

## Organic carbon leaching behavior from incinerator bottom ash

A.L. Guimaraes<sup>a,\*</sup>, T. Okuda<sup>b</sup>, W. Nishijima<sup>b</sup>, M. Okada<sup>a</sup>

<sup>a</sup> Department of Material Science and Chemical Engineering, Graduate School of Engineering, Hiroshima University, 1-4-1 Kagamiyama, Higashi Hiroshima, Hiroshima 739-8527, Japan

<sup>b</sup> Environmental Research and Management Center, Hiroshima University, 1-5-3 Kagamiyama, Higashi Hiroshima, Hiroshima 739-8513, Japan

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### Abstract

The understanding of the leaching behavior of organic carbon from incinerator bottom ash is an important aspect for the control of organic carbon emissions from landfills in order to minimize their potential risk to the environment. The leaching behavior of organic carbon from incinerator bottom ash samples, obtained from two different solid waste sources, as well as the effects of calcium (Ca) on organic carbon (DOC) leaching associated with pH were therefore investigated in this paper. The effect of pH on the leaching of DOC and other major elements from bottom ash was ascertained through pH-stat leaching experiments. Column leaching experiments were carried out to evaluate the dependence of the leached amount of DOC on Ca leaching. It was found that the bottom ash produced by incineration of municipal solid waste (MSW) was composed of two DOC fractions: DOC leached independent (fraction I) of Ca leaching, observed at alkaline–neutral pH, and DOC leached dependent (fraction II) on Ca leaching, observed at acid pH. However, the bottom ash produced by incineration of industrial solid waste (ISW) was composed of only DOC fraction I. The addition of calcium oxide during incineration and the differences in the paper to plastic ratio in the physical composition of the solid wastes incinerated would explain the distinct organic carbon leaching behaviors of bottom ash samples.

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### 1. Introduction

Municipal solid waste (MSW) is being incinerated in increasing quantities, leaving behind large amounts of bottom ash enriched with potentially harmful elements relative to soils and sediments [1]. MSW incinerator (MSWI) bottom ash makes up roughly 90% of MSWI residues [2]. The matrix elements of MSWI bottom ash are comparable in concentration with igneous rocks, the main components being Ca, Si, Al, Fe, Na, Mg and K [3]. MSWI bottom ash, however, also contains a fraction of unburned organic matter and organic by-products, which contains various hazardous organic substances [4,5], totaling approximately 4% w/w in concentration, and there is a concern that their release may pose a threat to surface and groundwater quality [6]. The material can be regarded as a potential risk to the environment, depending on the leaching of these contaminants [7].

The short- and long-term environmental impacts of bottom ash from MSW incineration have long been the subject of studies [8]. The emphasis of these studies is, generally, on the thermodynamics and kinetics of inorganic reactions. However, organic matter in bottom ash is responsible for increasing biochemical oxygen demand (BOD) and chemical oxygen demand (COD) in the leachate from landfill sites. Several studies [9–11] have reported that concentrations of total organic carbon (TOC) in MSWI bottom ash leachates were in the range of 200–800 mg/L. According to the guidelines for waste management in Japan, the leachate from landfills has to comply with the quality requirements defined by the “Waste Disposal and Public Cleansing Law”, which establishes BOD and COD limitations of 60 and 90 mg/L, respectively [12]. As a result, long-term treatment of the leachate from landfills to reduce BOD and COD is required. In this context, the leaching of organic carbon from MSWI bottom ash is of growing concern in Japan.

The understanding of the leaching behavior and main processes occurring with organic carbon in MSWI bottom ash is an important aspect for the control of organic carbon emissions from landfills. To address this problem, previous studies

\* Corresponding author. Tel.: +81 824 24 7622; fax: +81 824 24 7622.  
E-mail address: [alguim@hiroshima-u.ac.jp](mailto:alguim@hiroshima-u.ac.jp) (A.L. Guimaraes).

have examined the extraction of organic carbon from bottom ash to reduce the leaching potential from landfills [13]. It was demonstrated that pH value was a prominent parameter for the extraction; acid or neutral condition was necessary to achieve higher extraction efficiency. The authors also observed a noticeable effect of acid washing for organic carbon extraction in samples originated from distinct MSW incineration plants.

