Contribution of Each Combustible Waste to the Element Content of MSW Incineration Residue

Hirofumi Sakanakura\textsuperscript{1,}*, SeungKi Back\textsuperscript{1} and Tomohiro Naruoka\textsuperscript{2}

\textsuperscript{1} National Institute for Environmental Studies, 16-2, Onogawa, Tsukuba, Ibaraki, 305-8506 Japan
\textsuperscript{2} Tottori Prefectural Research Institute of Public Health and Environmental Sciences, 526-1, Minamidani, Yurihama, Tohaku-gun, Tottori, 682-0704, Japan

* corresponding author: sakanakura@nies.go.jp

Abstract. To investigate the origin of toxic/beneficial metals in incineration residues, we tried to estimate the contribution of each combustible waste to various elements in incineration residue. Municipal solid waste (MSW) sample to be incinerated was collected from a waste pit of incineration plant A by a crane and compressed according to the quarter method. After excluding metals and incombustibles, the MSW sample was divided along with the material such as kitchen waste, vinyl and plastic, wood, rubber, and so on. Each sample was dried at 105°C for 2 hours to obtain the water content, and cut into small piece. Each sample was placed in a magnetic crucible and closed with a lid, then, heated at 650°C in a muffle furnace to obtain constant weight after ignition loss. After the heat-up and cool down, two-stage extraction consisting of aqua regia decomposition and alkali fusion was carried out on each ash. Each extract was subject to ICP-OES and ICP-MS to determine the element content in each combustible. Element composition of the ash was well correlated with the element composition of actual incineration residues collected at the same incineration plant on the same day of sampling. As a source of toxic metals, 68% of Pb was proved to originate from vinyl and plastic. As for Cd, woods accounted for 38% and rubber and leather accounted for 29%, respectively.

1. Introduction

In Japan, total discharge amounts of municipal solid waste (MSW) decreased by around 20% in FY 2015 compared to the peak amount in FY 2000. The incineration ratio in total discharge is still close to 80%. Besides, the proportion of incineration residues in the total disposal amount to landfill has risen from 54% in FY 2000 to about 76% in FY 2015 [1]. Based on this situation, the importance of incineration residues in MSW management is increasing even from a nationwide perspective of Japan. Furthermore, this issue becomes more urgent for local governments where the prospect of final disposal is severe. Recycling of incineration residues, mainly by melting of ash and by feeding as cement raw materials, is progressing little by little along with the decrease of final disposal amount (Figure 1) [1]. In recent years, further attempts have been made to recycle of incineration residues as construction materials by mixing with cement and solidifying it. As one of such efforts, the authors have recommended to classify the target residue for recycling according to its characteristic. A main bottom ash has relatively low for heavy metal content. On the other hands, heavy metal content in a
grate shifting ash and boiler ash is relatively high. In addition, aging and weathering treatment is recommended to reduce heavy metal to be leached [2].

However, for all type of recycling, it is very important to reduce the total input of such harmful elements, because harmful heavy metals are concentrated in the incineration residue. Reducing the amount of harmful elements entering MSW incinerator could lead to increasing the possibility of recycling of residue.

Two types of methods have been applied to estimate the chemical properties of input MSW: the estimation using mass balance from analysis of residue [3–6] and the direct analysis of input MSW [7–10]. The mass balance method could present representative total amount of elements which put to MSW incinerator, however information on detailed MSW is not available. Direct analysis of the input MSW method could present supplementary information for identification of origin wastes containing beneficial/toxic element which behave in MSW incinerator. Law and Gordon (1979) reported the 20 elements contents in combustible fraction of MSW [7]. Olajire and Aodele (1998) presented the information of the 17 elements in MSW from 12 cities in Nigeria [8]. Zhang et al. (2008) reported the 6 elements contents of MSW in China [9]. These researches provided information on the entire of MSW without categorization. After that, Long et al. (2011) presented information of Cu and Zn contents from MSW categorized with 15 components (kitchen waste, plastic paper, and so on) [10]. However, information on the two target elements (Cu and Zn) has limitations to identifying the origin waste for elements contained in incinerator residues.

