which contains large amounts of organic matter. The organic matter in the MSW could be degraded into organic acid during the landfilling operation, forming a relatively acidic condition. In some cases in China, the pH of landfill during the acid formation phase could be less than 5.0 [7]. Such an acidic environment could greatly facilitate the mobility of heavy metals in landfill. As a matter of fact, the MSW landfill in China has already been associated with serious heavy metal pollution. For example, Long et al. [8] found that the leaching of Cu and Zn in MSW landfills in China could reach unacceptable levels. It is still unclear whether the co-disposal of MSWI bottom ash with MSW in landfill will exacerbate the heavy metal pollution in such an acidic condition. Therefore, it is necessary to evaluate the subsequent environmental impact when MSWI bottom ash is co-disposed with MSW in landfill sites. On the other hand, the bioreactor landfill with leachate recirculation has become a popular operation mode for landfill, which could enhance the decomposition of the MSW and speed up the stabilization of the landfill [9]. Several practical landfills have adopted this strategy to manage the landfill in China

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Fig. 1 Schematic of the simulated landfill systems. 1 Leachate outlet, 2 valve, 3 gravel layer, 4 MSW sampling port, 5 MSW, 6 MSWI bottom ash, 7 MSWI bottom ash sampling port, 8 sandy layer, 9 headspace, 10 vent port, 11 leachate recirculation port, 12 peristaltic pump, 13 leachate collection tank

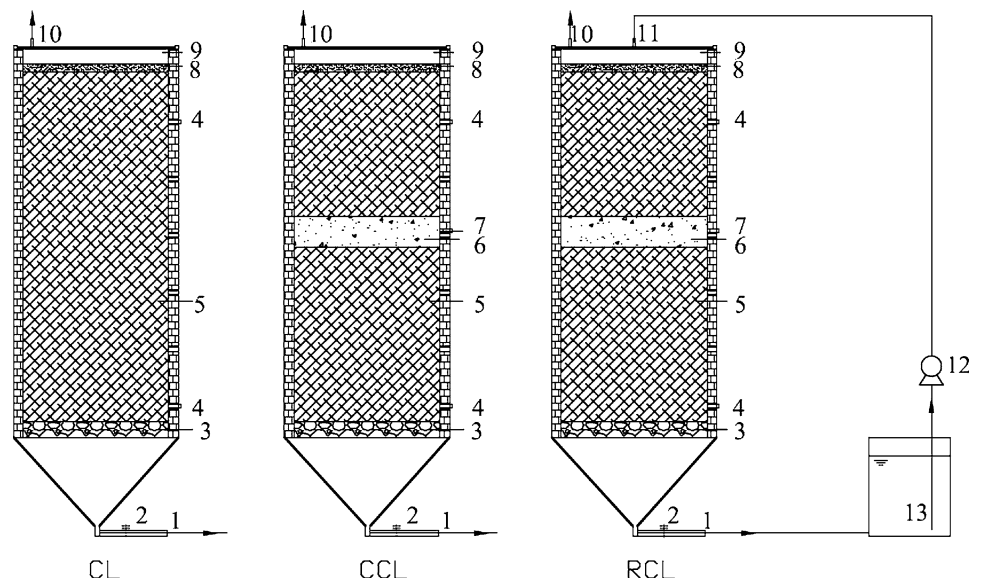


Table 1 Main characteristics of the MSWI bottom ash

Property	Value
Moisture content (%)	16.6
Bulk density (kg/m ³)	1278
Loss on ignition (%)	2.23
pH	11.2
Acid neutralization capacity at a titration point of 7.5 (ANC _{7.5})	1 mequiv. H ⁺ /g
Zn content (mg/kg dry weight)	1922 ± 33
Cu content (mg/kg dry weight)	315 ± 22

Mean ± standard deviation ($n = 3$)

MSW was collected continuously at different times of the day. Larger particles of the collected refuse were all shredded into approximately 20 mm. Then, they were manually homogenized by a shovel as thoroughly as possible prior to the loading of the simulated landfills. The moisture content of the MSW was 64.3 %. The main components of the MSW are presented in Table 2, which shows that food waste was the main component.