Organic carbon leaching from MSWI bottom ash may also be related to mineral leaching, which results from interplay of a number of different processes. Extensive mineralogical studies have been done on primary mineral identification in incinerator residues. Chimenos et al. [14] identified various minerals such as calcite ( $\text{CaCO}_3$ ), ettringite ( $\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12}\cdot 26(\text{H}_2\text{O})$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), quartz ( $\text{SiO}_2$ ), gypsum ( $\text{CaSO}_4\cdot 2(\text{H}_2\text{O})$ ), and silicates by mineralogical studies (X-ray diffraction). Johnson and Furrer [15] also detected minerals like portlandite ( $\text{Ca}(\text{OH})_2$ ), other soluble Ca salts such as ettringite and calcite, which play a predominant role in the buffering of leachate solutions. Calcite becomes the predominant buffering constituent in time as soluble basic Ca salts or forming relatively insoluble minerals such as clay. Meima and Comans [3] reported that dissolution/precipitation reactions controlled the leaching of major elements from waste materials such as MSWI bottom ash. Ca minerals in MSWI bottom ash mainly control leachate pH, which in turn has been identified as a major parameter controlling the leaching of many elements. Johnson et al. [16] confirmed that the acid neutralizing capacity of MSWI bottom ash is dominated by Ca minerals. It has been a common understanding among all authors that pH controls salt solubility and calcium carbonate solubility increases as pH decreases. However, the influence of pH on salts solubility may also affect other calcium species or complexes in the solid matrix. Our previous research [17] showed that organic carbon leaching from a sample originated by incineration of industrial solid waste (ISW) was independent of leaching pH conditions. These findings indicated that the ISW incinerator sample was mainly composed of leachable organic carbon independent of pH, and that organic carbon leaching was not related with Ca leaching. Nonetheless, the differences between MSW and ISW incinerator ash samples and the relationship of Ca with organic carbon leaching have not been explained yet.

The purpose of this paper is to investigate the leaching behavior of organic carbon from incinerator bottom ash samples obtained from two different solid waste sources (MSW and ISW incinerator facilities). The effect of Ca on organic carbon leaching associated with pH was also a focus of this paper.

## 2. Experimental

### 2.1. Bottom ash samples

Ash samples were taken from two distinct solid waste Incinerator Facilities in the Cities of Hiroshima and Fukuoka, Japan, in 2003. The incineration capacity of the plants is about 150 t/day of solid waste. Samples were abbreviated as M-ash (Hiroshima) and I-ash (Fukuoka) in this paper. M-ash consists of 100%

bottom ash originating from MSW incineration, while I-ash consists of a mixture of bottom ash (90%) and fly ash (10%), originating from ISW incineration. The incineration conditions were quite similar for both samples; the temperature ( $T$ ) in the combustion chambers was in the range  $1000^\circ\text{C} < T < 1200^\circ\text{C}$ . Major differences are in the physical composition of the solid waste incinerated; the MSW is mainly composed of 53% paper, 17% plastic and 21% food waste (9% incombustibles and other materials), whereas the ISW is composed of 8% paper, 64% plastic and 26% food waste (2% incombustibles and other materials)—and the addition of calcium oxide for MSW incineration. The addition of calcium oxide has been commonly applied in Japan in the case of MSW incineration to prevent the emissions of hazardous organic compounds, such as polycyclic aromatic hydrocarbons (PAH), polychlorinated biphenyls (PCB) and dioxins and benzofurans, into the atmosphere [18,19]. The composition of these two samples of solid waste are quite similar to those reported by Shim et al. [20], for samples collected from three different Incinerator Plants in Tokyo (Japan).

About 20 kg of bottom ash were taken from the two respective ash pits, homogenized and aliquots of 2 kg were crushed in centrifugal ball mills, sieved to obtain grain size between 450 and  $105\ \mu\text{m}$  and dried at  $105^\circ\text{C}$  for 24 h [21] until two consecutive masses were identical for both samples. The samples were stored in glass bottles to be used for all experiments. The concentration of Ca in bottom ash samples was determined by the United States Environmental Protection Agency (US EPA) Method 3050B [22]. The total carbon concentration (TOC) was determined by a pH-stat leaching test carried out at pH 7.0 [13]. The pH of bottom ash samples was determined by the US EPA Method 9045C [23].

