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Biotoxicity evaluation of fly ash and bottom ash from different municipal solid waste incinerators

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ABSTRACT

Different types of municipal solid waste incinerator (MSWI) fly and bottom ash were extracted by TCLP and PBET procedures. The biotoxicity of the leachate of fly ash and bottom ash was evaluated by *Vibrio fischeri* light inhibition test. The results indicate the following: (1) The optimal solid/liquid ratio was 1:100 for PBET extraction because it had the highest Pb and Cu extractable mass from MSWI fly ash. (2) The extractable metal mass from both fly ash and bottom ash by PBET procedure was significantly higher than that by TCLP procedure. (3) The metal concentrations of fly ash leachate from a fluidized bed incinerator was lower than that from mass-burning and mass-burning combined with rotary kiln incinerator. (4) The TCLP and PBET leachate from all MSWI fly ash samples showed biotoxicity. Even though bottom ash is regarded as a non-hazardous material, its TCLP and PBET leachate also showed biotoxicity. The pH significantly influenced the biotoxicity of leachate.

1. Introduction

Incineration is a common method to treat municipal solid waste in Taiwan. The advantages of incineration include reduction of waste volume and mass, detoxification, and sterilization. There are approximately twenty-two municipal solid waste incinerators (MSWIs) in Taiwan and 5.52 million tons of waste was treated by incineration from 2003 to 2007 [1]. Municipal solid waste may contain heavy metals which can be vaporized at high temperature during incineration and then condensed on fly ash in a low temperature zone like an air pollutant control device (APCD). Therefore, MSWI fly ash may contain high concentrations of heavy metals. The toxicity characteristic leaching procedure (TCLP) is often used to evaluate the hazardous properties of solid waste. Fly ash is regarded as hazardous materials because their metal concentrations in TCLP leachate, especially for lead, often exceed the levels of Taiwan EPA regulations, [2–5]. Fly ash should be treated to reduce its damage to the environment. A great amount of MSWI bottom ash (0.83 million tons) was also generated in Taiwan from 2003 to 2007. Because its TCLP leachate concentration is below the EPA regulatory level, MSWI bottom ash is often classified as non-hazardous material. The biotoxicity of MSWI fly and bottom ash have been evaluated by different assays such as on cells [6], bacteria [7,8], daphnia [7,9], alga [7,9], and plants [10].

Many factors can influence the leaching behaviors of fly ash and bottom ash, including chemical speciation, particle size, minerals, and incinerator type [11–13]. Abbas et al. [14] indicate that the bottom ash from two different kinds of fluidized bed MSWIs was mainly composed of silicates and aluminum–silicate minerals with low solubility, and the fly ash contained considerable amounts of soluble salts of alkali and alkaline earth metals. Several studies [15–17] report that most particle size ranges of MSWI fly ash and bottom ash were 53–75 and 1680–4750 μm, respectively. Shim et al. [18] indicated that Pb concentrations in the TCLP leachate of bottom and fly ash often exceeded the regulatory level in two countries.

The physiologically based extraction test (PBET) employs simulated stomach juice as extracting agent to evaluate the bioaccessible pollutant contents of solid waste in the human gastrointestinal system [19]. Many studies have used this test to evaluate the effectiveness of remediation on contaminated soil [19–23]. However, researches on extraction of fly ash and bottom ash by PBET are limited. The *Vibrio fischeri* light inhibition test has been widely used to evaluate the biotoxicity of aqueous solution because of its high sensitivity, fast results, and relatively low cost [24,25]. It can also be applied to evaluate the biotoxicity of solid samples like soil and sediment [19]. Acid extraction agents are often used to extract metals from solid waste for evaluation of the biotoxicity of leachate by various bioassays [13,26,27].

The aim of this study was to estimate the bioavailability and biotoxicity of MSWI fly ash and bottom ash from different incineration processes. MSWI fly ash and bottom ash from different incineration processes were extracted by PBET. After extraction,

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the biotoxicity of PBET leachate was also analyzed. The comparison of the biotoxicity of TCLP and PBET leachate was discussed in this study.

