The metal-leaching and acid-neutralizing capacity of MSW incinerator ash co-disposed with MSW in landfill sites

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Abstract

Municipal solid waste (MSW) incinerator (MSWI) bottom ash and fly ash were used as landfill cover or were co-disposed with MSW to measure their potential metal-releasing and acid-neutralizing capacity (ANC) in landfill sites. Five lysimeters (height 1.2 m, diameter 0.2 m), simulating landfill conditions, were used in the experiment. Four contained either bottom ash (BA) or fly ash (FA) with BA:MSW ratios of 100 and 200 $g L^{-1}$ and FA:MSW ratios of 10 and 20 $g L^{-1}$, and the fifth was the control, which contained no ash. The lysimeters were arranged so as to contain four layers, with BA or FA placed on top of MSW within each layer. Each lysimeter was recirculated with 100 mL leachate using peristaltic pumps, and 100 mL of the leachate was collected weekly to measure the soluble metal concentrations. The results showed that the concentrations of soluble alkali metals measured in the leachate were in the order $Ca > K > Na > Mg$. In addition, the concentrations of soluble alkali metals of Ca and K collected from the lysimeters containing FA were found to be higher than the concentrations from the lysimeters containing BA. The concentrations of heavy metals (Cd, Cr, Cu, Ni, and Zn) were found to be $<1 mg L^{-1}$ except for Pb, which reached $2 mg L^{-1}$. These results suggest that for alkali metals there might be an ANC consistent with the results of an acid titration curve, which would provide suitable conditions for anaerobic digestion of the MSW in the landfill. Furthermore, heavy metals and trace metals were found in concentrations, which were too low to exert inhibitory effects on anaerobic digestion, and thus they could serve as micronutrients to exert beneficial rather than detrimental effects on landfill biostabilization.

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1. Introduction

Municipal solid waste (MSW) incinerator residues, such as bottom ash (BA) and fly ash (FA), have been of environmental concern, as they may cause secondary pollution to the environment if they are disposed of or utilized in an improper way. Ash has been utilized for agricultural soil amendment, granular fill, road base, aggregate or mono-disposal, as well as for many other purposes [1–7]. However, the release of different kinds of ions, including heavy metals, was thought to have the potential to pollute the environment or affect the extent of further utilization [8]. As the release of metals was the major concern for the disposal or utilization of ash, the potential release of ions

was investigated extensively and the extraction methods that might affect the release of metals were also reported [2,9–13].

With regard to fly ash, the solubility could be affected by pH, silicate and non-silicate distribution, magnetic and non-magnetic fractions, and humic matter [14–16]. The release of metals controlled by individual ion solubility has shown that the acid-neutralizing capacity (ANC) potential of ash is mainly due to the release of Ca, Mg, K, Na, Fe, and Al metals [17–22]. The release of these alkali metals might increase the pH value that is suitable for anaerobic processes such as municipal solid waste (MSW) and wastewater anaerobic digestion [23]. Investigations of co-composting sewage sludge and coal ash residues, similar to this study, have also shown that ash has the potential to enhance alkalinity and enzyme activity except when a higher amount of fly ash was applied [24]. Furthermore, the adverse effect of thermophilic bacteria was not affected except for when 25% of fly ash was added.

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The increase in alkalinity caused by alkali metals could be expressed as ANC, which could neutralize the volatile acids in a beneficial way for the anaerobic digestion process [23,24]. In addition, the release of metals through the use of ash and their probable sorption to MSW might also affect the subsequent anaerobic digestion. Thus, the components of MSW such as cellulose, hemicellulose, and lignin [25,26] and the solubility of individual compounds [27,28] in the ash would be the key concerns associated with the ANC benefits from metal release. Thus, the study aimed to investigate metal release and the ANC potential of MSW incinerator ash co-disposed with MSW that might affect landfill biostabilization and other utilization under different environmental conditions, as it is known that such properties might play an important role both at the disposal and the utilization site.

2. Materials and methods

2.1. MSWI ash, MSW, and seeded sludge

The ash used in this study was derived from a mass burning incinerator operating at a temperature of 850–1050 °C in central Taiwan. Bottom ash was obtained from the furnace of the MSW incinerator (MSWI) as well as the siftings from the stoker. Fly ash was derived from the air pollution control residues such as the semi-dry scrubber and bag filter. Generally, the particle size of bottom ash was greater than that of fly ash. This phenomenon implies that fly ash would have a higher potential release of ions, including heavy metals and alkali metals, than bottom ash, mainly due to its higher specific surface area and higher total content than bottom ash [2,23].

