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Journal of Hazardous Materials 141 (2007) 793–802

Journal of Hazardous Materials

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# Heavy metal leaching from aerobic and anaerobic landfill bioreactors of co-disposed municipal solid waste incineration bottom ash and shredded low-organic residues

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Received 15 March 2005; received in revised form 21 July 2006; accepted 24 July 2006

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## Abstract

In this study, heavy metal leaching from aerobic and anaerobic landfill bioreactor test cells for co-disposed municipal solid waste incineration (MSWI) bottom ash and shredded low-organic residues has been investigated. Test cells were operated for 1 year. Heavy metals which were comparatively higher in leachate of aerobic cell were copper (Cu), lead (Pb), boron (B), zinc (Zn), manganese (Mn) and iron (Fe), and those apparently lower were aluminum (Al), arsenic (As), molybdenum (Mo), and vanadium (V). However, no significant release of heavy metals under aerobic conditions was observed compared to anaerobic and control cells. Furthermore, there was no meaningful correlation between oxidation–reduction potential (ORP) and heavy metal concentrations in the leachates although some researchers speculate that aeration may result in excessive heavy metal leaching. No meaningful correlation between dissolved organic carbon (DOC) and leaching of Cu and Pb was another interesting observation. The only heavy metal that exceeded the state discharge limits (10 mg/l, to be enforced after April 2005) in the aerobic cell leachate samples was boron and there was no correlation between boron leaching and ORP. Higher B levels in aerobic cell should be due to comparatively lower pH values in this cell. However, it is anticipated that this slightly increased concentrations of B (maximum 25 mg/l) will not create a risk for bioreactor operation; rather it should be beneficial for long-term stability of the landfill through faster washout. It was concluded that aerobization of landfills of heavy metal rich MSWI bottom ash and shredded residues is possible with no dramatic increase in heavy metals in the leachate.

*Keywords:* Heavy metal; Leaching; Landfill bioreactor; Aerobic landfill; Bottom ash

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## 1. Introduction

Landfills in Japan currently receive MSWI residues and shredded low-organic residues as main inputs. However, incineration residues and other low-organic wastes originating from recycling activities are not completely stabilized, and therefore do not comply with the “final storage quality”. Significant amounts of organic carbon, heavy metals and toxic organic pollutants are emitted to the environment via leachate and gas phases, requiring extensive treatment and monitoring.

On the other hand, European countries landfill directive requires low-organic wastes to be landfilled, and mechanical–biological pretreatment (MBP) process might have limitations for meeting the required organic carbon criteria. Accordingly, the share of incineration in European countries is also expected to rise in the future. This will result in increased amount of MSWI residues going to landfills creating higher potential of heavy metal leaching from these sites. Heavy metal leaching from the landfills is an important process due to the toxicity on the environment and human health.

Organic matter creates a highly reactive landfill that without additional measures will result in methane emissions long-after the landfill has been completed. It also leads to the formation of DOC capable of mobilizing metals and organic pollutants. Therefore, if low leachate concentration of metals and other constituents is aimed, then DOC levels should be decreased as quickly as possible [1].

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doi:10.1016/j.jhazmat.2006.07.042

Landfill bioreactor applications have been investigated in recent years to achieve a faster biodegradation of organic matter. Earlier studies have concentrated on anaerobic landfill bioreactor applications with a secondary target of methane recovery and electricity generation. In recent years, aerobic landfill bioreactors are gaining attraction due to faster biodegradation and elimination of fugitive methane emissions.

Sustainable landfills require landfill bioreactors for accelerated stabilization of landfilled wastes. Anaerobic bioreactor alternative is not well suited for landfills in which mostly low-organic wastes such as MSWI bottom ash and residues from recycling due to insignificance of methane generation for possible electricity production and slow reaction rates of biodegradation of organic matter. Aerobic landfill bioreactors promise a faster stabilization rates for achieving final storage quality. However, it is feared that excessive heavy metal leaching may occur under oxidized conditions. It becomes more important for the landfills which received MSWI residues due to high heavy metal contents of these wastes.

Under anaerobic conditions the majority of metals are bound in carbonate-, hydroxide- or in metal–sulfide complexes. By aeration, the mobility of metals in the waste matrix represents the transition from these stable binding forms to soluble or mobilized species of ions, complexes or other compounds. These mobile forms can be transported to groundwater via leachate [2]. However, the leaching behavior of heavy metals under aerobic conditions is not well known. Only a few studies have been reported for aerobic landfill bioreactors of municipal solid waste, and mainly for laboratory scale lysimeters [2–4]. Cossu et al. [5] have studied the impact of MSWI bottom ashes (10%) co-disposed with MBP waste (90%) on heavy metal leaching in column tests. They have observed slight increase in only Cr and Cu after starting intermittent and continuous air injection into the columns following 52 days of anaerobic operation, though the increases were almost ignorable (Cr from 0.001 to 0.01 mg/l, Cu from 2–3 to 4–5 mg/l level).

There is a need to understand the behavior of heavy metals in aerobic landfill bioreactors where MSWI bottom ashes are co-disposed at higher ratios. To our knowledge, there is no full-scale landfill aerobic bioreactor study for co-disposed MSWI residues and low-organic wastes at almost similar proportions. The only study reported is a test-cell investigation by the authors of this paper [6,7]. However, leaching behavior of heavy metals has not been discussed in the previous papers. In this paper, we present and discuss the leaching behavior of heavy metals from aerobic and anaerobic landfill bioreactor test-cells as well as from a control cell.