2. Materials and method

2.1. Fly ash and bottom ash collection

The incinerator types in Taiwan include fluidized bed, mass-burning, and mass-burning combined with rotary kiln. Most of the incinerators in Taiwan are the mass-burning type. There is one fluidized bed incinerator and one mass-burning combined with rotary kiln incinerator in Taiwan. The wastes treated in these three incinerators are about 90% household waste and 10% business waste [28]. MSWI fly and bottom ash were collected from three types of MSWIs (MSWI-A, MSWI-B and MSWI-C) in Taiwan. The incinerator of MSWI-A is a fluidized bed and its treatment capacity is 95 tons/day. The combustion temperature is 800–950 °C and the APCD is bag filter (BF). Lime and powder activated carbon are used as injection agents. MSWI-B is a mass-burning type consisting of a three-step movable grate system (first, second, and combustion section). Its treatment capacity is 900 tons/day and the combustion temperature is 850–1050 °C. The APCD includes semi-dry scrubber (SD) and BF. The injection agents are activated carbon, lime, and urea. MSWI-C includes mass-burning and a rotary kiln and its capacity is 900 tons/day. The operational temperature ranges from 850 to 1050 °C. The solid waste is burned by mass-burning incinerator. The unburned solid waste is further combusted by rotary kiln. Its ACPD consists of SD and BF. Lime and powder activated carbon are used as APCD injection agents. The lime powder is injected into SD after the addition of activated carbon. The major components of the fly ash and bottom ash were SiO_2, CaO, Al_2O_3, Fe_2O_3, Na_2O, and MgO. Generally, the most abundant components in fly and bottom ash were CaO and SiO_2 (7–30%), except for a small amount of SiO_2 (0.1–2.3%) in MSWI-B ash. The next most abundant components were Fe_2O_3 (0.5–17%) and Na_2O (4–8%) [28]. In this study, the fly and bottom ashes were dried and then preserved in a plastic bag in an air-tight condition to avoid contact between the ashes and the ambient air (including CO_2). Therefore, the possibility of carbonation reaction should be very low. The leached mass of fly and bottom ashes should be unchanged during storage

2.2. The TCLP and PBET procedures

The TCLP test was conducted according to the regulations of Taiwan EPA. The TCLP extracting agent was acetic acid, and pH was about 2.88. The solid/liquid (S/L) ratio of TCLP was kept at 1:20. The total extracting volume and time were 100 mL and 18 h, respectively.

The PBET procedure was according to that described by Ruby et al. [19]. Gastric solution was prepared by adding 1.25 g of pepsin (activity of 800–2500 units/mg), 0.50 g of citrate, 0.50 g of malate, 0.42 mL lactic acid, and 0.5 mL acetic acid into 1 L distilled and deionized water. The pH of the gastric solution was 2.0 and was adjusted by 12N HCl. The total volume of stomach solution was 40 mL. Different solid/liquid ratios (S/L ratio = 1:5, 1:20, and 1:100) were used. The MSWI fly ash and bottom ash were added to the stomach solution in a polyethylene vessel and mixed in a temperature-controlled water bath shaker (37 °C). Ten millilitre of samples were collected at 0.5, 1.0, and 2.0 h respectively. After each sampling, 10 mL of extracting agent was added to the vessel to maintain the total extraction volume of 40 mL. The solids and liquid were separated by centrifugation at 3000 rpm for 10 min. The concentrations of lead, cadmium, and copper were analyzed by ICP-OES. The analysis of samples in this study was triplicate and the measure values were averaged. The analysis instrument in this study was ICP-OES. The r^2 value of calibration curve of Pb, Cu, and Cd were higher than 0.995. Reagent blank control was also conducted. Because the S/L ratios for TCLP and PBET procedures might be different, the metal concentrations of fly ash and bottom ash that were extracted by the TCLP and PBET procedures were also provided to compare the extract ability of these two procedures.