The MSW substrate consisted of synthetic refuse comprising office paper (30%), newspaper (30%), food waste (5%), and hay (35%), which was assumed to be typical of the organic fraction of municipal refuse. The chemical composition of the synthetic refuse was around 46% C, 6% H, 41% O, 1.4% N, and 5.6% other elements. The refuse was shredded into pieces of about 1 cm, and was then further blended with deionized water and ground into particles about 1 mm in size (6% total solids; TS). The MSW was stored in a freezer before being used in the experiments. In order to simulate the anaerobic status of landfill sites, seeded sludge (TS 2.5%) was collected from a municipal wastewater treatment plant to be mixed with MSW at the start of the experiment, to create landfill anaerobic conditions. The metal contents of ash, MSW, and sludge are shown in Fig. 1.

2.2. Lysimeters

Each lysimeter was 20 cm in diameter and 120 cm in height, with a working volume of about 34 L. The control lysimeter contained a mixture of 22 L of synthetic MSW substrate (TS 6%; VS 5%) and 12 L anaerobic seeded sludge (TS 3%; VS 2.5%) from a municipal wastewater treatment plant. The other four ash-amended lysimeters contained about 34 L of MSW substrate and seeded sludge in the same proportions as the control lysimeter and were amended further with different ratios of bottom and fly ash. These ash-amended lysimeters were arranged so as to contain four layers each with 7.5 L of the mixture of MSW substrate and anaerobic seeded sludge followed by a 1 L mixture of MSW and seeded sludge blended with MSWI bottom and fly ash in ratios designed to simulate either landfill cover or co-disposal. The five experimental ash to MSW ratios corresponded to 0 (control lysimeter), 10 and 20 g L^{-1} (10 and 20 g fly ash per liter MSW for FA-amended lysimeters), and 100 and 200 g L^{-1} (100 and 200 g bottom ash per liter MSW for BA-amended lysimeters). The leachates in the five lysimeters were recirculated using peristaltic pumps with a volume of 100 mL each per day. Another 100 mL of leachate was sampled weekly to measure the amounts of metals released. The five lysimeters were placed on a homeostatic oven kept at a temperature of about 35 °C, simulating the landfill environment.

The ash's potential release of alkali metals and heavy metals and other ions such as Cl^{1-} and SO$_4^{2-}$ that might affect the biostabilization of MSW in the lysimeters was analyzed by ICP-OES (Thermo Electron Corporation) and IC (Dionex), respectively. COD, pH and TS in leachates from the lysimeters were analyzed to monitor their effects on metal release potential. All measurement procedures and methods were carried out in accordance with standard methods for the examination of water and wastewater [29].

2.3. The release of metals from ash according to an acid titration curve

In order to understand the potential release of metals in a broader pH range (pH 1–12), an acid titration curve for BA and
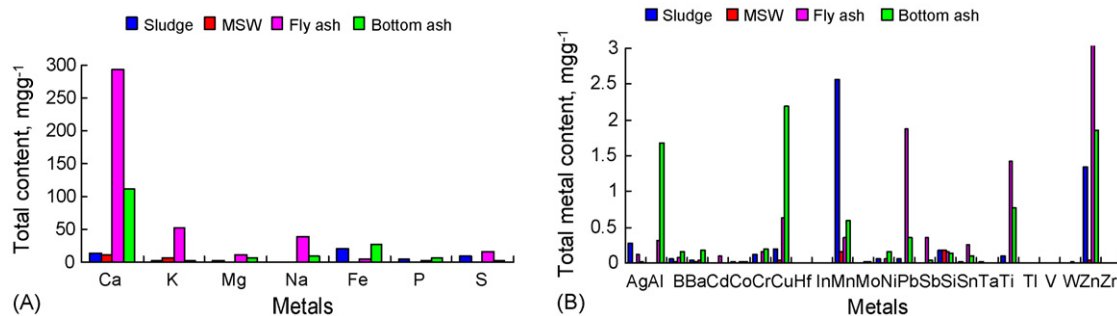

Fig. 1. Selected metal content of MSWI bottom ash, MSWI fly ash, MSW and seeded sludge.