Operation of simulated landfills

For CL, 50 kg of MSW was loaded and compacted using a shovel and a sledgehammer. For CCL and RCL, 20 kg of MSW was loaded and compacted first. Then, 10 kg of MSWI bottom ash was loaded. Finally, another 20 kg of MSW was loaded on top of the MSW and compacted. After the compaction, the density of the waste in the reactors was found to be 0.77 t/m³. The moisture content of the refuse was adjusted to 75 % [7, 8, 13] by adding tap water to the simulated landfill after loading. The volume of the tap water added to the CL, CCL, and RCL was 21.4, 17.1, and

17.1 L, respectively. Leachate was collected and stored in the leachate collection tank. The leachate of CL and CCL was directly discarded, while the leachate of RCL was continuously recirculated using peristaltic pumps with a flow rate of 0.01 L/min every day. The duration of the recirculation varied with the leachate volume. In order to keep the volume equilibrium of leachate from RCL before recirculation, the same volume of tap water (100 mL) was added back to the leachate of RCL after sampling every week. The total tap water added back to the RCL accounted for 5.5 % of the total water content in the reactor throughout the study, which lasted for 275 days.

Analyses

Leachate samples were collected weekly from leachate outlet ports (* 100 mL). Refuse was sampled biweekly from the sample ports at the side of simulated landfills. The collected leachate and refuse were monitored to track the migration of Cu and Zn in landfills.

Leachate samples were analyzed for volume (V), pH, dissolved organic carbon (DOC), and Cu and Zn concentrations. MSW samples were analyzed for Cu and Zn contents. All these analyses were performed in accordance with 'Standard Methods for the Examination of Water and Wastewater' [14]. Cu and Zn analyses were performed using an atomic absorption spectrophotometer (Shimadzu A-650). Prior to the analysis, each sample was digested with aqua regia according to the standard method [15]. All the analyses were carried out in triplicate to ensure the validity of the results. Besides, the fractionation of Cu in the leachate was determined according to the size charge fractionation (SCF) procedure suggested by Driscoll [16]. By the procedure, Cu in the leachate was size charged into

Table 2 Components and heavy metal contents of the 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
Cu content (mg/kg dry weight)	115	176	133	84.7	148	164	–	164	127
Zn content (mg/kg dry weight)	1541	775	929	169	708	762	–	834	846

three fractions, including particulate and colloidal matter >0.45 μm (F1), non-labile complex (F2), and labile complex and free cation (F3). The leachate was filtrated through a 0.45-μm screen filter, to obtain the fraction particulate and colloidal matter >0.45 μm. The filtered leachate was then passed through a cation exchanger (a cartridge packed with sulphonic acid cation exchange resin). Cu presented as labile complexes and free cation was adsorbed onto the cation exchange resin. Cu that remained in the leachate was the fraction non-labile complex.

Results and discussion

Amounts of Cu and Zn in landfills

The amounts of Cu and Zn in CL were entirely from the MSW, and could be calculated by Eq. 1:

$$M_{CL} = \sum_1^n A \times C_i \times wt_i \% \tag{1}$$

where M_{CL} is the amount of Cu or Zn in CL, A is the mass of the MSW (50 kg in this study), C_i is the Cu or Zn content in various fractions of MSW, and $wt_i \%$ is the mass proportion of the MSW fraction (Table 2).

Cu and Zn in CCL and RCL were partly from MSWI bottom ash and partly from MSW, and could be calculated by Eq. 2:

$$M_{CCL\ or\ RCL} = A_1 \times C_{Bottom\ ash} + \sum_1^n A_2 \times C_i \times wt_i \% \tag{2}$$

where $M_{CCL\ or\ RCL}$ is the Cu or Zn amount in CCL or RCL, A_1 is the mass of the input MSWI bottom ash and A_2 is the mass of the input MSW, $C_{Bottom\ ash}$ is the Cu or Zn content in bottom ash, C_i is the Cu or Zn content in various fractions of MSW, and $wt_i \%$ is the mass proportion of the MSW fraction.