### 2.2. Effect of pH on the leaching of organic carbon (DOC) and other major elements from bottom ash

Bottom ash samples were submitted to pH-stat leaching tests to find out the amount of DOC and other major elements leached as a function of pH. Hydrochloric acid (HCl) was used as the leaching solvent [13]. An automatic pH controller with the addition of acid (model TPD-51, TOKO Chemical Laboratories Corporation) was used to keep constant pH values. Bottom ash samples (10 g) were placed in glass beakers containing 1000 mL of leaching solution adjusted to pH 11, to reach a liquid to solid (L/S) ratio of 100. A high L/S ratio was employed to avoid the supersaturation of Ca with respect to other leachable species [16]. HCl was added to maintain the pH at 11, 9, 8, 7, 6, 4 or 2 and then the L/S ratio was kept in the range of 90–100 for all experiments. Mixtures were continuously stirred at  $20^\circ\text{C}$  for 2 h at each pH value. After 2 h of equilibration at each pH value, an aliquot of 10 ml was collected, filtered through a  $0.45\ \mu\text{m}$  membrane filter HA (MF-Millipore, Millipore Corporation) and the clear filtrate was analyzed for DOC by a TOC analyzer (TOC-5000, Shimadzu Corporation) and for Ca, Al and Si by an inductive coupled argon plasma atomic emission spectrometer—ICAP-AES (model ICAP-575II, Nippon Jarrell-Ash Corporation).

### 2.3. Dependence of the leached amount of organic carbon (DOC) on Ca leaching

Column leaching experiments were performed on the M-ash sample to simulate more realistic conditions of a landfill site [24]. A glass column, length 25 cm, internal diameter 2 cm, was filled with 10 g of ash and submitted to a two-step continuous leaching process. In the first step, ultra pure water at pH 5.8 continuously percolated through the column until DOC and Ca concentrations in the leachate presented negligible variations. Subsequently, HCl solution at pH 2 was applied in the second step. This second step was carried out to evaluate the dependence of the leached amount of DOC on Ca leaching from bottom ash in acid conditions. The experiments were carried out at 20 °C and using continuous flow of 1.5 cm<sup>3</sup> min<sup>-1</sup> maintained by a micro tube pump (model MP-3, Tokyo Rikakikai Corporation). Layers of quartz–wool (1 cm thick) were placed at the top and bottom of the column to ensure a uniform feed distribution and to prevent the formation of channels. Leachate samples were periodically collected, filtered through 0.45 μm membrane filters and analyzed for DOC, Ca, Al and Si. The pH of the leachate samples were measured using a pH meter (model HM-20P, DKK-TOA Corporation).

## 3. Results and discussion

### 3.1. Effect of pH on the leaching of organic carbon (DOC) and other major elements from bottom ash

The effect of pH on the leaching of organic carbon (DOC), Ca, Al and Si from M-ash and I-ash samples is shown in Fig. 1A and B, respectively. The leached amount of DOC from M-ash was almost indistinguishable, 520 mg-C/kg-ash, at the pH range from 11 to 6 (Fig. 1A). However, DOC leaching increased to 800 mg-C/kg-ash when the pH was lowered from 6 to 2. This suggests that DOC can be divided into two fractions: the DOC leached independent (fraction I) and dependent (fraction II) on the leaching pH. These results are in agreement with the findings of Dijkstra et al. [7], who also found that higher DOC leaching from a MSWI bottom ash took place at lower pH values (acid range). On the other hand, the leached amount of DOC from the I-ash was independent of the leaching pH (Fig. 1B); approximately 1600 mg-C/kg-ash of DOC was leached regardless of the pH range (fraction I in Fig. 1B).