FA was carried out in batch flasks. A sample of 4 g of FA exposed with 200 mL deionized water and 200 mL MSW substrate was used to obtain the acid titration curve [17,23]. Similarly, 30 g of BA was exposed with 200 mL deionized water and 200 mL MSW substrate to conduct the acid titration experiment. These exposed ratios were close to the BA and FA to MSW ratios in the lysimeters. Alkali metals of Ca, K, Mg, and Na; six heavy metals; and other trace metals were measured by ICP-OES (Thermo Electron Corporation) Different normalities of 0.1–5N HNO_3 were employed during the titration process depending on the ANC potential of the ash suspended within the deionized water and MSW substrate. The release of metals in an acid titration curve could provide an indication of heavy metals, alkali metals, and trace metals, which might affect landfill biostabilization or other utilization.

3. Results and discussion

3.1. The release of metals from lysimeters

The alkali metals Ca, K, Mg, and Na, and six heavy metals (Cd, Cr, Cu, Pb, Ni, and Zn) in the leachates were detected by ICP/OES. The results are presented in Fig. 2. The alkali metals were found in greater concentrations than the six heavy metals and were considered as the major components of the MSWI ash, as shown in Fig. 1. Among the alkali metals, Ca was found in a higher soluble concentration than the other three alkali metals (Fig. 2A–D). This phenomenon was consistent with the higher total content of Ca (Fig. 1), resulting in a higher Ca release. In addition, the soluble Ca concentration in the $20\,gl^{-1}$ ash-amended lysimeter was also found to be higher than in the other
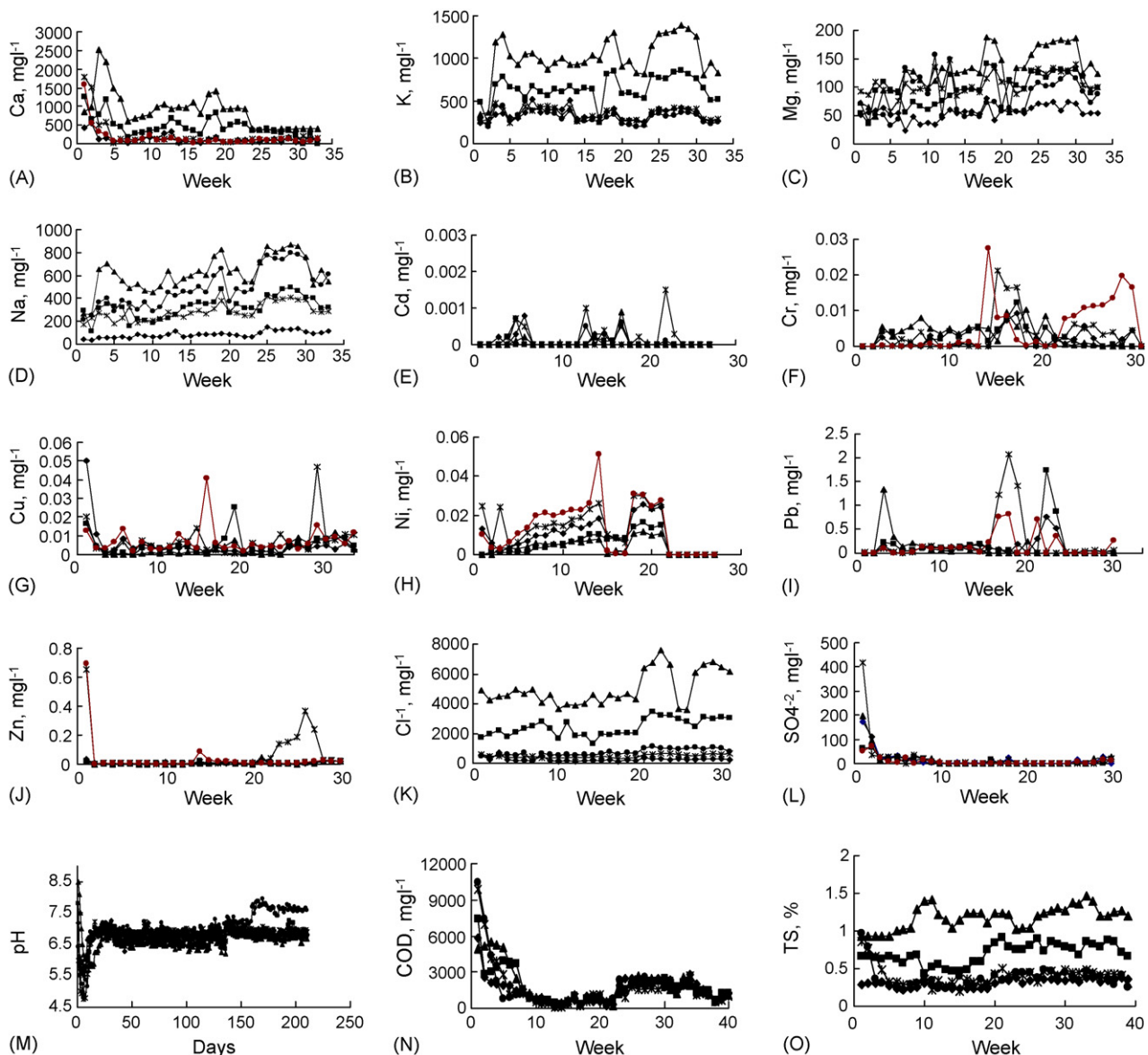

