

Chemical extraction of organic carbon to reduce the leaching potential risk from MSWI bottom ash

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Abstract

The performance of extraction solvents, including organic and inorganic solvents, for organic carbon extraction from municipal solid waste incinerator (MSWI) bottom ash was evaluated. The total carbon (TC) extracted was used to ascertain the efficiency of extraction solvents and the reduction of dissolved organic carbon (DOC) leaching potential was used to evaluate the capacity of solvents to minimize environmental impacts of MSWI bottom ash over short- and long-term considerations in landfill sites. Extract final pH value was a prominent parameter affecting TC extraction. The higher efficiency was obtained at the lower extract final pH and acid or neutral condition was necessary to achieve approximately 30% of TC extraction from bottom ash. On the basis of the results of TC extraction, the efficiency of organic carbon reduction was evaluated using organic carbon leaching potential. Hydrochloric acid was the best solvent to extract organic carbon in controlled pH conditions. Hydrochloric acid reduced the organic carbon leaching potential of MSWI bottom ash by about 68% at neutral leaching pH.

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1. Introduction

Incineration of municipal solid waste (MSW) is widely used to decrease the large volume of waste that the modern society produces [1]. In Japan, more than 70% of MSW is incinerated and the residue generated from this is then deposited in landfills. Incineration of MSW is a management option, which has the potential to reduce the solid waste volume by 90%. Municipal solid waste incinerator (MSWI) bottom ash represents about 80% of the residues from incinerated MSW and is considered a heterogeneous slag, which mainly consists of glass, magnetic and paramagnetic metals, minerals and ceramics [2]. MSWI bottom ash, however, also contains a fraction of unburned organic matter and organic byproducts, which contain various hazardous

organic substances [3,4] that may pose a threat to surface and groundwater quality [5].

The major environmental concern in relation to MSW disposal is the short- or long-term environmental impacts by leaching of harmful substances from landfill sites. Disposal methods must be accomplished in a sustainable manner [6]. Johansson and Bavel [1] found that the maximum polycyclic aromatic hydrocarbon concentration in a weathered bottom ash was higher than the generic guidelines for sensitive land use in Europe. Brunner et al. [7,8] and Belevi et al. [9] reported that concentrations of total organic carbon (TOC) in MSWI bottom ash leachates were in the range of 200–800 mg/l. Brocca et al. [10] found TOC levels even >2000 mg/l in incinerator ashes. European countries have been implementing regulations requiring that TOC in landfilled waste must be lower than 5% since the year 2000 [11]. Materials to be disposed of in such landfills should be almost chemically stable and have properties similar to the earth's crust. In Japan, waste management is regulated

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by the “Waste Disposal and Public Cleansing Law”, where the Ministry of Environment is responsible for setting up the standards pertaining to final disposal sites for MSW [12].

Several studies have been carried out to clarify the effects of pH on the leaching of heavy metals from incinerator residues, such as fly ash and bottom ash, through lysimeter and leaching tests by acid water solutions [13,14]. Tateda et al. [15] and Katsuoka et al. [16] investigated the removal of heavy metals from MSWI fly ash. The main methodologies described from these authors are the use of adsorbents, such as silica, alumina and activated carbon, the use of chelating agents, thermal treatment to remove organic hazardous substances (dioxins and furans) and heavy metals, as well as chemical extraction by inorganic acid solutions. Therefore, the behavior and interaction of heavy metals in incinerator residues have been studied by many researchers and well established. On the other hand, few studies have been reported on the behavior of organic compounds in MSWI bottom ash, especially, the effect of extraction solvent or solution pH on the removal of organic compounds.

According to the specific criteria developed by the US Environmental Protection Agency (US EPA) [17] for the evaluation of the toxicity of incinerator ashes, acetic acid is the best acid for heavy metal extraction from solid materials. Chang et al. [18] evaluated the ability of different extraction tests, i.e., toxicity characteristic leaching procedure (TCLP), extraction procedure (EP) and American Society for Testing and Materials (ASTM) Standard Procedure, aiming at extracting metals from incinerator bottom ash and chemical sludge. They found that extraction by TCLP and EP gave the highest metal concentration due to acetic acid effect. Nagib and Inoue [19] also used acetic acid for heavy metal extraction from MSWI fly ash. Nakamiya et al. [20] developed optimum washing conditions for contaminated soil with four to eight chlorinated dibenzo-*p*-dioxins and dibenzofurans. However, the use of an organic acid for organic carbon removal from incinerator ash has not been studied yet.