## 2. Materials and methods

### 2.1. Test cell construction

Three test cells with geomembrane bottom liners were constructed in landfill A located in Kanto Region of Japan. Namely, aerobic cell with air injection and leachate recirculation, anaerobic cell with leachate recirculation only and control cell with no air or leachate injection [6]. In landfill A, wastes are landfilled in 3 m layers (2.5 m waste + 0.5 m cover soil), therefore the height of the cells were determined by the operational practices of the landfill. The shape of the cells were

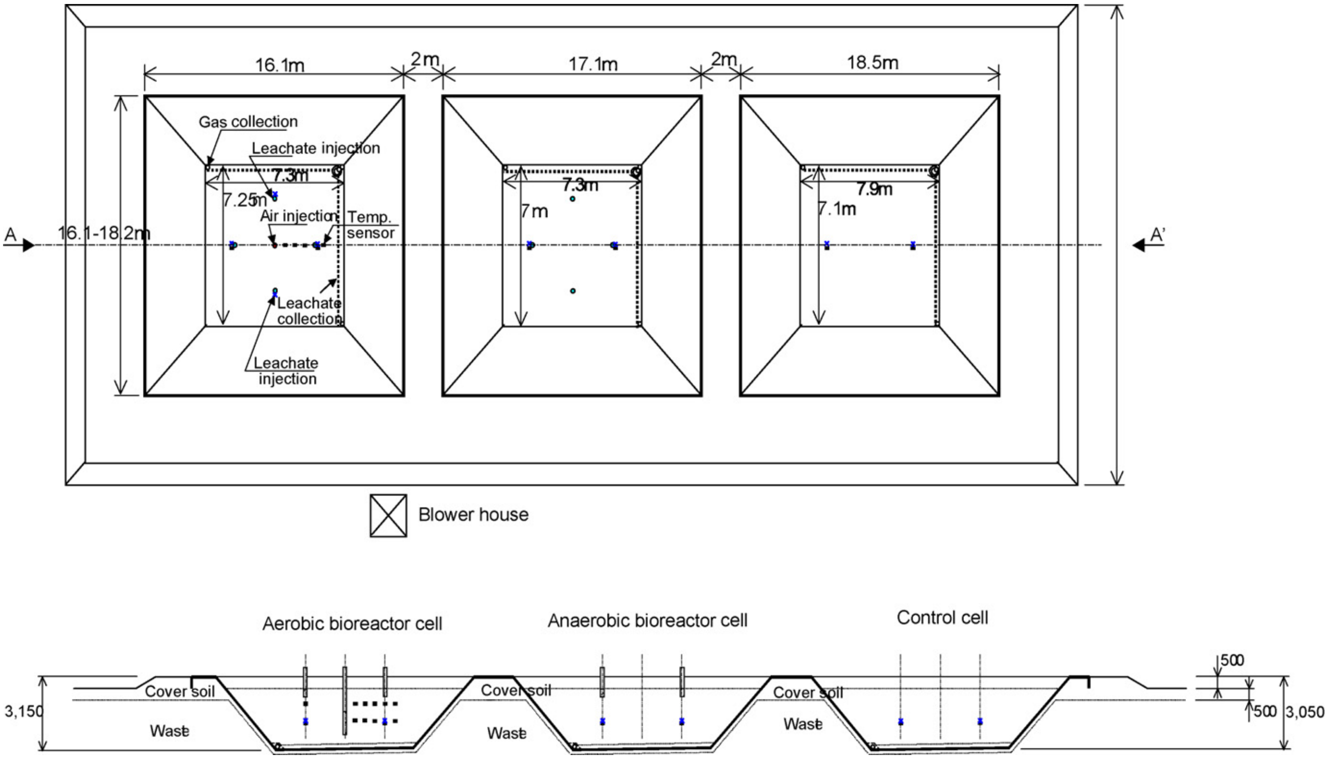

Fig. 1. Schematic layout of the bioreactor test cells.