Fig. 2. Released alkali metals, six heavy metals and Cl^{-1} and SO_4^{-2} in five lysimeters and the pH, COD and SS in leachates in five lysimeters: (♦) control; (▲) FA $20\,g\,L^{-1}$; (■) FA $10\,g\,L^{-1}$; (*) BA $100\,g\,L^{-1}$; (●) BA $200\,g\,L^{-1}$.
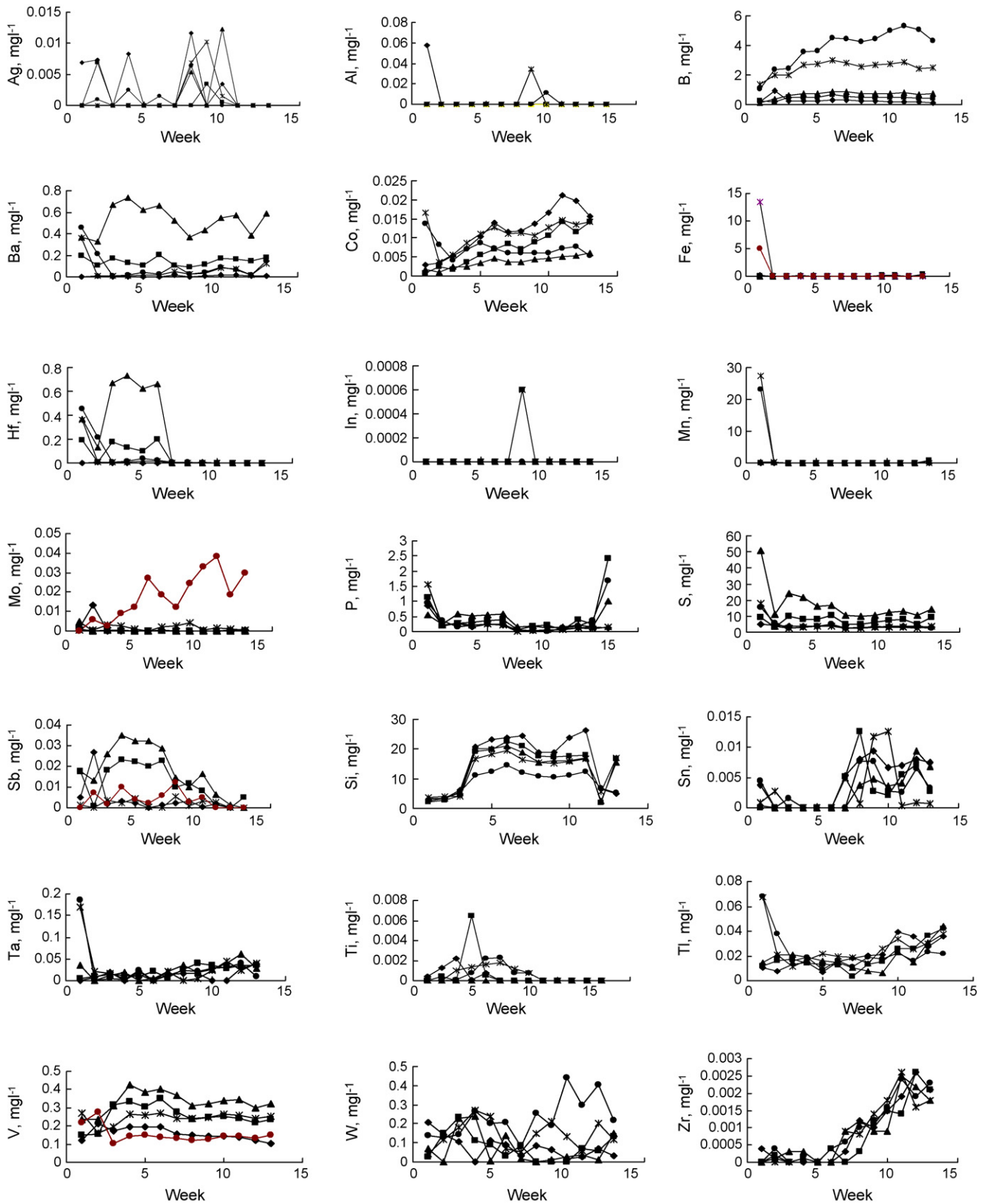
Fig. 3. Released trends of 21 trace elements in five lysimeters: (♦) control; (▲) FA 20 g L^{-1}; (▦) FA 10 g L^{-1}; (*) BA 100 g L^{-1}; (●) BA 200 g L^{-1}.