The purpose of this study is to evaluate the performance of extraction solvents, including organic and inorganic solvents, for organic carbon extraction from MSWI bottom ash. The total carbon extracted was used to ascertain the efficiency of extraction solvents and the reduction of organic carbon (DOC) leaching potential was used to evaluate the capacity of solvents to minimize environmental impacts of MSWI bottom ash over short- and long-term considerations in landfill sites.

2. Experimental

2.1. Sample preparation

Bottom ash was acquired from a Municipal Solid Waste Incinerator facility in Higashi Hiroshima City, Japan, in

March 2002. The plant incinerates 150 t/day of MSW and discharges 7 t/day of bottom ash. This MSW mainly consisted of 53% paper, 17% plastic, 2% wood and 21% food waste (7% incombustibles and other materials). The temperature in the combustion chamber was $>850^{\circ}\text{C}$. About 20 kg of bottom ash was taken from the ash pit, homogenized and an aliquot of 2 kg was crushed in a ball mill, sieved to obtain grain size between 450 and $105\ \mu\text{m}$ and dried at $100\text{--}105^{\circ}\text{C}$ for 24 h until two consecutive masses were identical. We have used $100\text{--}105^{\circ}\text{C}$ for sample drying, as this is the standard procedure for bottom ash treatment [21]. The final sample was stored in a glass bottle to be used for all experiments. The pH of bottom ash was determined by the US EPA Method 9045C [22].

2.2. Total carbon extraction from bottom ash

2.2.1. Solvent selection

Different organic and inorganic solutions were evaluated for the extraction of total carbon (TC) from bottom ash, in order to know the prominent factor for TC extraction from bottom ash. Extraction solvents were selected based on the regulatory extraction procedures developed by the US Environmental Protection Agency, such as extraction procedure (US EPA Method 1310A), toxicity characteristic leaching procedure (US EPA Method 1311) and automated soxhlet extraction (US EPA Method 3541).

Bottom ash samples (1 g) were placed in vial bottles and extraction solvents were added at a liquid to solid (L/S) ratio of 20 (20 ml solvent) and then continuously agitated on a rotary shaker at 30 rpm (model MR-5, Iuchi) for 24 h at 20°C . The L/S ratio was fixed at 20 based on the US EPA regulatory leaching tests [17] and according to Vehlow [23]. Leaching at higher L/S ratios (>10) are mainly characterized by solubility-controlled substances release and the chemical equilibrium between solid and liquid phase is generally attained [6]. Table 1 shows the extraction solvents and the conditions used in this study. After extraction, the solid fractions were separated from the mixtures by filtration through $1.2\ \mu\text{m}$ membrane filters (Whatman GF/C), washed by ultra pure water and dried at $100\text{--}105^{\circ}\text{C}$ for 24 h to remove residual organic solvent. This drying treatment condition was selected based on the methodology proposed by Zhang et al. [21] and Bai et al. [24] for the analysis of total carbohydrates

Table 1
Conditions of extraction solvents (solutions) for total carbon extraction from bottom ash

Extraction solvent	Condition/concentration	Final pH
Acetic acid	pH 3.0/60 mM	9.4
Hexane/acetone	pH 3.1/50% (v/v)	9.9
Ethanol/water	pH 6.0/80% (v/v)	11.6
Hydrochloric acid (HCl)	pH 3.0/1 mM	11.8
Sodium hydroxide (NaOH)	pH 10.0/0.1 mM	13.2
Water (H ₂ O)	pH 5.6	12.0

content in bottom ash and drying shrinkage properties of ash residues, respectively. Total carbon content of the final material was determined by Elemental Analyzer (model MT-700, Yanaco). Total carbon extracted from bottom ash was calculated based on the remaining carbon in the final material and fresh ash.

2.2.2. Effects of extract final pH on TC extraction

Acetic acid solution was used to investigate the effects of pH and extraction time on TC extraction. Bottom ash samples were placed in glass beakers and acetic acid solution at different concentrations, ranging from 6.0 to 6.7×10^{-4} M ($2.0 < \text{pH} < 4.0$), were added at L/S ratio of 20. The mixtures were continuously stirred for 6, 12 and 24 h at 20 °C. The solid fractions were separated from the mixtures by filtration through 1.2 μm membrane filters (Whatman GF/C), washed by ultra pure water, to remove residual acid and dried at 100–105 °C for 24 h. Total carbon was determined by Elemental Analyzer as previously described.