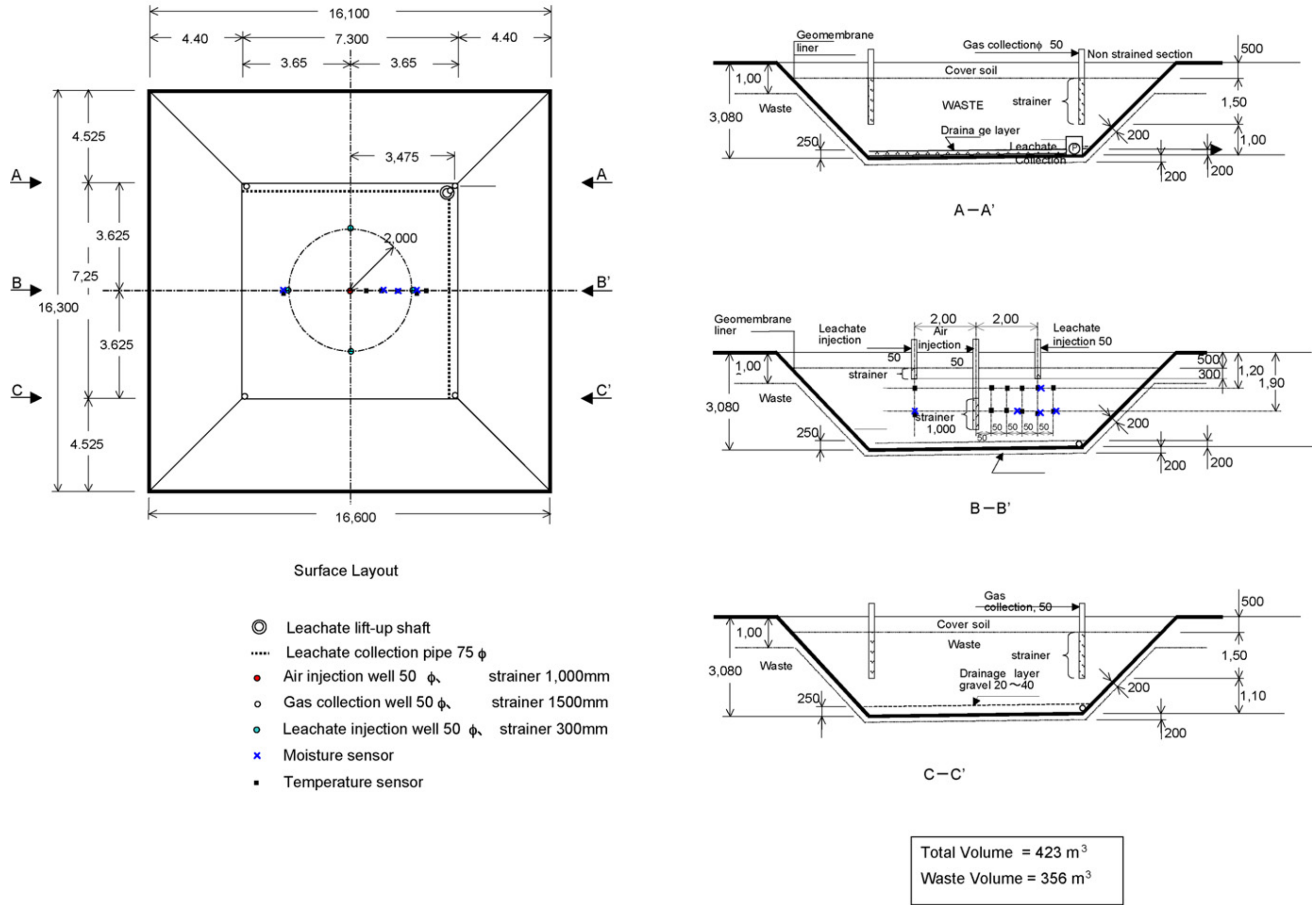

Fig. 2. Aerobic bioreactor test cell design and instrumentation layout.

like inverted truncated pyramids (bottom: 7.5 m × 7.5 m; top: 17 m × 17 m), although the exact size of each cell was slightly different due to limitations of construction equipment. Waste volumes in aerobic, anaerobic and control cell were 356.4, 378.8 and 426.8 $m^3$, respectively (Figs. 1 and 2).

### 2.2. Waste characteristics

Landfill A receives unweathered MSWI bottom ashes and shredded incombustible wastes (<10 cm) from varying sources such as MSW recycling centers and automobile recycling plants. Each cell has a capacity of almost the amount of the waste received in 1 day, and the cells were filled within consecutive 3 days. It was noticed that amount of ashes and shredded incombustibles differed in each cell. Since it was not possible to accumulate and homogenize the wastes before filling, this result was inevitable. Table 1 shows the ratios of ash and shredded residues in each cell.

### 2.3. Operation of the landfill bioreactor test cells

Air was injected into aerobic cell via one injection well (50 mm diameter). The length of the slotted part was 100 cm. The blower was operated between 08:30 and 16:30 (8 h) on the

Table 1
Characteristics of the wastes deposited in the test cells

| | Aerobic cell | | Anaerobic cell | | Control cell | |
|---|---|---|---|---|---|---|
| | Wet weight (kg) | % | Wet weight (kg) | % | Wet weight (kg) | % |
| MSWI bottom ash | 130200 | 43.9 | 185300 | 47.3 | 238800 | 55.1 |
| Industrial waste ash | 0 | 0.0 | 11800 | 3.0 | 20700 | 4.8 |
| Sludge incineration ash | 0 | 0.0 | 3600 | 0.9 | 0 | 0.0 |
| Incombustibles | 80000 | 26.9 | 120600 | 30.8 | 104200 | 24.0 |
| Glass–ceramics | 2200 | 0.7 | 2400 | 0.6 | 3700 | 0.9 |
| Melting slag | 8100 | 2.7 | 8200 | 2.1 | 25400 | 5.9 |
| Plastic rich waste | 76600 | 25.8 | 59800 | 15.3 | 40900 | 9.3 |
| Total | 297100 | | 391700 | | 433700 | |

Table 2
Heavy metal levels in the raw leachates of the test cells

| Heavy metal | Order of quantity (abundance) in the test cells | Observed concentrations in aerobic cell (mg/l) | Japanese state discharge limits[a] (mg/l) | Local discharge limits[b] (mg/l) |
|---|---|---|---|---|
| Zn | Aerobic > anaerobic > control | <1.0 | 5 | 0.5 |
| Mn | Aerobic > anaerobic > control | <2.0 exceptions: 16.7 on Day 50, 11.5 on Day 58 (before start of aeration) | 10 | 1 |
| Fe | Aerobic > anaerobic ≈ control | <1.6 | 10 | 1 |
| B | Aerobic > anaerobic ≈ control | <25 | 10[c] | |
| Si | Aerobic > anaerobic ≈ control | <18 | | |
| Cr | Aerobic > anaerobic > control | <0.03 as Cr | 2 | 0.2 |
| $Cr^{6+}$ | | | | 0.05 |
| Cu | Aerobic > anaerobic ≈ control | <0.45 | 3 | 0.3 |
| Pb | Aerobic > anaerobic ≈ control | >0.1 for the first 120 days (leachate is in anaerobic conditions), <0.1 for the rest of the experiment | 0.1 | |
| Ba | Anaerobic > aerobic > control | | | |
| Sr | Anaerobic > aerobic > control | | | |
| Al | Control > anaerobic > aerobic | | | |
| As | Control > anaerobic ≈ aerobic | <0.01 | 0.1 | |
| Mo | Control > anaerobic > aerobic | | | |
| V | Control > anaerobic > aerobic | | | |
| Hg | | | | 0.0005 |
| Ni | Control > aerobic > anaerobic | | | |

[a] Ministerial ordinance determining engineering standards to final disposal site for municipal solid wastes and final disposal site for industrial wastes (appended Table 1), issued on 14 March 1977 and revised on 28 April 1989.

[b] Stricter discharge limits agreed between the landfill owner/operator and the local residents.

[c] To be enforced after April 2005.

business days only. Airflow rate has been set as $2.0\,\mathrm{m}^3$/min ($120\,\mathrm{m}^3$/h, $960\,\mathrm{m}^3$/day). This corresponds to $2.7\,\mathrm{m}^3/\mathrm{m}^3$/day ($V = 354.6\,\mathrm{m}^3$).

In aerobic and anaerobic bioreactor cells, leachate lifted-up to collection tanks on the surface is recycled to the cells beneath the cover soil through four vertical leachate injection wells with 50 mm diameter. The leachate recycle rate was set as 15 l/min and 16 times 15 min operation of the pumps ($Q = 3600$ l/day), respectively. However, leachate recirculation was halted after Day 180 due to excessive infiltration and leachate accumulation in the cells caused by high precipitation.