four lysimeters. This higher soluble concentration of Ca might come from the dissolution of $Ca(OH)_2$, $CaSO_4$, CaO, etc. The higher Ca and alkali metals and their hydroxides and carbonates were thought to be the major ANC for the provision of an alkaline buffer. Thus, they might offer an alkalinity buffer if used as landfill cover in landfill sites as the anaerobic process progresses.

Cd, Cr, Cu, Ni, Pb, and Zn in the leachate were measured in the ranges 0–0.002, 0–0.03, 0–0.05, 0–0.06, 0–2, and 0–0.4 $mg\,L^{-1}$, respectively (Fig. 2E–J). Among these, Pb was found to have a higher release than the other five heavy metals. Heavy metal concentrations were below 1 $mg\,L^{-1}$ (except for Pb), which is less than the concentration, which would inhibit the anaerobic digestion process [30]. This small amount of heavy metal release could probably be adsorbed by MSW and sludge, leading to a lower amount of leaching as compared with mono-ash disposal [25,26,31]. Moreover, heavy metals might serve as micronutrients rather than detrimental concentrations under these circumstances [32]. The release of 21 trace metals (Fig. 3) was also detected, in order to measure their soluble concentration for the purposes of landfill biostabilization and leachate treatment control. These concentrations of metals released were also found suitable to serve as nutrients rather than inhibitory toxicants [32]. Other elements such as Fe, Al, Si, Mn, P, and S were found in higher concentrations at lower pH values than the other trace metals such as Ag, B, Ba, Hf, In, Mo, Sb, Sn, Ta, Ti, Tl, V, W, and Zr (see Fig. 3). The TS values (2O) in leachates were found in the same order as for alkali metal release: $20\,g\,L^{-1} > 10\,g\,L^{-1} > 200\,g\,L^{-1} \fallingdotseq 100\,g\,L^{-1} >$ control. COD (2N) seemed to show a decreasing trend in the five lysimeters. pH (2M) was found to be the same (near neutral pH in all five lysimeters except in the first stage of about 3 weeks, which was mainly attributed to the MSW hydrolysis resulting in a higher concentration of volatile acids and a lower pH). Chloride ions (2K) were found to be higher in FA-amended lysimeters. However, sulfate ions (2L) showed the same decreasing trend, which might be attributable to their consumption by sulfate-reducing bacteria.

3.2. Release of metals examined using an acid titration curve

The release of alkali metals, heavy metals, and trace metals was further examined using acid titration curves from batch flasks in a broader pH range between pH 1 and 12 (Fig. 4). It was noted that the release of metals was found to be higher in the lower pH range, especially below pH 3. In most cases, metal release from both kinds of ash (4 g FA and 30 g BA) exposed with media such as deionized water and MSW substrate showed the order: 200 mL (deionized water) >200 mL (MSW substrate). The release of the alkali metals of Ca, K, Mg, and Na was found to be higher than the release of heavy metals and the other 21 trace elements. Among alkali metals, Ca showed the highest release as compared to K, Mg, and Na, and other heavy and trace metals. In the FA-amended batch flasks, Ca, K, Mg, and Na release from pH 1 to 12 ranged between 100 and 400, 30 and 70, 0.01 and 12, and 20 and 60 $mg\,g^{-1}$, respectively, whilst in BA-amended batch flasks, Ca, K, Mg, and Na release ranged

between 10 and 60, 1 and 6, 0 and 7, and 0 and 2 $mg\,g^{-1}$, respectively. Similarly, the release of the six heavy metals was found to be higher at lower pH values. The release of the six heavy metals Cd, Cr, Cu, Ni, Pb, and Zn in the FA-amended acid titration curve was found to be 0–35, 0–20, 0–160, 0–0.08, 0–600, and 0–1500 $\mu g\,g^{-1}$, respectively. The release of the six heavy metals in the BA-amended acid titration curve was found to be lower than that in the FA-amended acid titration curve. These six heavy metals: Cd, Cr, Cu, Ni, Pb, and Zn in the BA-amended acid titration curve were found to be 0–0.02, 0–0.15, 0–15, 0–0.003, 0–1, and 0–30 $\mu g\,g^{-1}$, respectively.