2.3. Organic carbon leaching potential from treated bottom ash samples

The efficiency of solvents for organic carbon extraction was confirmed by their capacities to reduce the DOC leaching potential from fresh samples. Bottom ash was extracted by ultra pure water (pH 5.8) or solutions of acetic or hydrochloric acid controlled at pH 7.0. For hydrochloric or acetic acid extraction, automatic pH controller with acid addition (model TPD-51, TOKO Chemical Corporation) was used to keep the pH at 7.0. The mixtures were continuously stirred at 20 °C for 24 h. Up to 2 ml of acid was added to the system in order to keep the pH at 7.0. Therefore, the L/S ratio was in the range from 20 to 22. With regard to extraction by water, the mixture at L/S 20 was agitated in a shaker for 24 h (200 rpm) at 20 °C. The pH of the resulting solution from water extraction was 11.9. After 24 h of equilibration, the solid fractions were separated from the mixture as described before and submitted to the following evaluation of DOC leaching potential.

Inorganic buffer solutions were used to evaluate the DOC leaching potential from bottom ash. Phosphate ($\text{NaH}_2\text{PO}_4/\text{KOH}$) buffer solution at pH 4.5 and 7.4 and borate (NaB_2O_7) buffer solution at pH 9.6 were mixed with the solids obtained from the extractions by water, hydrochloric or acetic acid at L/S 20 and agitated in vial bottles for 24 h at 20 °C on a rotary shaker (model MR-5, Iuchi) at 30 rpm. Experiments were run for 24 h because kinetic experiments have shown that in general two steps can be observed in element leaching from bottom ash: a fast process, which is generally completed within 24 h, followed by a slow process that continues for more than a week [25]. Consequently, in this paper, we focus on the initial fast reactions between bottom ash and aqueous solution. After 24 h of equilibration, the resulting solutions were filtered through 0.45 μm membrane filters (Millipore HA) and analyzed for DOC by TOC Analyzer (TOC-5000, Shimadzu).

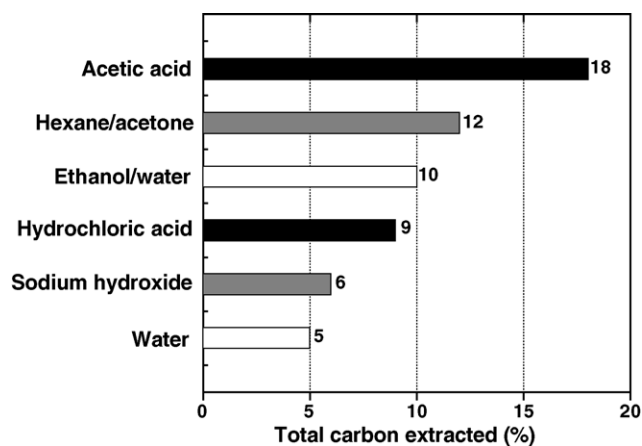


Fig. 1. Performance of extraction solvents for total carbon extraction from bottom ash.

3. Results and discussion

3.1. Performance of extraction solvents for TC extraction

Fig. 1 shows the performance of extraction solvents for TC extraction from bottom ash. The TC content in fresh bottom ash was approximately 7000 mg-C/kg-ash. Acetic acid showed the highest TC extraction (18%). The capacity of the solvents for TC extraction from bottom ash was: acetic acid > hexane/acetone > ethanol/water > hydrochloric acid > sodium hydroxide > water.

Extraction by acetic acid resulted in final pH value of 9.4 and it was the lowest among all solvents (Table 1). This fact suggests that the higher efficiency was due to the lower extract final pH. It seems that acetic acid introduces some additional buffering effects during extraction in comparison with other acids, because initial pH values were the same between acetic acid and hydrochloric acid solutions [19,26]. Moreover, the bottom ash pH was 12.8, determined by the US EPA Method 9045C [23]. Our previous research [27] also showed that acetic acid was the most efficient solvent for TC extraction from MSWI bottom ash sampled at another plant. In the same research, among all inorganic acids (hydrochloric acid (HCl), H_2SO_4 and HNO_3) evaluated, HCl and H_2SO_4 showed relatively higher efficiency for TC extraction than HNO_3 owing to lower final extract pH. These results are corroborated by the observations of Chang et al. [18] and Wang et al. [28], who also found higher leaching of diverse organic and inorganic species from bottom ash at the lower solution pH.