2.3. V. fischeri light inhibition test

The marine luminescent bacteria *V. fischeri* (NRRL B-11117), obtained from DSMZ (Germany), was employed to evaluate the biotoxicity of TCLP and PBET leachate of fly ash and bottom ash. The cultivation of luminescent bacteria and biotoxicity evaluation procedure were according to ISO 11348-1[Water quality-determination of the inhibitory effect of water samples on the light emission of *V. fischeri* (Luminescent bacteria test)][29]. The biotoxicity of leachate before and after pH adjustment (pH 6.2) was evaluated. *V. fischeri* was exposed to the leachate samples for 5, 15, and 30 min as determined by a luminometer at 15 °C. Phenol was used as the positive control with EC_{50} ranging from 13 to 26 mg L^{-1}. Biotoxicity was expressed as the light inhibition ratio and was calculated as follows (ISO 11348-1):

$$\text{Inhibition ratio}(\%) = \frac{I_0 \times f_k - I_f}{I_0 \times f_k} \times 100\%$$

where: I_0: the luminescence intensity of the control sample at $t = 0$ min, f_k: the correction factor at $t = 5$ min, $f_k = I_k/I_0$, I_k: the luminescence intensity in the control sample at $t = 5$ min, I_f: the luminescence intensity of the sample at $t = 5$ min.

3. Result and discussion

3.1. The optimum solid/liquid ratio of PBET

In our previous study, we indicated that TCLP leachate of MSWI-C fly ash contained the highest metal concentrations among these three fly ash samples [28]. Therefore, the MSWI-C fly ash was selected to determine the optimum S/L ratio used for PBET extraction. The volume of extracting agent was 40 mL. The effects of S/L ratios on metal mass extraction from fly ash were evaluated. Fig. 1a shows that the highest mass of Pb was extracted from fly ash at S/L ratio of 1:100. The extracted Pb mass decreased with an increase of sampling time. This is because after each sampling, the extraction volume was replenished with 10 mL of gastric solution, resulting in the dilution of extractable Pb in the leachate. The same trends were observed at the S/L ratio of 1:5 and 1:20. In this study, a higher Pb mass was extracted at lower S/L ratio, even when less fly ash was used. It may be that at higher S/L ratio more fly ash matrix (anions and cations, like Cl^-, $NO_3{}^-$, $SO_4{}^{-2}$, Cd^{+2}, and Cu^{+2}) dissolved in the solution and interfered with the dissolution of Pb. Similar trends were found for PBET extraction of Cu from fly ash (Fig. 1b). In contrast, the highest Cd extractable mass was at S/L ratio of 1:5. This may be because Cd was easily extracted by PBET and Cd extraction was less interrupted by fly ash matrix dissolved in the leachate (Fig. 1c).

The biotoxicity of fly ash PBET leachate at different S/L ratios was also evaluated. First, the background biotoxicity of extracting agents used in the TCLP and PBET procedures was assessed. I_5, I_{15}, and I_{30} were the inhibition ratios of either the extracting agent or leachate measured at 5, 15, and 30 min exposure time, respectively. In this study, I_5 and I_{30} represented the acute and chronic toxicity, respectively. Table 1 shows that the I_5, I_{15}, and I_{30} of TCLP extracting agent were 5.23%, 9.96%, and 15.0%, respectively and the I_5, I_{15}, and I_{30} of PBET extracting agent were 1.32%, 1.98%, and 10.7%, respectively. The background biotoxicity of both extracting agents was insignifi-