3.3. Implications

The higher content of Ca and its dissolution were considered to be the major contribution of ANC in MSWI ash [23,28,33]. The dissolution of many compounds was controlled by their solubility constant and can be modeled by MINTEQA2 [28]. Many investigations of the ANC of bottom ash and fly ash from miscellaneous substances such as waste, wood, coal, tires, peat, and bark have been reported [21,22]. These results showed that $ANC_{pH\,1}$ of bottom ash was about 10 L of 1 M H_2SO_4 per kg of dry solids and slightly lower than that of about 12 L/kg for waste fly ash.

The higher Ca content and release in the FA-amended lysimeters was mainly attributed to the air pollution control device using semi-dry $Ca(OH)_2$ as a scrubber reagent. Thus, $Ca(OH)_2$ contributed to the large amount of Ca in FA that might affect its further utilization. Ca, K, Mg, and Na were thought to offer the alkalinity and had ANC potential that might be beneficial to landfill biostabilization, acid neutralization, or other utilization. The unit pH drop for acids added equivalent mequiv. (mequiv. H^+ added/unit pH drop; that is, ΔANC (mequiv. H^+)/Δ pH) in the FA and BA acid titration curves can be seen in Fig. 5, which shows that FA had a higher acids added equivalent than that of BA, particularly in the pH ranges of 10–12 and 1–2 (Fig. 5). Both FA and BA had an ANC potential mainly attributed to the Ca, K, Mg, and Na. This phenomenon suggests that FA and BA have the potential to provide the necessary alkalinity, especially for landfill biostabilization, anaerobic digestion, soil amendment, and other purposes needing alkaline conditions. However, the potential higher release of alkali metals might exert detrimental effects on the utilization of ash. Therefore, suitable ratios of ash addition need be chosen in order to improve the utilization purpose, because addition ratios will affect the leaching characteristics, leaching concentrations, and therefore the final results, with regard to the different scenarios of utilization.