3.2. Effects of extract final pH and extraction time on TC extraction

Total carbon extraction by acetic acid solution at distinct pH ranges was studied in order to clarify the importance of extraction pH on the TC extraction process. Fig. 2 shows the percentage of extracted TC with time of extraction by

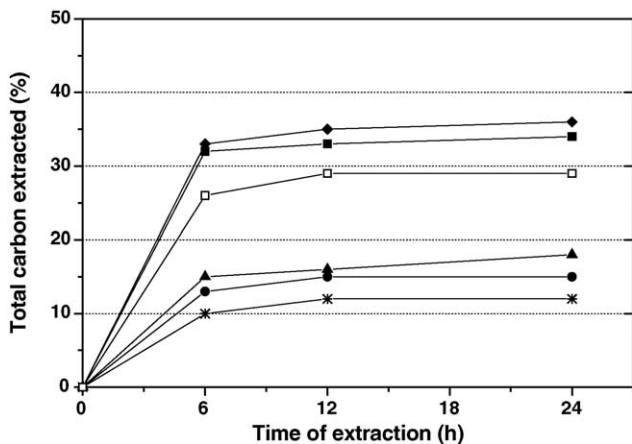


Fig. 2. Total carbon extraction along the time of extraction: (◆) 6 M (initial pH 2.0); (■) 0.6 M (initial pH 2.5); (▲) 0.06 M (initial pH 3.0); (●) 6×10^{-3} M (initial pH 3.5); (✱) 6×10^{-4} M (initial pH 4.0); (□) controlled pH at 7.0.

acetic acid solution at several concentrations. Total carbon extraction leveled off at about 6 h after the start of extraction. Extractions of 36 and 34% were achieved at initial pH of 2.0 (6.0 M) and 2.5 (0.6 M), respectively. Nevertheless, only 12, 15 and 18% were extracted at initial pH of 3.0 (0.06 M), 3.5 (6×10^{-3} M) and 4.0 (6×10^{-4} M), respectively. Extraction controlled at pH 7.0 was able to extract around 29% of TC from bottom ash. These results suggest that extraction pH is a prominent factor for TC extraction.

Fig. 3 shows the effects of final pH on TC extraction from the data in Fig. 2. Final pH values in the acid-neutral zone showed higher efficiency of acetic acid, where approximately 30% of TC was extracted. Nonetheless, TC extraction was lower than 18% at pH values >9 (alkaline zone). Extract final pH considerably affected TC extraction and thus acid or neutral condition was necessary to achieve higher extraction efficiency.

These results may be due to the carbon species in bottom ash. Total carbon can be qualitatively divided into two frac-

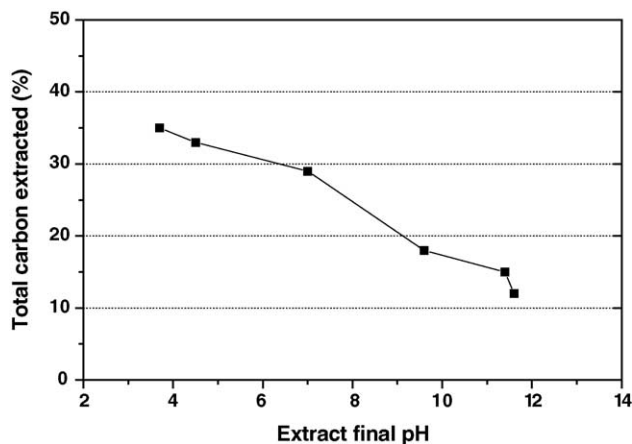


Fig. 3. Influence of extract final pH on total carbon extraction by acetic acid solution.

tions [29]: organic carbon (OC) and carbonate carbon (CC). Highest efficiency of acid extraction for TC extraction would be mainly attributed to higher extraction of CC by neutralization of carbonates in bottom ash, controlled by buffering reactions [30]. Recent research [31] suggested that organic carbon leaching from MSWI bottom ash was controlled by the presence of calcium containing minerals, which were identified by mineralogical measurements (X-ray diffraction). Consequently, the solubility of calcium and/or calcium species at different pH levels might explain the dependence of TC extraction efficiency on the extraction final pH [25]. Current experiments have corroborated these statements; OC was extracted in comparable levels irrespective of the pH changing in the alkaline-neutral range ($7 < \text{pH} < 11$). However, OC extraction was enhanced by about 40% with pH decrease from neutral to acid range (pH 2), mainly attributed to the dissolution of calcium containing species from MSWI bottom ash.