According to Table 2 and the moisture content of MSW, the Cu and Zn contents in MSW were calculated, which were 46.2 and 410.4 mg/kg wet weight, respectively. The Cu and Zn contents in MSWI bottom ash were 262.7 and 1602 mg/kg wet weight, respectively. Therefore, the amounts of Cu and Zn in the reactors could be calculated. The Cu and Zn amounts in CL were 2285 and 20290 mg,

respectively. The amounts of Cu and Zn in CCL and RCL were 4445 and 32261 mg, respectively. Due to the co-disposal, the Cu and Zn amounts increased by 94.5 and 59.0 %, respectively. The increase of the Cu and Zn amounts might aggravate the potential threat of the landfill if the non-linear releasing behavior of Cu and Zn occurred, a concern that has been expressed by several researchers [8, 17, 18].

Variation of the leachate characteristics

The variation of the leachate characteristics, including the pH, DOC, volatile fatty acid (VFA), and NH_4^+ , are shown in Fig. 2. The initial pH was low in all three landfills, which was 4.96, 5, and 5.06 for CL, CCL, and RCL, respectively. It showed a fast increase from days 1 to 22, which might be due to the aerobic degradation of the organic acid by the residual oxygen in the landfills. The pH of CCL and RCL leachate was higher than that of CL from days 59 to 129, which might be due to the high acid neutralizing capacity of the MSWI bottom ash. An interesting phenomenon was observed after day 129. The pH of the RCL leachate showed a remarkable increase to 7.5 and remained relatively steady afterwards, which was close to the pH of a mature landfill. However, the pH of CL and CCL remained at about 6.0 from day 129 to the end of the study. Furthermore, the DOC and VFA of the leachate showed a variation corresponding to the variation of the pH. The initial DOC was about 20000 mg/L for the three landfills. It showed an increasing trend from days 1 to 88. The DOC of the leachate of CL was higher than that of CCL and RCL, which was consistent with the MSW amount in the three landfills. The DOC of the leachate of RCL showed a significant decrease from 16753 to 4182 mg/L after day 129. However, the DOC of leachate of CL and CCL was still above 20000 mg/L. The variation of the VFA in the leachate was similar to that of DOC. The VFA of RCL showed a significant decrease from 16358 to 945 mg/L after day 129, while the VFA of the leachate of CL and CCL was still above 20000 mg/L. This result was generally consistent with the gas generation and settlement of the reactors. The generated gas from RCL was sometimes beyond the upper monitoring limit of the Smith fermentation tube, while the gas volume of the CL and CCL remained relatively mild. The settlement of RCL was