As MSWI ash was employed as a landfill cover, the potential toxicity of heavy metals to the microorganisms and the ecological environment and the human health risk were the major considerations. From the results of the lysimeter experiments, Cd, Cr, Cu, Ni, and Zn were released at concentrations under 1 $mg\,L^{-1}$, which may not exert any inhibitory effects on MSW anaerobic biodegradation [30]. In addition, these five released concentrations did not exceed the groundwater regulatory standards in Taiwan (Cd: 0.05; Cr: 0.5; Cu: 10; Ni: 1.0;
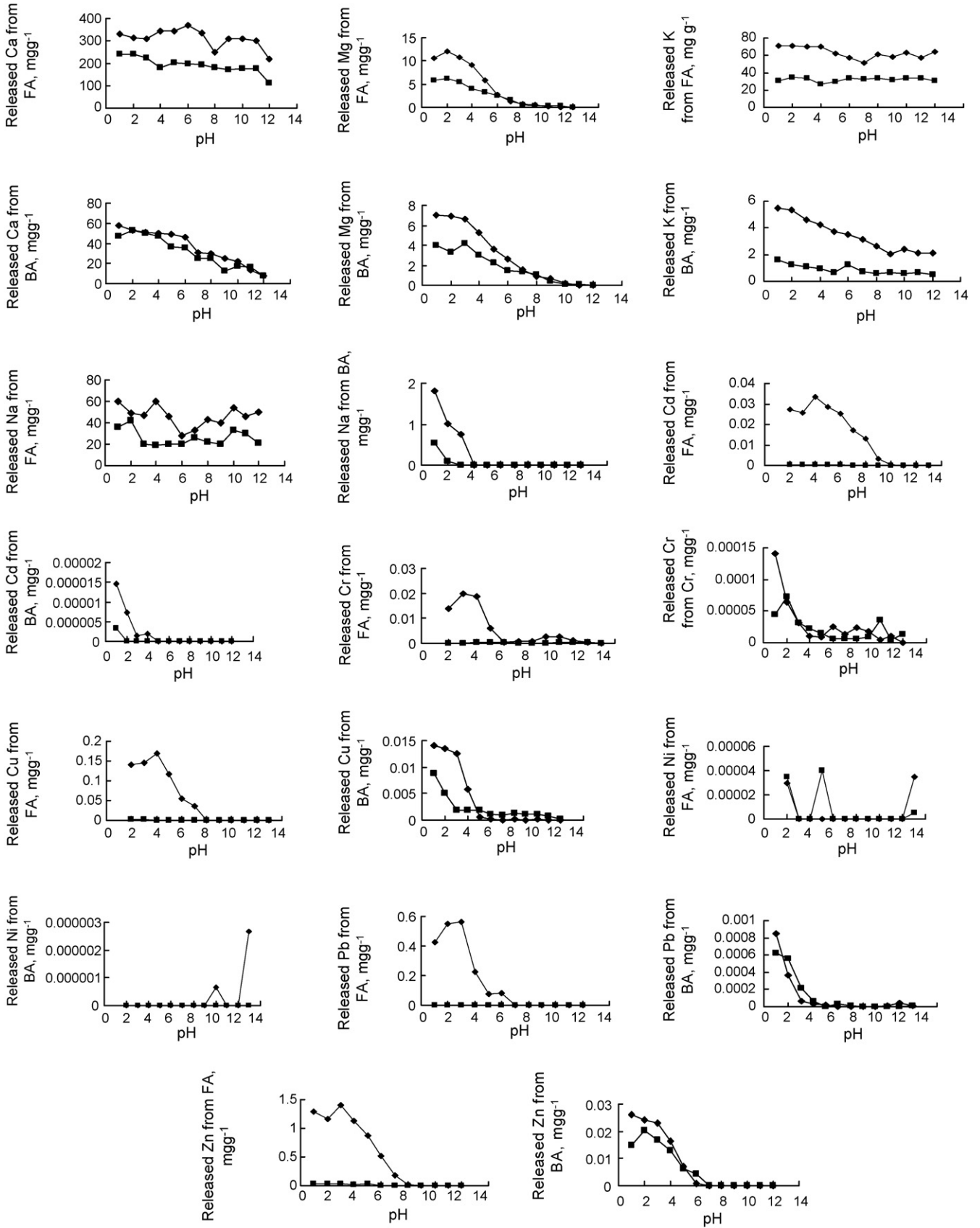
Fig. 4. Release of alkali metals and six heavy metals from BA and FA exposed with 200 mL deionized water (♦) and 200 mL MSW (▒) in acids titration curve.
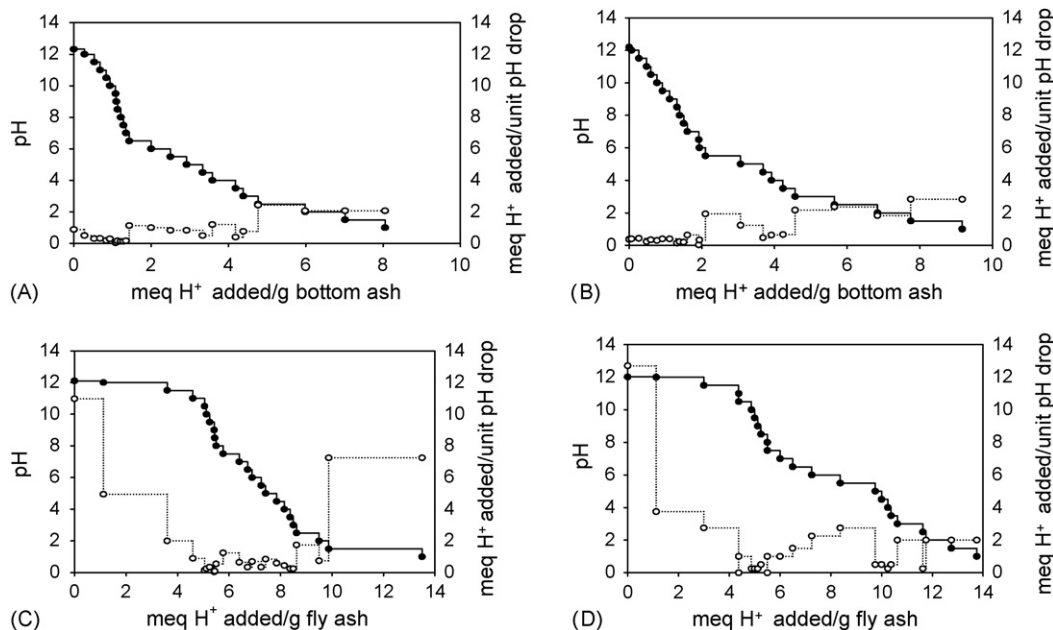
Fig. 5. pH (●) and mequiv. H⁺ added/unit pH drop (○) in acids titration curves of 30 g BA (A and B) and 4 g FA (C and D) exposed with 200 mL deionized water and 200 mL MSW, respectively.