### 2.4. Leachate sampling analysis

Leachate samples were brought to the laboratory within a few hours and filtered with 0.45 μm membrane filters and stored at +4 °C. All the heavy metal concentrations reported in this paper represents the dissolved metals (Table 2). Blank samples with deionized water were prepared in the same way to identify possible contaminations. Samples were acidified with nitric acid at a final concentration of 1%, and analyzed with ICP-OES (Vista Pro, Seiko Instruments Inc.). Following the ICP-OES analyses, samples were investigated by ICP-MS (Agilent 7500 Series) for low concentration metals such as cadmium, lead, arsenic and chromium, copper, manganese, nickel, selenium and vanadium. The dissolved organic carbon (DOC) was measured by combustion (NPOC) with a Shimadzu TOC-5000 instrument. pH and ORP values were measured on the site immediately after sampling.

## 3. Results and discussions

### 3.1. pH and ORP

In MSW landfills where organic rich municipal wastes are deposited aeration generally increases the pH of leachate reduced by accumulation of volatile fatty acids. However, the case is opposite for most of the landfills in Japan at which either

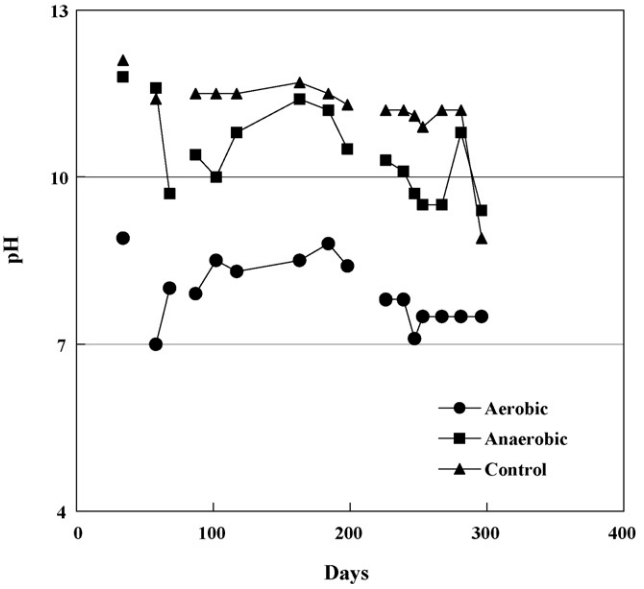

Fig. 3. Behavior of pH in the leachates of the test cells.

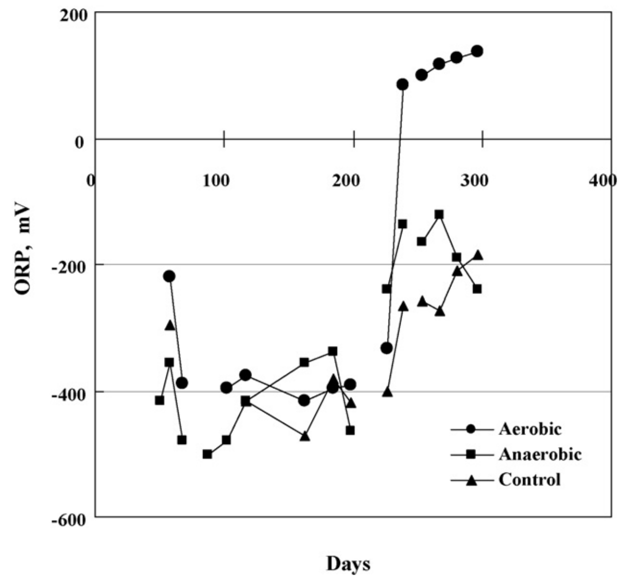

Fig. 4. Behavior of ORP in the leachates of the test cells.

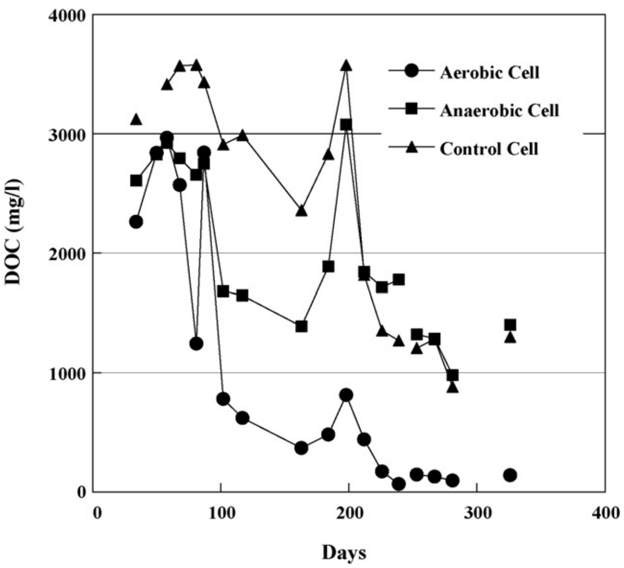

Fig. 5. Behavior of DOC in the leachates of the test cells.

incineration residues deposited alone (monofill) or together with shredded incombustible wastes. pH values in the test cells have differed significantly as a result of differences in ash to shredded incombustible waste ratios. Aerobic cell had the lowest amount of bottom ash (40%) yielding initial pH values below 9, which permitted the early initiation of microbial activity. In the control cell (ash: 65.7%) initial pH was around 12, and remained above 11 for most of the experimental period. In the anaerobic cell (ash: 53%) pH has decreased from 11.5 up to 9.5 with fluctuations (Fig. 3). ORP values in all the cells exhibited similar behavior ranging between −350 and −500 mV during first 200 days (Fig. 4). Low ORP values for aerobic cell leachate samples were caused by leachate accumulation in the cell. Since injected air can travel in the unsaturated upper zone only, leachate accumulated at the bottom remained anaerobic. It was also confirmed by black color and hydrogen sulfide smell of the samples. On Day 226, all the leachate was pumped out after sampling. As a result, ORP has jumped to +85 mV on the next sampling (Day 239). ORP values of anaerobic and control cells also make an increase from −400 mV level to −200 mV level as leachates in these cells were also discharged. New leachates produced as a result of surface infiltration had higher ORP.