In this study, we estimated the contribution of each combustible waste category to the 52 elements in MSW incineration residue. The collected combustible waste was separated to 6 categories and each category item was ignited and grinded for Wavelength Dispersive X-ray fluorescence (WDXRF) analysis. In addition, chemical properties were analyzed by ICP-MS/OES analysis after acid digestion and alkali fusion steps. This result could be utilized to reduce harmful elements for the recycling of MSW incinerator residues.

![Figure 1](image.png)

**Figure 1.** Landfilling and recycling amounts and recycling ratio of incineration residues in Japan. Categories “Melting” and “Raw Material for Cement” were added in FY 2005 and FY 2007, respectively, in the statistic options. [1]

2. Experimental

2.1. MSW Sample collection and preparation

In an MSW incineration facility A in Japan, a grasp of MSW to be incinerated was collected from a waste pit by a crane and divided according to quarter method. Metals and bigger incombustibles such as glass and ceramics more than 5 mm were removed before analysis.
2.2. MSW composition, water content and Loss on Ignition (LOI)

The MSW sample was categorized as 6 groups; “Kitchen waste”, “Paper and Cloth”, “Wood, Bamboo and Straw”, “Vinyl and Plastic”, “Rubber and Leather”, and “Small particles”. The weight of each sample was measured to derive the composition of MSW. Each sample was dried at 105°C for 2 hours to obtain the water content, and cut into small piece less than 10 mm. Each sample was placed in a magnetic crucible and closed with a lid, then, heated at 650°C in a muffle furnace to obtain constant weight after ignition loss.

2.3. Element content analysis

After ignition, the samples were milled to pass the sieve opening of 125 µm for the two-step element analysis as follows. One gram of the milled sample in triplicates for each sample was digested by adding aqua regia and heating up to 120°C. After cooling down, filtered liquid was diluted and analyzed by ICP-OES (Agilent 720) and ICP-MS (Agilent 7500CX). The solid residue after filtration was again digested by the alkali fusion method using lithium metaborate in platinum dish. Then, the filtered liquid was analyzed in the same way of aqua regia solution. Total elemental content was calculated by the sum of both extraction amounts of aqua regia and alkaline fusion solutions. Triplicates were analyzed for each sample. As for Li, B and Pt, results from alkali fusion is excluded. In addition, WDXRF (Rigaku Primus II) was adopted on the milled and compacted sample for determination of Si, P, S and Cl.

2.4. Incineration residue sample collection and handling

Beside combustibles, all types of incineration residue such as bottom ash, grate shift ash, gas cooler ash and air pollution control (APC) residue were collected and analyzed in the same way dictated above, and generation amount of each ash was counted. The element partitioning during the incineration process was discussed, and the average element content of all incineration residues was obtained [11]. In this paper, the elemental balance between MSW to be incinerated and the residues after the incineration process was compared.

3. Results and discussion

3.1. MSW composition, water content and Loss on Ignition (LOI)

Table 1 shows the result of the composition of combustibles and small particles. The “Paper and Cloth” (39.3%) and “Vinyl and Plastic” (35.9%) were accounted for 75.2% of the total weight of combustibles and small particles. Various kinds of paper and pieces of clothes were included in “Paper and Cloth”. And, packaging film, plastic bag and plastic container were included in “Vinyl and Plastic”. The relatively small amount of “Wood, Bamboo and Straw” (11.2%), “Kitchen waste” (8.4%), “Small particle” (3.9%), and “Rubber and Leather” (1.3%) were included in collecting MSW. Table 2 shows water content, LOI, and ash content of each waste category. However, as the sample was already almost dry condition when it arrived in the laboratory, water content is not a fresh value. For the ash content, the value of “Small Particles” (49.9%) was the highest, followed by “Rubber and Leather” (21.3%) and “Vinyl and Plastic” (11.6%). The ratio of total ash from the total waste was calculated 10.3%. By multiplying each ash content with the dry weight ratio of each category, proportion of the ash originated from each category was calculated. “Vinyl and Resin” has the highest value by nearly 40%, then it was in the order of “Paper and Cloth” (30.6%) and “Small Particle (18.2%).
Table 1. Composition of combustibles and small particles.