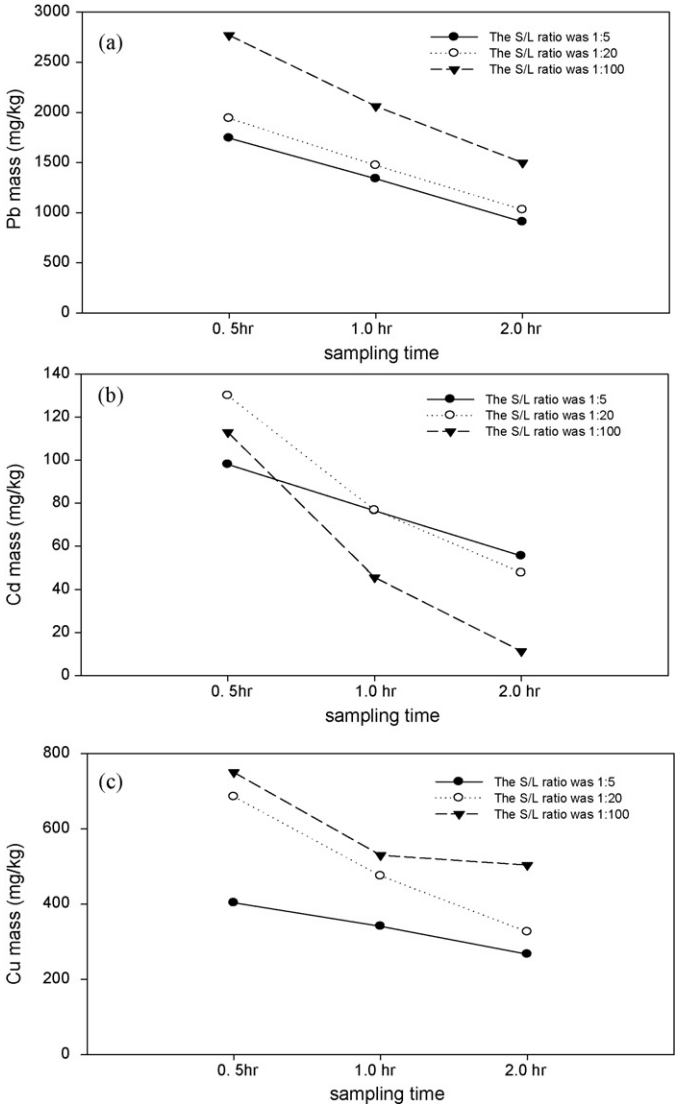

Fig. 1. The extracted metal mass of MSWI-C fly ash with PBET procedure at different solid/liquid ratios (a) Pb (b) Cd (c) Cu. Total extracting volume was 40 mL.

cant. The extracting agents were suitable for biotoxicity evaluation of both TCLP and PBET leachate. Generally, I_5 can be used to measure the acute biotoxicity of solution in the *V. fischeri* light inhibition test. The difference between I_5 and I_{30} can be used to reflect chronic toxicity in the leachate.

Table 2 exhibits the light inhibition of PBET leachate at different S/L ratios. The leachate at all applied S/L ratios (1:5–1:100) showed toxic. The biotoxicity of leachate increased with the S/L ratio. Table 2 also shows a high acute toxicity of the 0.5 hr leachate at S/L = 1:5 and 1:20 (I_5 = 30–85%). The high acute toxicity may have been caused by matrix substrates (anions and cations) or organic matters. The presence of acute toxicity would interfere with the measurement of chronic contaminants like heavy metals in the leachate. In contrast, when the S/L ratio decreased to 1:100, the I_5 of 0.5 h-leachate was close to 0. At this S/L ratio (1:100), the interference of the acute toxicity can be ignored; the biotoxicity of leachate is mainly caused by the chronic toxic substrates (I_{30} = 58%). The highest Pb and Cu mass could be extracted from fly ash at S/L ratio of 1:100, and the interference of acute toxic matrix substrates was insignificant. The S/L ratio of 1:100 was selected for sequential PBET extraction. The extraction time was 2 h without sub-samples being taken during the leaching period.

Table 1
The light inhibition ratio of TCLP and PBET extracting agents.

	Light inhibition ratio (%)		
	I_5	I_{15}	I_{30}
Blank of TCLP leachate	5.23	9.69	15.0
Blank of PBET leachate	1.32	1.98	10.7

Table 2
The light inhibition ratio of PBET leachate at different S/L extraction ratios.

	Light inhibition ratio (%)		
	I_5	I_{15}	I_{30}
S/L = 1:5			
0.5 h sample	85.2	89.5	96.3
1.0 h sample	81.7	98.1	100
2.0 h sample	60.0	88.1	97.2
S/L = 1:20			
0.5 h sample	30.1	51.7	82.8
1.0 h sample	20.0	49.5	82.4
2.0 h sample	0.0	21.4	38.2
S/L = 1:100			
0.5 h sample	0.0	24.6	55.7
1.0 h sample	1.37	26.9	47.6
2.0 h sample	0.0	0.0	47.4