### 3.2. DOC and IC

DOC (dissolved organic carbon) values in the cells were relatively high (up to 3500 mg/l), and rapid reduction in DOC and increasing dissolved IC (inorganic carbon) levels in aerobic bioreactor cell compared to other cells clearly showed the acceleration of bio-stabilization in this cell. In anaerobic and control cells, DOC decreased mainly due to dilution (Fig. 5). However, it is very clear that DOC reduction in aerobic cell is larger than that caused by dilution, since initial concentrations were very close in both anaerobic and aerobic bioreactor cells, and chloride concentrations indicated rather higher dilution in anaerobic and control cells (data not shown). BOD (biological oxygen demand for 5 days) values showed distinctly different pattern between the leachates of the three test cells. BOD values in leachate of aerobic cell has decreased faster than DOC and became negligible (<10 mg/l) after around Day 120 (data not shown). Significant increase in DOC in the leachates of all the cells around Day 200 was due to leachate discharge and arrival of new leachate from the unsaturated middle and upper parts of the cells.

### 3.3. Nitrate

The observations have indicated that nitrogen removal through nitrification and denitrification with aerobic landfill bioreactor operation is possible even under extremely high salt and heavy metal concentrations [6].

### 3.4. Leaching behavior of heavy metals form the test cells

Few studies are available in the literature reporting the leaching behavior of heavy metals from landfill bioreactors. Hantsch et al. [3] have reported no noticeable increase in heavy metal concentration in the leachates of aerobic lysimeters compared to anaerobic lysimeters which were filled with excavated waste from an old landfill. Ritkowski and Stegmann [4] have observed increases in Ni, Cu and Cd with no noticeable change in Cr leaching at ORP values between +200 and 250 mV created by aeration of lysimeters. However, concerning the leachability of total content, only 0.02–0.5% of the heavy metals were mobilized into leachate. Since, the leachability was correlated with ORP values, they recommended aeration rates low enough not to increase ORP values significantly. The only study on co-disposed MBP waste and MSWI residues by Cossu et al. [5] has investigated the impact of aerobic conditions in lab-scale lysimeters. Among the heavy metals monitored, Cr and Cu only have slightly increased with start of aeration after 52 days under anaerobic conditions.

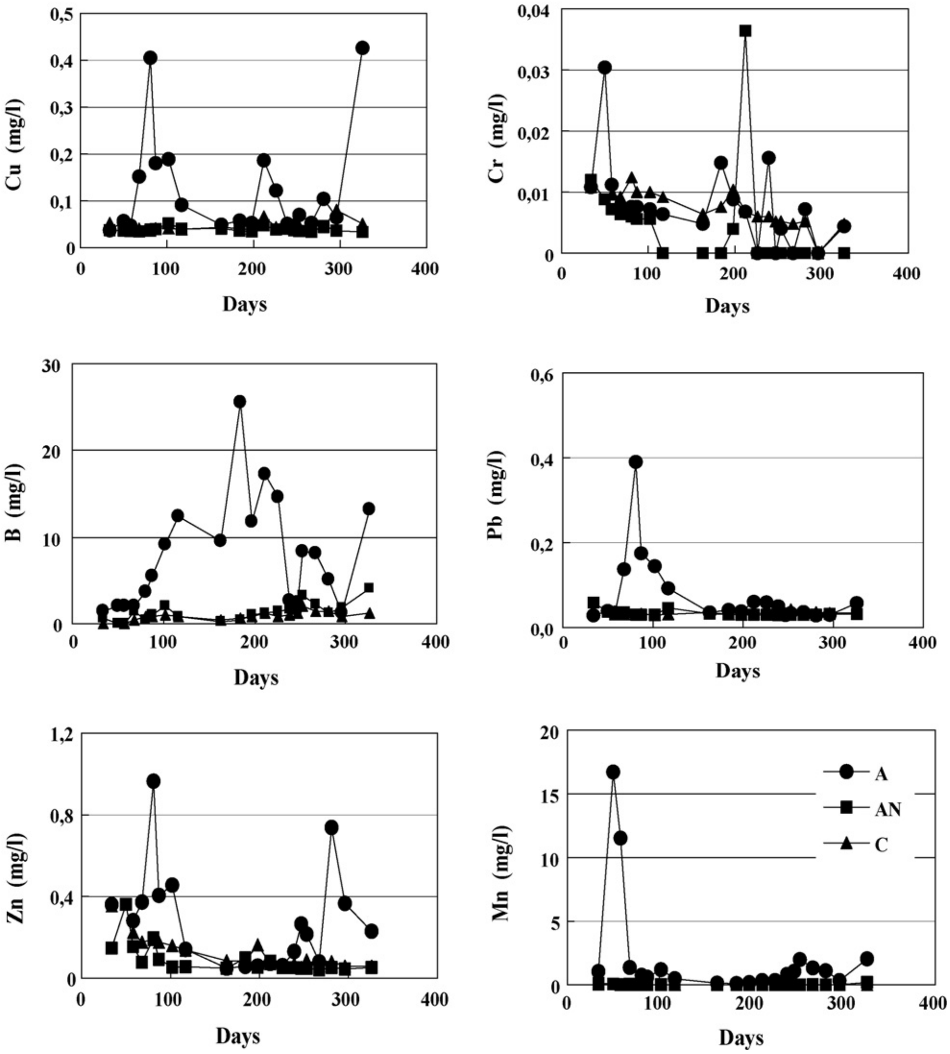

Fig. 6. Leaching behavior of Cu, Cr, B, Pb, Zn and Mn in the test cells (A, aerobic cell; AN, anaerobic cell; C, control cell).

Heavy metal concentrations observed in this study are given in Figs. 6 and 7. Heavy metals which were comparatively higher in leachate of aerobic cell were copper, lead, boron, zinc, manganese and iron, and those apparently lower were aluminum, arsenic, molybdenum, and vanadium. However, no excessive release of heavy metals under aerobic conditions was observed compared to anaerobic and control cells.

Copper had comparatively higher concentrations in the leachate of aerobic cell. This could be due to two factors: higher initial amounts of these metals in aerobic cell or enhanced leaching under aerobic conditions. Jung et al. [8] have shown that shredded bulky wastes have several times higher copper content than MSWI bottom ashes, mainly due to power cords and electronic circuit boards in these wastes. However, it should be noted that observed copper concentrations (below 0.5 mg/l) are far below the state discharge limit of 3.0 mg/l and even most of the time below the local limit of 0.3 mg/l. Zinc had somehow similar behavior, exceeding the local discharge limit of 0.5 mg/l two times only, but mostly less than this limit. State discharge limit for the zinc is 5.0 mg/l.