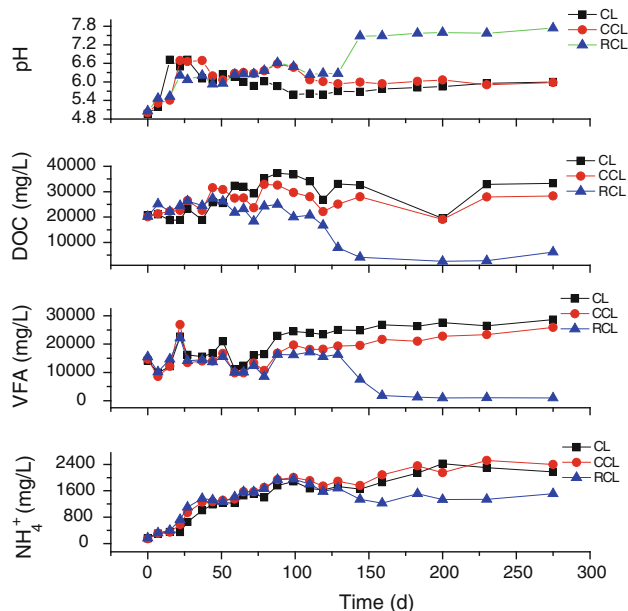


Fig. 2 Variation of pH, DOC, VFA, and NH_4^+ in the leachate of CL, CCL, and RCL

also more rapid than that of CL and CCL. Besides, the NH_4^+ of the leachate remained steady at about 1400 mg/L, even after day 129. The results showed that the RCL had reached the stabilization phase, while the CL and CCL were still in the acetogenic phase. It appeared that the recirculation of leachate in MSWI bottom ash and MSW co-disposed landfill could enhance the stabilization of landfill. On the one hand, the recirculation of the leachate increased the moisture content of the landfill and it provided better conditions to release the alkaline buffer from the bottom ash, which could neutralize the acidic condition of the landfill. These processes were beneficial for the biostabilization of the landfill. As the leachate pH increased with the stabilization of the landfill, the recirculation of the leachate might, in turn, facilitate the immobilization of heavy metals in the landfill.

Cu and Zn concentrations in leachates

Cu

The concentration of Cu in the leachate of CL, CCL, and RCL is shown in Fig. 3, demonstrating a decreasing trend for all three landfills, which was generally consistent with previous reports [8, 19]. The initial concentration of Cu was relatively high, which was 3.14, 3.06, and 3.06 mg/L for the leachate of CL, CCL, and RCL, respectively. It was thought that most of the unstable Cu in MSW and MSWI bottom ash was apt to leach out at the beginning of the operation. The concentration of Cu decreased to 0.02 mg/L

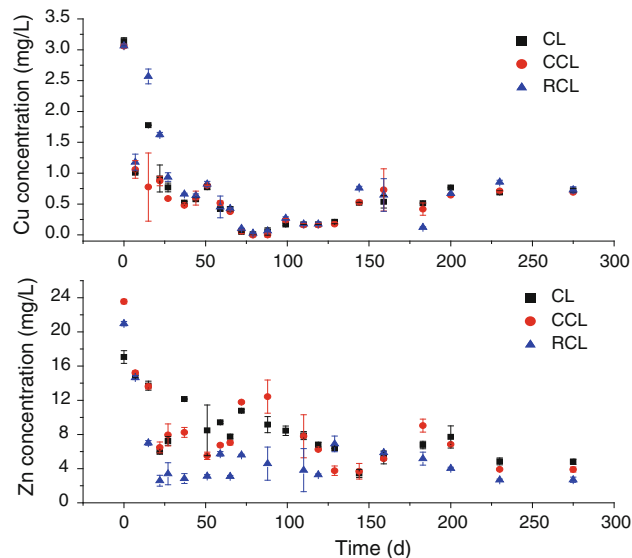


Fig. 3 Concentrations of Cu and Zn in the leachate of CL, CCL, and RCL

when the landfills were operated for 79 days. It seemed that the leaching and immobilizing of the Cu had reached a balance in the landfill, leading to the low leaching of Cu. However, the Cu concentration showed a slight increase afterwards, ranging from 0.17 to 0.87 mg/L. This might result from the degradation of MSW in landfill, which released a part of the Cu bound with the organic matter in MSW. The concentration of Cu in the leachate of the three landfills could meet the Discharge Standard for Waste Water of China (GB 8987-1996, Grade II: $\text{Cu} \leq 1.0$ mg/L as the threshold) after 15 days of the experiment [20]. Therefore, special attention should be paid at the beginning of the landfilling, as far as the Cu pollution is concerned.

No significant difference was observed among the leachates of the three landfills, although the pH of the leachates varied greatly between each other (Fig. 2). This result agreed well with other studies. For example, Long et al. [7] investigated the Cu concentration in the leachates of the conventional landfill and leached recirculating landfill. No significant difference was observed between the landfills. This phenomenon was often attributed to the adsorption and precipitation capacity of the refuse [8]. However, besides the adsorption and precipitation, the bounding of Cu on the organic matter should also be considered. As is well known, Cu has a high affinity for organic ligands, which is abundant in all three landfills. Several researches have found that the leaching of Cu is controlled by the organic ligands [21, 22]. It could also be proved by the fractionation of Cu in the leachate of this study (Fig. 4), which showed that most of the Cu was present as particulate and colloidal matter $>0.45 \mu\text{m}$ and non-labile complex, indicating that most of the Cu in the