Leaching of Ca was strongly dependent on the leaching pH for both ash samples. When the pH was lowered from alkaline to acid range, Ca leaching significantly increased. Approximately 90 and 42 g/kg-ash of Ca were leached at pH 2 from the M-ash and I-ash samples, respectively, while those at pH 11 were 6 and 4 g/kg-ash, respectively. Al and Si exhibited different leaching behavior from Ca; significant leaching from the M-ash sample was observed only at pH lower than 4, showing strong interaction with the solid matrix in the pH range from 11 to 6. This result is supported by the findings of Johnson et al. [16], where Si dissolution also occurred in acid pH (pH ≈ 3.5) [16]. Al leaching may be due to the decomposition of amorphous Al(OH)<sub>3</sub>, as reported by several other authors [5,14,25]. The I-ash sample presented

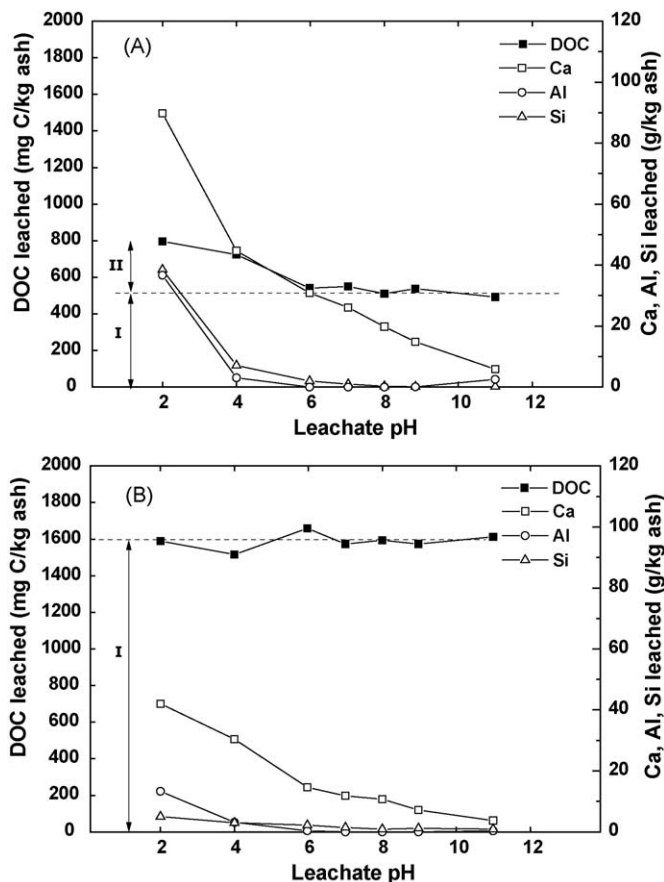


Fig. 1. Effect of pH on the leaching of organic carbon (DOC), Ca, Al and Si from bottom ash samples: (A) M-ash and (B) I-ash.

an analogous leaching trend, but much lower amounts of Al and Si were leached at pH 2; 13 and 5 g/kg-ash, respectively.

Fig. 2 shows the dependence of the leached amount of DOC on Ca leaching in M-ash and I-ash samples from the data presented in Fig. 1A and B. The dotted profile shows a positive correlation pattern ( $r = 0.93$ ) between Ca and DOC leached from M-ash (fraction II<sub>M-ash</sub>). Nevertheless, about 520 mg-C/kg-ash

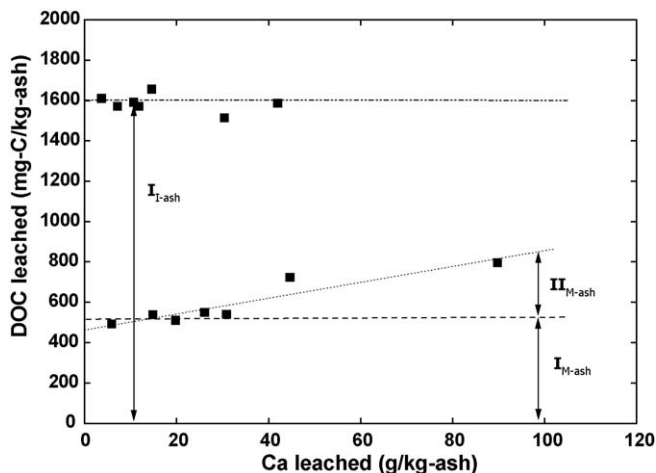


Fig. 2. Dependence of the leached amount of organic carbon (DOC) on calcium (Ca) leaching in M-ash and I-ash samples.