3.2. Extractable metal mass of fly ash and bottom ash by PBET and TCLP

The extractable metal mass of MSWI fly ash and bottom ash by the PBET and TCLP procedures was evaluated. Table 3 indicates that the extractable metal mass of fly ash was obviously higher than that of bottom ash for both the TCLP and the PBET procedures, except for the extractable Cu and Zn mass of bottom ash for the TCLP procedure, which were higher than those of fly ash. Even though heavy metals of solid waste are vaporized and condensed on MSWI fly ash during the combustion process, the high pHs (8.3–11.7) of fly ash TCLP leachate reduced Cu and Zn leaching from fly ash. Table 3 also shows that the extractable metal mass of fly ash and bottom ash in the PBET leachate was significantly higher than in the TCLP leachate from bottom ash of three MSWIs, especially for Cu and Pb. The higher extractable metal mass by PBET than by TCLP can be attributed to the low pH of the extracting agent in the PBET extract.

Metal concentrations in TCLP leachate of bottom ash from three MSWIs were lower than stipulated by EPA regulation (Cd: 1; Cu: 15 and Pb: 5 mg/L) and were regarded as non-hazardous materials. However, a much higher metal mass could be extracted by PBET than by TCLP, especially for Cu (81–558 mg/kg) and Pb (28–267 mg/kg). Even though bottom ash is classified as non-hazardous substrate, it should still be handled carefully and ingestion by residents should be avoided. Table 3 indicates that the pH adjustment from original condition to pH 6.2 did not significantly change metal concentrations in the TCLP and PBET leachate of fly ash and bottom ash, expect for Pb concentrations in the TCLP leachate of MSWI-B and -C fly ash.

3.3. Biotoxicity of PBET leachate and TCLP leachate

3.3.1. MSWI fly ash

The biotoxicity of TCLP and PBET leachate of fly ash and bottom ash was evaluated by *V. fischeri* light inhibition test. The biotoxicity of TCLP and PBET leachate of fly ash and bottom ash before and after pH adjustment was also compared. Fig. 2a shows that the I_5 for TCLP leachate of MSWI-A, B, and C fly ash was 2%, 100%, and 100%, respectively. The leachate of MSWI-B and C fly ash was acutely toxic.

Table 3
TCLP and PBET extractable metal concentrations of MSWI fly ash and bottom ash and the metal concentrations in the leachate.

	Metal concentration extracted by TCLP procedure (mg/kg)				Metal concentration extracted by PBET procedure (mg/kg)				Metal concentration in TCLP leachate (mg/L)					Metal concentrations in PBET leachate (mg/L)				
	Cd	Cu	Pb	Zn	Cd	Cu	Pb	Zn	pH	Cd	Cu	Pb	Zn	pH	Cd	Cu	Pb	Zn
Fly ash																		
Original																		
MSWI-A	N.D.	0.5	15	N.D.	0.85	1271	849	1757	8.3	N.D.	0.02	0.7	N.D.	2.0	0.01	12.7	8.5	17.6
MSWI-B	N.D.	1.0	161	39	2.05	940	2048	3270	11.7	N.D.	0.05	8.1	1.9	2.0	0.02	9.4	20.5	32.7
MSWI-C	N.D.	16.8	825	136	2.17	702	2168	3061	11.7	N.D.	0.84	41.3	6.8	2.0	0.02	7.0	21.7	30.6
pH adjusted																		
MSWI-A	0.48	0.2	15	N.D.	0.71	1147	708	1600	6.2	0.02	0.01	0.8	N.D.	6.2	0.01	11.5	7.1	16.0
MSWI-B	0.55	6.0	162	53	0.43	882	435	3128	6.2	0.03	0.30	8.1	2.7	6.2	N.D.	8.8	4.4	31.3
MSWI-C	N.D.	15.5	874	164	0.63	662	634	2759	6.2	N.D.	0.77	43.7	8.2	6.2	0.01	6.6	6.3	27.6
Bottom ash																		
Original																		
MSWI-A	N.D.	25.7	N.D.	467	0.03	81	28	283	5.7	N.D.	1.28	N.D.	23.3	2.0	N.D.	0.8	0.3	2.8
MSWI-B	N.D.	19.8	N.D.	43	0.27	558	267	1596	7.4	N.D.	0.99	N.D.	2.1	2.0	N.D.	5.6	2.7	16.0
MSWI-C	N.D.	182	1.3	418	0.06	262	59	243	7.8	N.D.	9.10	0.1	20.9	2.0	N.D.	2.6	0.6	2.4
pH adjusted																		
MSWI-A	N.D.	27.8	N.D	496	N.D.	70	27	250	6.2	N.D.	1.39	N.D	24.8	6.2	N.D.	0.7	0.3	2.5
MSWI-B	N.D.	23.0	N.D	66	N.D.	536	267	1479	6.2	N.D.	1.15	N.D.	3.3	6.2	N.D.	5.4	2.7	14.8
MSWI-C	N.D.	142	N.D	405	N.D.	240	53	228	6.2	N.D.	7.11	N.D.	20.3	6.2	N.D.	2.4	0.5	2.3