| Category                  | Weight ratio (%-wet) |
|---------------------------|----------------------|
| Kitchen waste             | 8.4                  |
| Paper and Cloth           | 31.9 39.3            |
| Wood, Bamboo and Straw    | 11.2                 |
| Vinyl and Plastic         | 22.9 35.9            |
| Rubber and Leather        | 1.3                  |
| Small particle (<5 mm)    | 3.9                  |
| Total                     | 100                  |

Table 2. Content of water, combustible and ash fraction of each waste category.

|            | Water %-wet | LOI %-dry | Ash %-dry | Proportion in total ash, % |
|------------|-------------|-----------|-----------|---------------------------|
| Kitchen waste | 7.5 | 91.6 8.4 | 6.4       |
| Paper and Cloth | 6.2 | 91.5 8.5 | 30.6      |
| Wood, Bamboo and Straw | 10.2 | 97.3 2.7 | 2.6       |
| Vinyl and Plastic | 2.2 | 88.4 11.6 | 39.5      |
| Rubber and Leather | 0.69 | 78.7 21.3 | 2.7       |
| Small particle (<5 mm) | 4.1 | 50.1 49.9 | 18.2      |
| Total         | 100.0       |           |           |                           |

3.2. Element Content

Table 3 shows the element content analysis results of each waste category. In this analysis, although the crucible was covered with a lid during ignition, its hermeticity was not perfect. Therefore, there was a possibility that volatile elements such as chlorine and sulfur are partly lost during heating at 650°C. Besides, as we consider a fraction highly likely shift to bottom ash, the results can be regarded as effective to some extent.

“Kitchen waste” had the highest Cl content among the 6 categories, and the second highest Na, P, and K content after “Small Particle”. As for “Paper and Cloth”, Cu content was the second highest after “Small Particle”, “Wood, Bamboo and Straw” has a relatively high content of Cd and Sn.

In “Vinyl and Plastic”, Ti, Nb, and Sb were the highest among the categories, and Cr, Ni, Sr, and Ba were the second highest after “Small Particle”. “Rubber and Leather” had the highest content of Mg, S, Zn, and Cd. “Small particles” had highest content in many elements. LOI of “Small particle” was 50% and other categories were in the range of 78 to 97% (Table 3). This means large amounts of element are contained in incombustible small particles.

Table 3. Element content of each waste category.