Zn: 50 mg L^{-1}). However, the Pb concentration, which reached 2 mg L^{-1}, does exceed the Taiwan groundwater regulatory standard of 0.5 mg L^{-1} and could have potential detrimental effects on the ecological environment and anaerobic processes. Furthermore, the amounts of Cd, Cu, Ni, and Zn found near the landfill site after MSWI had been co-disposed with the MSW were found to exceed the Taiwan soil regulatory standard. This result indicated that heavy metal release has the potential to cause a risk to human health and the ecological environment. Thus, it is essential that leachates from MSW landfill sites co-disposed with ash need to be treated carefully to prevent groundwater pollution. Further, remediation of the contaminated soil in the landfill site is also required to avoid ecological and human health risks.

In addition, releases of Ca, K, Mg, and Na in lysimeters were reported to be under inhibitory concentrations. However, released concentrations of Ca at a lower pH, as can be seen from the titration curve, were found to have potential strong inhibitory effects on anaerobic biodegradation [30]. In fact, the practical MSW anaerobic biodegradation should be maintained at neutral pH values between 6.5 and 7.5 to maintain an active anaerobic process. In this pH range, Ca, K, Mg, and Na (Figs. 2 and 4) seemed not to exert detrimental effects on anaerobic biodegradation. Rather, they were able to offer an alkaline buffer that was suitable for the anaerobic biological process. Released concentrations of other trace metals such as Co, Mo, Ni, W, Se, etc in the lysimeters and in the batch flasks by acid titration curves also showed that they might serve as micronutrients rather than as inhibitory metals [32]. Therefore, it was assumed that alkali metal release from 10 and 20 g L^{-1} FA-amended lysimeters and 100 and 200 g L^{-1} BA-amended lysimeters could provide the potential ANC and heavy metal release (except for Pb) that might exert beneficial rather than detrimental effects on the anaerobic process. It was therefore proposed that an enhancement of land-

fill biostabilization could be expected if suitable ash addition ratios were employed.

The release of metals was found to be lower from BA and FA exposed with MSW substrate as compared with that exposed with deionized water. This result implies that a large proportion of the released metals and ions could probably be adsorbed by the MSW substrate. Metal-binding behaviors have been investigated by several researchers [23,25,31,34,35]. These studies have revealed that metal-binding was mainly due to the functional groups such as organic and amino acid-type compounds and seven binding ligands. MSW components have also been found to contain cellulose, hemicellulose, lignin, lipophilic extractives, crude protein, and ash [26]. Furthermore, the functional groups of MSW were found to contain carbonyl, aromatic, O-alkyl, and alkyl groups. In addition, elemental analysis has shown that MSW contains mostly cellulose, carbon and O-alkyl. (O + N)/C and O-alkyl/alkyl ratios have been reported to affect the alkyl-benzene sorption to MSW [26]. This phenomenon suggests that it might show a similar trend for the potential released metal and compound sorption to MSW in this study. Generally, three isotherm adsorption models could be used to predict the potential sorption of released ions to the MSW including the Freundlich equation, the Langmuir equation, and the BET equation. These isotherm adsorption models for the metals and ions released from the MSW suggest that they might affect the anaerobic digestion of MSW in landfill. However, these sorption mechanisms need further investigation.

4. Conclusions

The leachates from BA- and FA-amended lysimeters showed that they contained large amounts of Ca, K, Mg, and Na. The release of these alkali metals could contribute to the higher ANC

and provide the necessary alkalinity. Alkaline conditions are suitable for anaerobic processes such as landfill biostabilization, soil amendment, and other utilization. This phenomenon of higher alkali metal release was further verified and was consistent with the results of BA and FA acid titration curves. Furthermore, heavy metal release is of great concern due to its potential detrimental effects when used for landfill cover and subsequent risks to human health and the ecological environment. Except for Pb, the five metals of Cd, Cr, Cu, Ni, and Zn released in the lysimeters were found below the inhibitory concentrations for anaerobic process. Thus, they might serve as micronutrients rather than as inhibitory toxicants. These released concentrations were also found to be below the Taiwan groundwater regulatory standard. This potential lower release might be due to MSW adsorption as compared to that exposed with the deionized water. However, the use of BA and FA in landfill cover and other utilization still needs further investigation, and stringent regulations and strategies still need to be developed to prevent the potential risks to human health and the ecological environment.

Acknowledgement

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