Table 1  
Concentrations of calcium (Ca) and total organic carbon (TOC) and pH in bottom ash samples

Bottom ash sample	pH	Concentration (g/kg ash)	
		Ca	TOC
M-ash	12.8	125	1.0
I-ash	12.2	55	2.1

of DOC, represented by the dashed profile (horizontal line), was leached without Ca leaching (fraction  $I_{M\text{-ash}}$ ). Table 1 shows the concentrations of Ca and TOC and pH in bottom ash samples. The far higher Ca/TOC ratio in M-ash (125), compared to I-ash (26), and the presence of alkaline species (calcium minerals), due to the ash pH values higher than 12 [14,16], indicates that DOC dependent on pH might form complexes with Ca; and would be leached with Ca at low pH conditions [26]. On the other hand, we did not find any relationship between DOC and Ca leaching in I-ash (fraction  $I_{I\text{-ash}}$ ). It is most likely that the DOC leached from the I-ash did not form complexes with Ca.

### 3.2. Behavior of organic carbon leaching from bottom ash samples

Column leaching experiments were carried out to confirm the relationship between DOC and Ca leaching from the M-ash sample, simulating more realistic environmental conditions of a landfill site. In most landfill sites, the pH of bottom ash is alkaline at the beginning, drops down and finally reaches neutral or slightly acid values by uptake of atmospheric carbon oxide and acid rain precipitation after several years. The M-ash was therefore submitted to the two-step continuous leaching process using water and an acid solution as described in the experimental section. Experiments were carried out only for the M-ash sample to evaluate the behavior of the two DOC fractions.

Fig. 3 shows the DOC, Ca, Al and Si leached from the two-step continuous leaching process. The DOC/Ca ratio and pH variation in the leachate during these experiments are shown in Fig. 4. They are shown as a function of the volume of leaching solution percolated through the bottom ash mass in the experi-

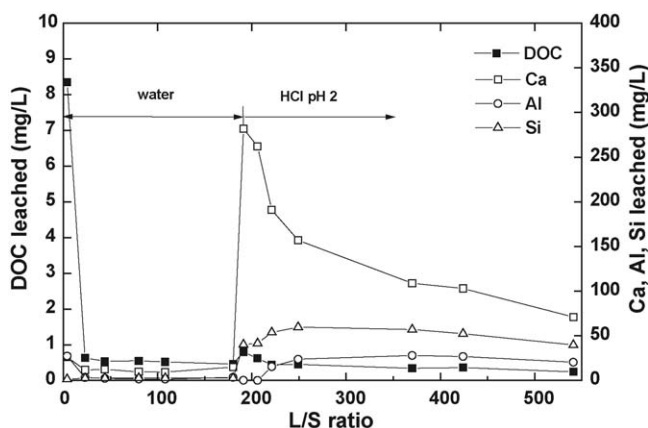


Fig. 3. DOC, Ca, Al and Si leached from the two-step continuous leaching process; water (first step), followed by HCl solution at pH 2 (second step).

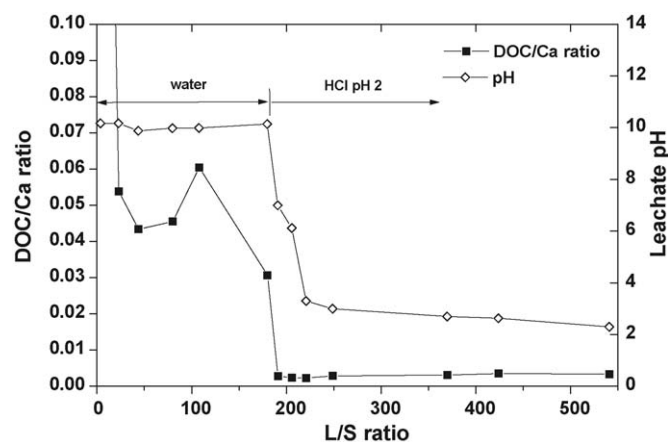


Fig. 4. DOC to Ca (DOC/Ca) ratio and pH variation in the leachate from the two-step continuous leaching process; water (first step), followed by HCl solution at pH 2 (second step).