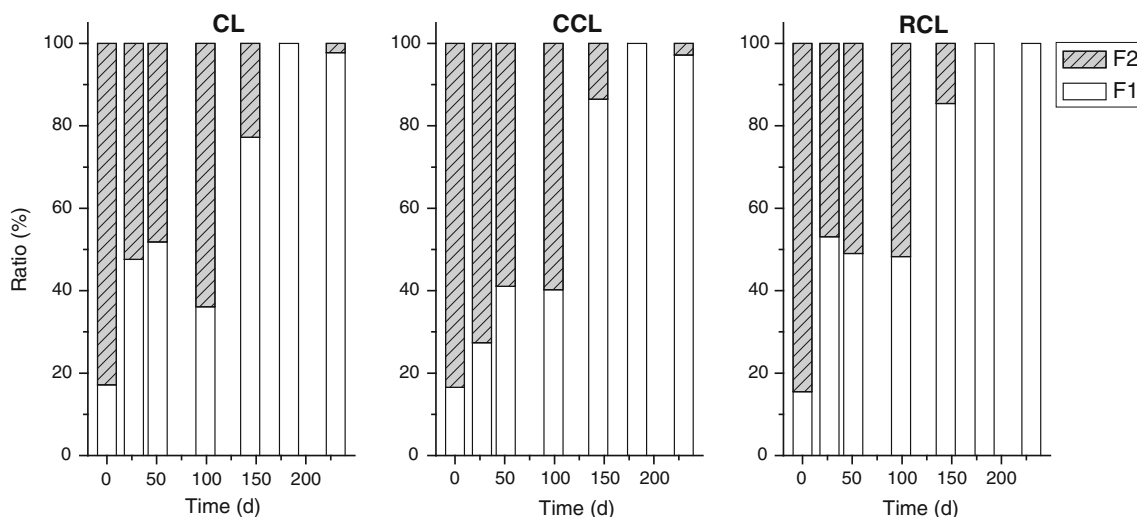


Fig. 4 Fractionation of Cu in the leachate of CL, CCL, and RCL (data are shown as the percentage relative to the total concentration). F1 Particulate and colloidal matter >0.45 μm. F2 Non-labile complex

leachate was bounded with the organic matter [23]. Therefore, the co-disposal of MSWI bottom ash as well as the recirculation of the leachate cannot greatly affect the leaching behavior of Cu.

Zn

At the beginning of the operation (days 0–27), the Zn concentration of CCL leachate was higher than that of CL (Fig. 3). For example, the initial Zn concentration was 23.6 mg/L for the CCL leachate, which was higher than that of CL (17.1 mg/L). This might be due to the fact that the co-disposal of MSWI bottom ash brought more leachable Zn to the landfill. After day 27, the Zn concentration of CCL leachate was lower than that of CL, which generally corresponded to the variation of pH (Fig. 3). Due to the high ANC of the bottom ash, the pH of CCL leachate was higher than that of CL after day 27 (Table 1). As the mobility of Zn is greatly dependant on the pH, the concentration of Zn in CCL leachate was restricted to a low level.

The concentration of Zn in RCL leachate was generally lower than that of CL and CCL throughout the study. Due to the leachate recirculation, Zn in the RCL leachate was brought back to the landfill. Some of the particulate matter in the recirculated leachate was retained by the refuse due to the action of filtration, which could decrease the Zn concentration at a certain content, as some of the Zn was present as the particulate matter [24]. Besides, the dissolved Zn in leachate could be immobilized by the VFA (Eq. 3, with Ac as an example), S²⁻ (Eq. 4), and humic substance (Eq. 5, with fulvic acid [FA] as an example) inside the landfill [8, 25]. Furthermore, a part of Zn was adsorbed by the refuse [19, 26], which could be proved by the variation of the Zn content in the refuse of RCL, as

discussed in the following section. These might be the possible reasons for the low concentration of Zn in RCL leachate before day 129. After day 129, the pH of RCL leachate increased from 6.3 to 7.5, while the pH of CL and CCL leachate remained at about 5.7 and 6.0. The high pH of RCL leachate could keep the Zn concentration at a low level:



The results above suggested that the co-disposal of MSWI bottom ash with MSW would not increase the leaching of Zn from landfill, no matter if the leachate was recirculated or not. As a matter of fact, the leaching of Zn was remarkably decreased due to the co-disposal and leachate recirculation.

Besides, although the Zn concentration in leachate of CL, CCL, and RCL showed a decreasing trend, it was apt to exceed the Discharge Standard for Waste Water of China (GB 8987-1996, Grade II: Zn ≤ 5.0 mg/L as the threshold) throughout the study. Therefore, the pollution of Zn should be an important concern throughout the landfilling.

To further explore the environmental impact of the co-disposal and leachate recirculation, the accumulated amounts of the leached Cu and Zn from the three landfills were calculated. The total leached Cu from CL, CCL, and RCL was 11.1, 11.3, and 4.6 mg, respectively. The total leached Zn from CL, CCL, and RCL was 151, 144, and 14.7 mg, respectively, suggesting that the recirculation of the leachate also decreased the heavy metal released from the landfill.

Cu and Zn in the refuse

Cu

The variation of Cu content in the refuse of the upper and bottom layers of landfill is shown in Fig. 5a, b. The content of Cu in the refuse experienced a reduction and an increase throughout the study, which was consistent with our previous research [7]. At the beginning of the operation, no significant difference of Cu content was observed between the upper layers of CL, CCL, and Fig.

limited in landfill without leachate recirculation. However, the Zn content of the refuse in the bottom layer of RCL was notably higher than that of the upper layer, implying that the leachate recirculation facilitated the migration of Zn. Besides, the Zn content of the refuse in the bottom layer of RCL was higher than that of CL and CCL. After day 77, the Zn content of refuse in the bottom layer of RCL ranged from 279 to 545 mg/kg, with an average content of 441 mg/kg. This value was higher than that of CL (210 mg/kg) and CCL (236 mg/kg). This suggested that the MSWI bottom ash and MSW co-disposed landfill with leachate recirculation could significantly increase the Zn content of the refuse in the bottom layer of landfill. The result was also consistent with the low level of Zn concentration in the RCL leachate. Moreover, the average Zn contents of the refuse in the bottom layer of CL and CCL were lower than the China Environmental Quality Standard for Soils (GB15618-1995, Grade II for soil pH <6.5: Zn \leq 250 mg/kg) [27]. However, the average Zn content of the refuse in the bottom layer of RCL exceeded the standard.

The results suggest that the MSWI bottom ash and MSW co-disposed landfill with leachate recirculation can increase the Cu and Zn contents of the refuse in the bottom layer of the landfill. Therefore, the environmental threat of the landfill can be aggravated. Special seepage-proofing measures are needed if the landfill is operated by this mode.

Conclusions

Municipal solid waste incinerator (MSWI) bottom ash and municipal solid waste (MSW) co-disposed landfill with leachate recirculation could facilitate the stabilization of the landfill. It could also reduce the release of Cu and Zn from the landfill. However, the co-disposal could increase the Cu and Zn contents of the refuse, especially when the leachate of the co-disposed landfill was recirculated. The average Cu and Zn contents of the refuse in the bottom layer of landfill increased from 56.9 to 72.9 mg/kg and from 210 to 441 mg/kg, respectively, due to the co-disposal and leachate recirculation, which exceeded the China Environmental Quality Standard for Soils. This might be the main concern when MSWI bottom ash was disposed of with MSW in landfill, as allowed in the newly enacted standard. Special seepage-proofing measures must be considered to prevent the non-liner leaching of Cu and Zn from the bottom layer of the landfill when the MSWI bottom ash is co-disposed with MSW.

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