Fig. 2a and 2b indicate that the adjustment of pH of MSWI-A fly ash leachate from 8.3 to 6.2 (Table 3) significantly increased the biotoxicity (I_5 = 70%). This can be attributed to the fact that the decrease in leachate pH increased dissolution of toxic contaminants. In contrast, the adjustment of pH of leachate of MSWI-B and–C fly ash from pH 11.7 to 6.2 obviously reduced the leachate biotoxicity. It is clear that because the extreme alkali solutions (pH 11.7) were toxic to *V. fischeri*, Leachate pH was an important factor influencing leachate biotoxicity.

Fig. 2c illustrates that PBET leachate of all MSWI fly ash was highly biotoxic (I_5 = 100%).This is because extreme acidity of PBET leachate (pH 2) were toxic to *V. fischeri*. The increase in the leachate pH from 2 to 6.2 significantly decreased the biotoxicity of PBET leachate of all three fly ash samples (I_5 for MSWI-A, B, and C fly ash leachate was 27%, 44%, and 27.4%, respectively). Comparison of I_5 and I_{30} of PBET leachate of all fly ash showed that the I_{30} of PBET leachate of MSWI-B and MSWI-C were obviously higher than their I_5. This suggests that chronically toxic substrates were present in those two leahcates. It should be noted that a low solid/liquid ratio of 1/100 was used for PBET extraction and the leachate was toxic; the toxicity of the leachate may increase at high S/L ratio. Acute toxicity of the TCLP and PBET leachate of fly ash may be caused by anions/cations like Cl^-, NO_3^-, SO_4^{-2}, Cd^{+2},Cu^{+2}, and Pb^{+2} [8], phenolic compounds [30], and other toxic residues. As mentioned