There could be several reasons for higher boron concentrations observed in aerobic cell leachate: pH, ORP, higher amount of boron rich waste, etc. MSWI bottom ashes have relatively low boron contents ranging between 38 and 510 mg/kg [9]. Waste glasses are suspected to be the most probable sources of boron. Possibility of their contribution to increased boron leaching under aerobic landfill bioreactor conditions is being investigated. Since the pH and ORP were the clearly different conditions, their possible roles are discussed here. It has been shown that the leachability of boron from APC residues are higher and independent from pH until pH 10, and decreases sharply at pH greater than 10 [9]. Lower pH would be the major reason for significantly higher boron concentration in the leachates of aerobic cell compared to those of anaerobic and control cells. There was no meaningful correlation between DOC and B concentrations. On the other hand, aerobic conditions seem to result in reduction of B leaching rather than an increase, since boron concentrations start to decrease while ORP increase. This can also be due to dilution. Higher B concentrations before Day 200 corresponds to lower ORP values, an indication of anaerobic

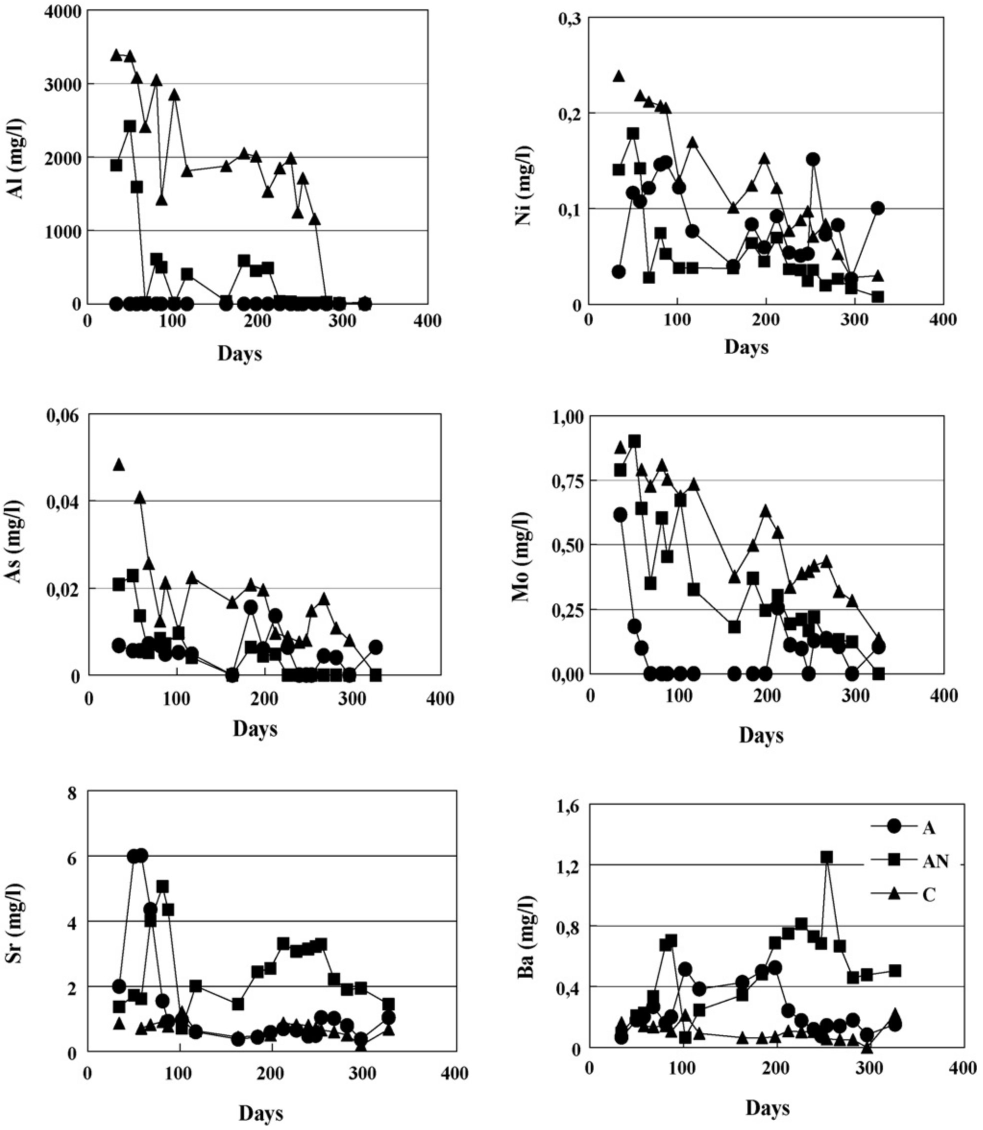

Fig. 7. Leaching behavior of Al, Ni, As, Mo, Sr and Ba, in the test cells (A, aerobic cell; AN, anaerobic cell; C, control cell).

condition of the leachate accumulated at the bottom of the aerobic cell. Increasing ORP values correspond to higher removal of the leachate from aerobic cell. Boron leaching data from the literature is not sufficient for comparing the results obtained in this study.

Lead concentrations were slightly higher in the aerobic cell and exceeded the state limit of 0.1 mg/l in the first half of the experimental period where ORP values of the leachate were around −400 mV. However, it decreased and never increased again even when the ORP has increased above +130 mV. Cadmium concentrations were even below the detection limits of ICP-MS (<0.004 mg/l), in all the cells.

Chromium concentrations were quite below the discharge limits and similar for all the cells. Polettini and Pomi [10] has observed higher Cr leaching from bottom ash samples treated with air, and commented that it would be due to higher solubility of oxidized chromium forms. Unfortunately, it is not possible to make a conclusion on our observations since we did not quantify the oxidized and reduced forms of chromium in this study.