mental column (L/S ratio – volume of leaching solution (ml)/10 g M-ash). The DOC leaching followed a typical leaching curve, in which the largest concentration was noted at the beginning of the experiment. This “first flush” scenario is mainly due to low L/S ratios [24,27]. Then, DOC leaching quickly leveled off after  $L/S = 25$ , at approximately 0.5 mg/L. Water percolated through the column produced alkaline leachates ( $pH \approx 10$ ) and then low amounts of Ca, Al and Si were leached regardless of L/S value, owing to their slower dissolution rate [8]. As shown in Fig. 4, the DOC/Ca ratio was high (0.320, not represented) at the beginning of the experiment ( $L/S < 25$ ), decreased quickly to around 0.040 and remained unstable during the percolation of water ( $L/S < 175$ ). The DOC/Ca ratio was much higher than that of the DOC fraction II (0.0037) in the pH-stat experiment (Fig. 2), which corresponds to the DOC dependent on Ca leaching. Moreover, the pH during the percolation of water was around 10, when the DOC fraction I independent of Ca leaching was leached in the pH-stat leaching experiment. This suggests that DOC leached by water would fall into fraction I. It should be noted, however, that the total amount of DOC leached by water was around 250 mg-C/kg-ash, which was far lower than that of the leached DOC fraction I (520 mg-C/kg-ash) in the pH-stat leaching experiment. This could be due to the change from water to the acid solution before the total leaching of fraction I was completed.

The leached amount of all elements increased when the HCl solution at pH 2 (second step of column experiment) was percolated through the column; Ca leaching significantly increased from 15 to 280 mg/L, whereas DOC leaching sharply increased from 0.4 to 0.8 mg/L (Fig. 3), when the pH decreased from 10.2 to values slightly lower than 7 (Fig. 4). When the pH was lowered to acid values, the DOC/Ca ratio rapidly decreased and remained constant at approximately 0.0030 until the end of the experiment ( $175 < L/S < 550$ ). The DOC/Ca ratio during the flow of HCl solution was similar to the one found during the leaching of DOC fraction II in Fig. 2 (0.0037). This indicates that the DOC fraction II (dependent on Ca leaching) was mainly leached at neutral and acid conditions produced by the percolation of the acid solution through the column. In addition to that, the DOC



fraction I would still be leached at neutral condition because the period of water percolation was not enough to totally leach the DOC fraction I. Regarding Si and Al, their leached amounts gradually increased and clearly showed different behavior from Ca and DOC. These elements did not seem to directly affect the DOC leaching from bottom ash samples.

The foremost organic compounds present in the extractable matter of MSWI bottom ash were reported to be carboxylic acids [28] and these negatively charged organic groups can be bound to cations like  $\text{Ca}^{+2}$ , possibly forming DOC–Ca complexes on the surface of bottom ash [26]. As the pH decreased to the acid range, these DOC–Ca complexes would dissociate and DOC would be leached with Ca [14,29,30]. DOC fraction II in our research would be the DOC which would form complexes with Ca. Furthermore, most of the organic matter in the two bottom ash samples investigated was not organic matter forming complexes with Ca (fraction II) but free organic matter (fraction I).

#### 4. Conclusions

The behavior of organic carbon leaching from incinerator bottom ash samples obtained from different solid waste sources and the effects of Ca on organic carbon leaching associated with pH were studied.

It was found that the leachable organic carbon (DOC) in the bottom ash samples was composed of two fractions: DOC leached independent (fraction I) of Ca leaching, observed at alkaline-neutral pH, and DOC leached dependent (fraction II) on Ca leaching, observed at acid pH. The bottom ash produced by incineration of a MSW composed of 53% paper, 17% plastic, 21% food waste (9% incombustibles and other materials), and the addition of calcium oxide, was composed by both DOC fractions and presented a calcium to total organic carbon (TOC) ratio (Ca/TOC) of 125. Nevertheless, the bottom ash produced by incineration of a ISW composed of 8% paper, 64% plastic and 26% food waste (2% incombustibles and other materials) was composed of only DOC fraction I and presented a Ca/TOC ratio of 26. In the MSW incinerator bottom ash, the fraction I would be leached first because of high pH conditions (bottom ash  $\text{pH} > 12$ ) and fraction II would be leached with pH decrease to neutral range afterwards. The fraction II was continuously leached at the same DOC–Ca ratio, likely caused by the formation of complexes of organic matter with Ca in bottom ash. The addition of calcium oxide during incineration as well as the differences in the paper to plastic ratio in the physical composition of the solid waste incinerated would explain why the two bottom ash samples presented distinct organic carbon leaching behaviors.

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