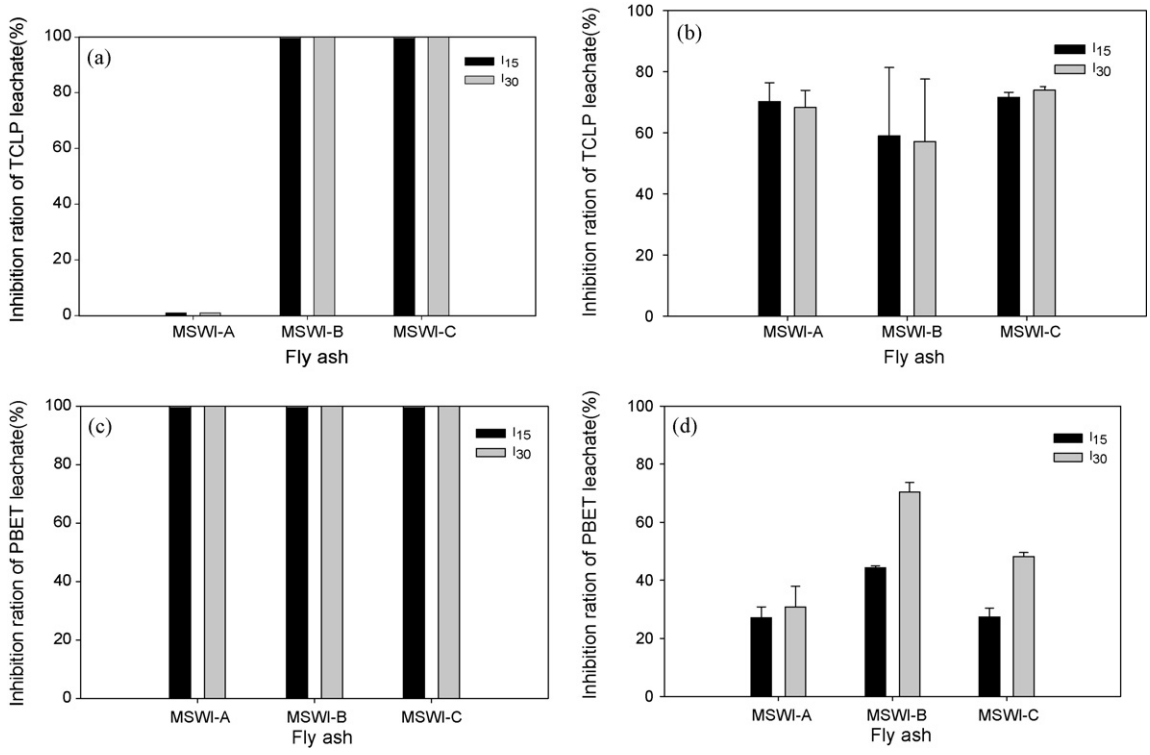

Fig. 2. The toxicity of MSWI fly ash with different extraction methods: TCLP leachate: (a) original solution (b) pH adjusted solution (pH 6.2); PBET extraction: (c) original (d) pH adjusted solution (pH 6.2). The S/L ratios for TCLP and PBET were 1:20 and 1:100, respectively.