3.3. Efficiency of solvents to reduce organic carbon leaching potential from bottom ash

Fig. 4 shows the DOC leaching potential from fresh and the other three ash samples, obtained from extraction by water, acetic acid and hydrochloric acid evaluated at three distinct leaching pH values. Extraction of organic carbon by HCl or acetic acid was carried out at controlled pH 7 based on the optimum conditions obtained in the previous section and the two extraction acids were compared in order to clarify the differences between inorganic and organic acids for carbon extraction. Distinct pH values, such as acid (4), neutral (7) and alkaline (10) ranges, were selected for DOC leaching potential evaluation, so as to represent the major specific phases in landfill sites. In a landfill site, the leachate pH of bottom ash was alkaline at the beginning, drastically dropped down to the acid range and finally reached neutral or slightly alkaline

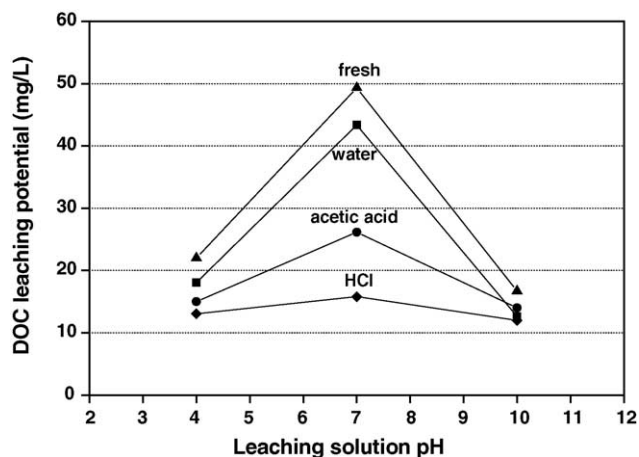


Fig. 4. Evaluation of solvent extraction for reduction of DOC leaching potential from fresh MSWI bottom ash. Samples: (▲) fresh bottom ash; (■) bottom ash extracted by water; (●) bottom ash extracted by acetic acid; (◆) bottom ash extracted by hydrochloric acid.

values, when the leachate contained higher organic carbon [32]. The acid condition might be due to uptake of atmospheric carbon oxide and acid rain precipitation after several years [24].

The maximum DOC leaching potential was noted at neutral pH for all samples. Water extraction slightly reduced the potential from fresh ash by 12%, showing 43 mg/l. On the other hand, acetic acid and HCl remarkably reduced the potential from fresh bottom ash by about 47 and 68%, respectively. The remaining solids presented potential of 26 and 16 mg/l, respectively. The extraction by HCl showed the highest reduction of DOC leaching potential at pH 7. Ferrari et al. [29] also found that the majority of organic carbon in bottom ash cannot be extracted either by water or alkaline solution. Zhang et al. [21] also used water for evaluation of extractable organic carbon fraction in bottom ash. However, the authors had to use HCl to extract the hardly leached components, which comprised 40–80% of the organic matter in bottom ash.

The potentials at acid and alkaline pH were also lower in the sample extracted by HCl than in the other samples. Indeed, the difference in the DOC leaching potential among leaching pH of 4, 7 and 10 was smaller than other samples; approximately, a straight profile was obtained. This was confirmed by laboratory experiments showing that pre-washing of bottom ash by HCl reduced the time of bottom ash to achieve the condition of “final storage quality” or “stabilization” [9] by about 80% in a simulated landfill situation [33]. Moreover, it was estimated that the leachate from washed bottom ash would satisfy with the guidelines for waste management in Japan within 25% of the time required for fresh ash. These results show that HCl is the best solvent to reduce the leaching potential and that pre-washing of bottom ash by HCl would result in a significant minimization of organic carbon leaching over short- and long-term considerations.

Regarding the differences between HCl and acetic acid for organic carbon extraction, one possibility is the remaining extraction solution (acetic acid) in the sample, which increases the DOC leaching potential. Acetic acid was actually detected by chromatography in the leachate from a sample extracted by acetic acid. Another possibility is the difference of calcium solubility as discussed in the previous section; the solubility of calcium chloride (3.8 mol/l at 20 °C) is higher than that of calcium acetate (1.6 mol/l at 20 °C) [34].

4. Conclusions

The performance of solvents for total carbon extraction from MSWI bottom ash was studied and it was found that the extract final pH value was a prominent parameter affecting total carbon extraction. The higher efficiency was obtained at the lower extract final pH and acid or neutral condition was necessary to achieve approximately 30% of total carbon extraction from bottom ash. Furthermore, hydrochloric acid

was the best solvent to extract organic carbon in controlled pH conditions. Hydrochloric acid reduced the organic carbon leaching potential of MSWI bottom ash by about 68% at neutral leaching pH.

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