The most significant different leaching pattern was observed for aluminum. Al concentrations in leachate samples of aerobic cell were below 1.2 mg/l throughout the experimental period, while it ranged between 500 and 3500 mg/l in anaerobic and control cells with the highest concentrations in the control cell. Al leaching from bottom ash is strongly pH-dependent and results in the characteristic V-shaped log-concentration/pH curve with minimum leaching around neutral pH values. Almost negligible Al leaching in aerobic cell and the highest leaching from control cell indicates strong pH dependence as can be seen from pH values in Fig. 3. On the other hand, calcite formation (carbonation) and neoformation of amorphous aluminosilicates might have also played a role at some extent through adsorption phenomenon [11].

Nickel concentrations have decreased with the time in all the cells. Nickel has increasing solubility with decreasing pH for

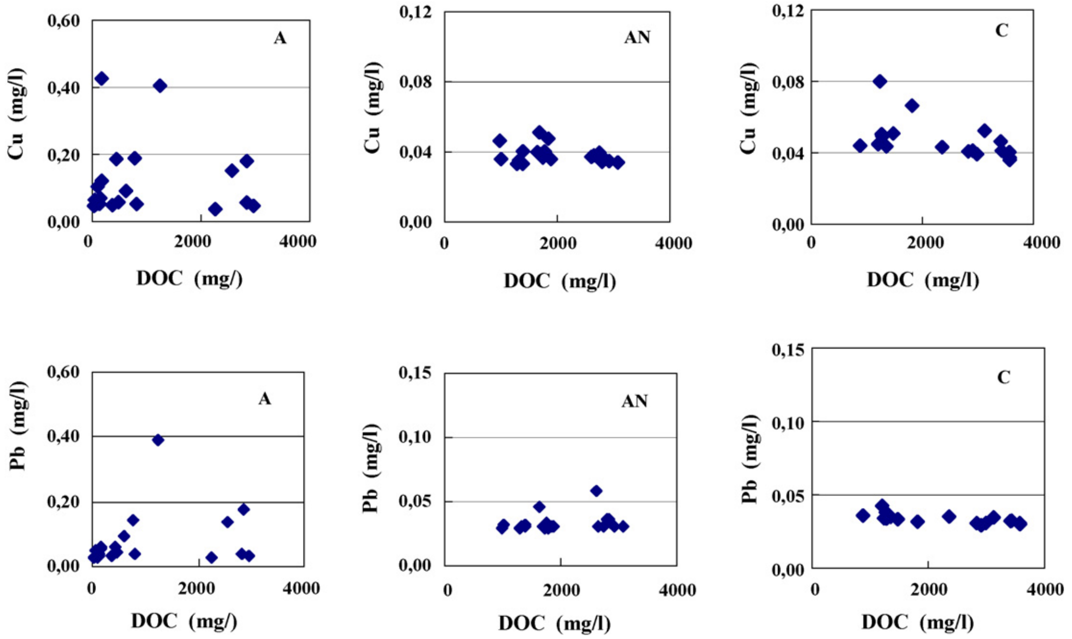

Fig. 8. Relationship between Cu and Pb leaching and DOC concentrations in the leachates of the test cells (A, aerobic cell; AN, anaerobic cell; C, control cell).

bottom ash [9]. However, we could not observe meaningful correlation between neither pH nor oxidation–reduction potential differing in each cell and Ni leaching behavior. It seems that DOC had more influence on Ni leaching as can be seen from Fig. 9.

In our study, no significant increase in heavy metals was observed as a response to increased ORP levels. One explanation would be higher easily leachable concentrations of heavy metals. That means effect of oxidized environment on leaching behavior of heavy metals and also probably for other organic pollutants can be observed after these easily leachable fraction has been washed out. Since the test cells were operated for only 1 year, the amount of water entering into the waste has played a stronger role than ORP changes. On the other hand, it is expected that huge amounts of $CO_2$ generated during degradation of organic matter has resulted in carbonation of the wastes, as can be understood from the substantial reduction in oxygen utilization and ANC (acid neutralization capacity) pattern with a strong pH resistance against acid addition for the excavated waste samples from aerobic cell. The same behavior could not be observed for the samples from other cells [6]. These observations have indicated that carbonation might have played an important role resulting in no increased leaching behavior of heavy metals in the aerobic cell. After operating aerobic lysimeters for more than 360 days, Ritkowski and Stegmann [2] has also observed a decreased metal leaching following a reduction in the leachate exchange rates (resulting in longer retention-contact times), and concluded that this could be due to stronger binding of metals to organic components, carbonates and hydroxides.

Heavy metal leaching, especially for Cu and Pb has been suggested to be strongly dominated by interaction with particulate matter and DOC [1]. However, no correlation between DOC levels and Cu and Pb leaching was observed in our study (Fig. 8). This indicates that leaching of these heavy metals was not being controlled dominantly by DOC in the test cells. Rather, it was controlled mainly by simple dissolution into water phase from the solid matrix at this early stage of landfilling. On the other hand, Ni, Zn, Mo and Cr leaching patterns were well correlated with DOC leaching with exception for Zn and Mo in aerobic cell (Fig. 9).

### *3.5. Further experiments on the solid waste samples to explain the observed leaching patterns*

Behavior and potential of heavy metal leaching from representative solid waste samples of the test cells are now being investigated. Due to difficulties in obtaining representative samples from large-scale test cells, a new approach was followed in this study for better characterization of the solid waste samples to quantify the development of stabilization. During filling the test cells, a homogeneous representative mixture of MSWI bottom ash and shredded incombustible solid wastes was prepared by mixing equal weights of each on wet weight basis. Then it was divided into subsamples and put into the bags which allow air and water to pass through but retain even the smallest fractions in the waste. These bags containing homogenous waste samples were named “Bio-probe Bags”. Several Bio-probe Bags containing 1–2 kg of identical waste samples were placed in the cells, mainly near the sensors and the corners at different depths during filling the cells. Aerobic cell has received 20 Bio-probe Bags and 10 Bio-probe Bags were placed in anaerobic and control cells, respectively. After 1-year operation, the test cell experiment was terminated and the cells were excavated for sampling.