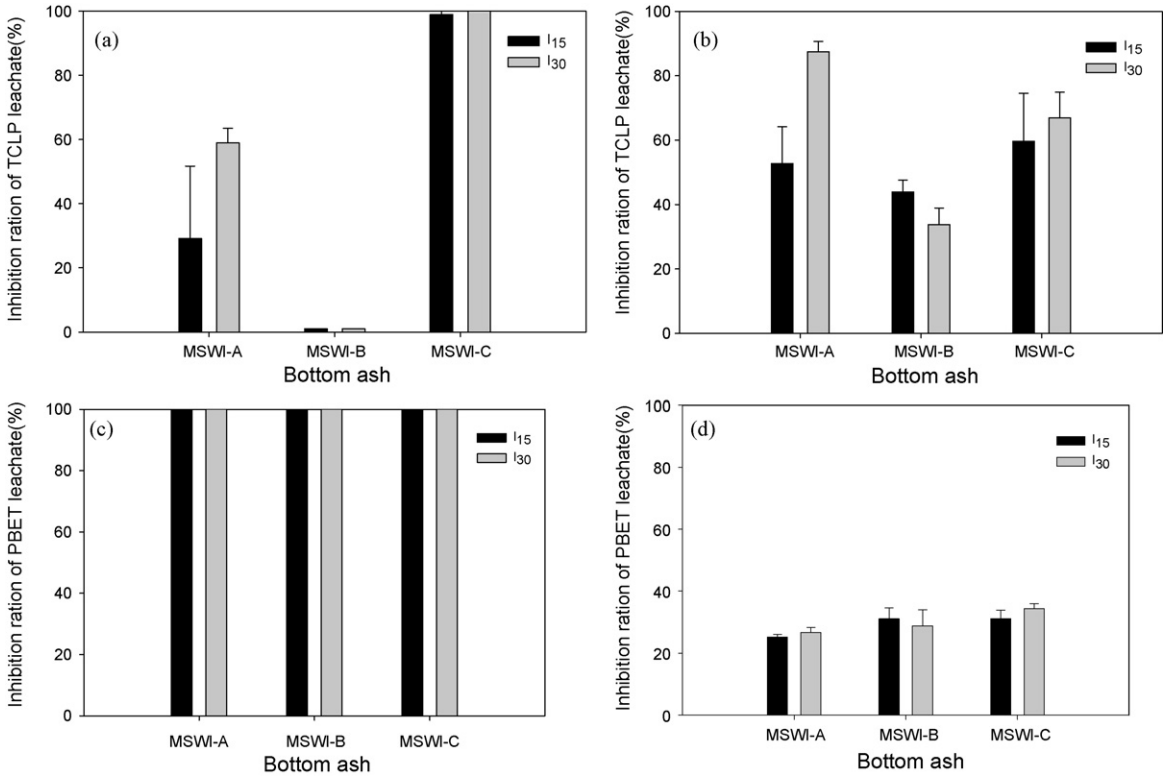

Fig. 3. The toxicity of MSWI bottom ash with different extraction methods: TCLP leachate: (a) original (b) pH adjusted solution (pH 6.2); PBET extraction: (c) original (d) pH adjusted solution (pH 6.2). The S/L ratios for TCLP and PBET were 1:20 and 1:100, respectively.

above, the difference between I_5 and I_{30} can be used to evaluate the influence of chronic substrates on light production of bioassay, *V. fischeri*.

3.3.2. MSWI bottom ash

Fig. 3a illustrates the biotoxicity of the original TCLP leachate from different MSWI bottom ash samples. Even though bottom ash is regarded as non-hazardous material, the TCLP leachate of bottom ash from MSWI-A and MSWI-C showed acute toxicity. The I_5 was 29% and 99% for MSWI-A and MSWI-C, respectively. Fig. 3b indicates that after pH adjustment, all three TCLP leachates of bottom ash were biotoxic. For example, when the pH of TCLP leachate of MSWI-B fly ash was adjusted from 7.43 to 6.2, the leachate biotoxicity significantly increased from 1% to 59%. This may be because the decrease in the leachate pH increases the dissolution of contaminants. In contrast, when the pH of TCLP leachate of MSWI-C fly ash was adjusted from 7.8 (original) to 6.2 (pH adjustment), the biotoxicity (I_5) of the leachate decreased from 100% to 60%. The difference between I_5 and I_{30} of the TCLP leachate from MSWI-A bottom ash indicates that it contained chronic toxicity (Fig. 3b). Fig. 3c illustrates the biotoxicity of PBET leachate from different MSWI bottom ash samples. The leachate of all three MSWI bottom ash samples had highly acute toxicity (I_5 = 100%). This is because the low pH of PBET leachate (pH 2) was toxic to *V. fischeri*. Fig. 3d indicates that when the leachate pH was adjusted from 2.0 to 6.2, the biotoxicity of PBET leachate from bottom ash was significantly reduced to 25%–34%. A comparison of I_5 and I_{30} of the pH adjusted PBET leachate of bottom ash indicated that the chronic toxicity was not significant for any of the PBET leachates of the bottom ash samples (Fig. 3d).

To conclude, the pH significantly influenced the biotoxicity of TCLP and PBET leachate of fly ash and bottom ash from these three MSWIs. Bioassy should be conducted to evaluate their leachate biotoxicity. Even though bottom ash is regarded as a non-hazardous material, its TCLP and PBET leachates show biotoxicity. Proper treatment of MSWI bottom ash is necessary when it is used as recycling material. For the possible applications of MSWI bottom ash, its biotoxicity should be considered in EPA regulation.

4. Conclusion

Based on the results of this study, we conclude the following:

(1) At solid/liquid ratio of 1:100, PBET extracted the highest Pb and Cu extractable mass from MSWI fly ash.
(2) The extractable metal masses of MSWI fly ash and bottom ash extracted by PBET were higher than those extracted by TCLP.
(3) The metal concentrations of fly ash from a fluidized bed incinerator was lower than those from mass-burning and burning mass combined with rotary kiln.
(4) The TCLP and PBET leachate from all MSWI fly ash samples showed biotoxicity (I_{30} = 57–74% for TCLP leachate and 31–70% for PBET leachate at pH 6.2). Even though bottom ash is regarded as a non-hazardous material, its TCLP and PBET leachate showed biotoxicity (I_{30} = 34–87% for TCLP leachate and 27–34% for PBET leachate at pH 6.2). The pH significantly influenced the biotoxicity of TCLP and PBET leachate.

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