The number of Bio-probe Bags that could be recovered from aerobic, anaerobic and control cells were 8, 8, and 4, respectively. Beside the Bio-probe Bags, more than 100 samples from the three cells were collected. Total content and detailed pH dependent leaching behavior of heavy metals are now being

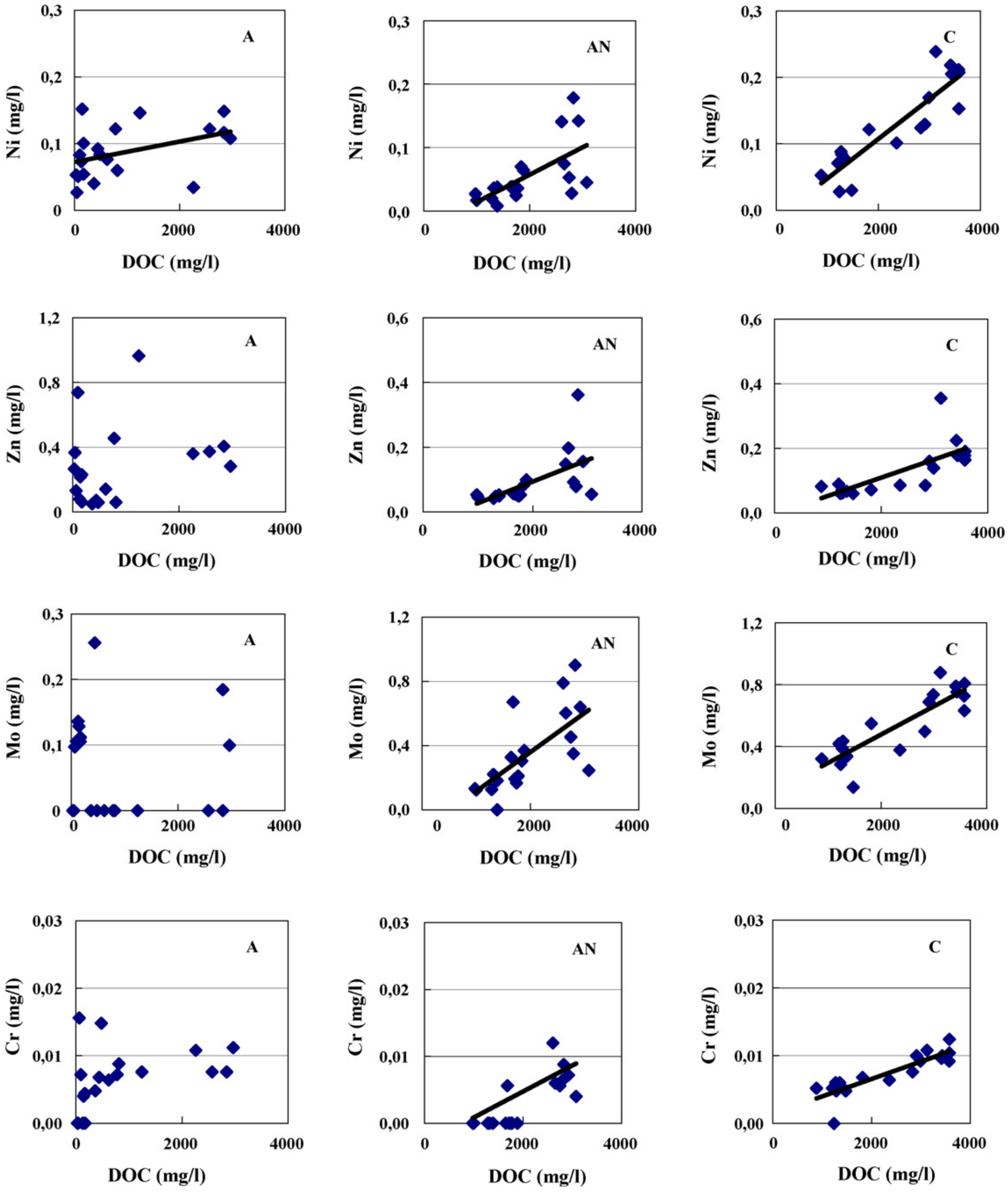

Fig. 9. Relationship between DOC and Ni, Zn, Mo and Cr leaching (A, aerobic cell; AN, anaerobic cell; C, control cell).

investigated with both Bio-probe Bag and other samples. Benefit of aerobic landfill bioreactor operation will be understood better after evaluating the solid waste samples.

## 4. Conclusions

Aerobic landfill bioreactor operation has proved to be beneficial for reducing the DOC leaching and nitrogen removal through nitrification for co-disposed MSWI bottom ash and low-organic residues. It was concluded that aerobization of landfills of heavy metal rich MSWI bottom ash and shredded residues is possible with no dramatic increase in heavy metals in the leachate.

There was no meaningful correlation between DOC and Cu and Pb for all the cells, and this was also case for Zn and Mo in aerobic cell. On the other hand, oxidation–reduction potential values ranging between −400 and +130 mV as a result of aeration in aerobic cell, have also did not indicate any correlation with heavy metal leaching. Almost negligible aluminum leaching from aerobic cell should be due to lower pH and carbonation, since Al has very low leaching at neutral pH values.

The only heavy metal that exceeded the Japanese state discharge limits (10 mg/l, to be enforced after April 2005) in the aerobic cell leachate samples was boron and there was no meaningful correlation between boron leaching and oxidation–reduction potential and DOC. Higher boron levels in aerobic cell should be due to comparatively lower pH values in this cell. However, it is anticipated that this slightly increased concentrations of boron (maximum 25 mg/l) will be beneficial for long-term stability of the landfill through faster washout of B. Leached B can be treated during bioreactor operation and until termination of post-closure care. Then the landfill should be much safer for long-term risk. On the other hand, B containing wastes such as glass cullet need to be diverted from the landfills for reducing the leaching potential of this metal.

Further leaching tests and total content measurements on solid samples excavated from the test cells will provide further information on the effect of aerobic landfill bioreactor operation for leaching behavior of heavy metals.

## Acknowledgements

The authors gratefully acknowledge the contributions by staff at Yorii Environmental Management Center (Saitama, Japan) and Mr. Toshiro Oshikata and Mr. Masaaki Ebihara of Taisei Corporation (Tokyo, Japan) for the construction of the test cells. This work was financially supported by Ministry of Environment, Japan (Project no. K1535). The authors also thank to Dr. Bulent Mertoglu of Marmara University, Turkey for his help during editing the manuscript.

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