| Atomic Number | Element | Kitchen waste | Paper and Cloth | Wood, Bamboo and Straw | Vinyl and Plastic | Rubber and Leather | Small particle (<5mm) |
|---------------|---------|---------------|-----------------|------------------------|------------------|-------------------|-----------------------|
| 3             | Li      | 7.7           | 4.5             | 4.1                    | 9.7              | 7.3               | 18.7                  |
| 4             | Be      | <0.1          | <0.1            | <0.1                   | <0.1             | <0.1              | 0.3                   |
| 5             | B       | 144           | 49              | 29                     | 288              | 12.5              | 1160                  |
|   |   |   |   |   |   |
|---|---|---|---|---|---|
|   | Na | 12100 | 2200 | 1090 | 4320 |
|   | Mg | 991  | 2980 | 386  | 3380 |
|   | Al | 786  | 4620 | 683  | 1990 |
|14 | Si*| 1000 | 1660 | 661  | 1210 |
|15 | P* | 3050 | 352  | 228  | 254  |
|16 | S* | 639  | 473  | 315  | 2700 |
|17 | Cl*| 14900| 1470 | 200  | 7830 |
|   | K  | 6600 | 1400 | 1080 | 977  |
|20 | Ca | 9300 | 20700| 2530 | 32800|
|21 | Sc | 0.1  | 0.4  | 0.1  | 0.3  |
|22 | Ti | 56.0 | 144  | 128  | 3740 |
|23 | V  | 0.8  | 2.3  | 1.0  | 1.5  |
|24 | Cr | 6.0  | 5.0  | 16.5 | 21.4 |
|25 | Mn | 172  | 157  | 784  | 165  |
|26 | Fe | 800  | 816  | 2120 | 1310 |
|27 | Co | 1.0  | 1.4  | 2.2  | 2.5  |
|28 | Ni | 3.8  | 4.4  | 22.3 | 29.6 |
|29 | Cu | 33.5 | 550  | 95.8 | 23.4 |
|30 | Zn | 195  | 147  | 573  | 662  |
|31 | Ga | 0.2  | 1.2  | 0.2  | 0.5  |
|33 | As | 0.8  | 0.6  | 1.0  | 1.1  |
|34 | Se | 0.2  | <0.1 | <0.1 | 0.1  |
|37 | Rb | 7.5  | 2.3  | 2.3  | 1.7  |
|38 | Sr | 16.7 | 21.0 | 22.4 | 61.0 |
|39 | Y  | 3.9  | 5.0  | 14.9 | 6.0  |
|40 | Zr | 0.7  | 1.1  | 1.3  | 4.9  |
|41 | Nb | 0.1  | <0.1 | 0.1  | 4.1  |
|42 | Mo | 1.9  | 0.3  | 0.8  | 1.1  |
|46 | Pd | <0.1 | <0.1 | <0.1 | <0.1 |
|47 | Ag | 0.1  | 0.1  | 0.3  | 0.2  |
|48 | Cd | 0.4  | 0.4  | 4.4  | 0.3  |
|49 | In | <0.1 | <0.1 | <0.1 | <0.1 |
|50 | Sn | 1.5  | 2.3  | 11.6 | 4.1  |
|51 | Sb | 1.0  | 0.6  | 2.4  | 12.8 |
|55 | Cs | 0.1  | 0.1  | 0.1  | 0.1  |
|56 | Ba | 30.8 | 18.6 | 43.2 | 57.9 |
|57 | La | 1.8  | 3.5  | 6.1  | 3.1  |
|58 | Ce | 1.6  | 3.8  | 3.6  | 2.5  |
|60 | Nd | 0.3  | 1.1  | 0.8  | 0.6  |
|63 | Eu | 0.4  | 0.4  | 1.3  | 0.5  |
|66 | Dy | <0.1 | 0.1  | 0.1  | 0.1  |
|72 | Hf | <0.1 | <0.1 | <0.1 | <0.1 |
|73 | Ta | <0.1 | <0.1 | <0.1 | <0.1 |
|74 | W  | 0.2  | 0.3  | 0.2  | 0.7  |
|78 | Pt | <0.1 | <0.1 | <0.1 | <0.1 |
|79 | Au | <0.1 | <0.1 | <0.1 | <0.1 |
|81 | Tl | <0.1 | <0.1 | <0.1 | <0.1 |
|82 | Pb | 35.1 | 17.6 | 99.5 | 231  |
|83 | Bi | 0.1  | 0.1  | 0.1  | 0.1  |

Note: “<0.1” represents element content was less than 1 mg/kg. “*” represents that the elements are determined by WDXRF, and the rests are determined by two step digestion and ICP-MS/OES.
3.3. Comparison with Incineration Residues

Figure 2 shows the balance of ash composition calculating from both side of an incinerator, i.e., input of waste to the incinerator (this study) and output from the incinerator [11]. Both axes are shown in the total ash content in mg/kg in log scale, and each point represents each element. Input amount to the incinerator was calculated from the proportion of waste shown in Table 1 and element content shown in Table 3. Output amount of the incinerator was calculated from weight ratio and the element content of all incineration residues sampled from the same incinerator on the same day of the combustible samples [11]. In the figure 2, five straight lines are shown referring to the 0.1, 0.3, 1 (input = output), 3 and 10 times difference, respectively. The differences between the both results were within the range of 0.1 to 10 times, mostly 0.3 to 3 times, which shows that a good correlation was obtained through these analyses.

![Figure 2: Relations between element content calculated from all combustible waste categories and small particles (this paper) and combined analytical results of incineration residues sampled on the same day from facility A. (Source: Sakanakura, 2016.)](image)

3.4. Contribution of Each Combustible to Each Element

Figure 3 shows the calculation results of the contribution ratio of elements contained in each combustible waste to incineration residues. In elements where the environmental standards of soil are set in Japan (B, Cr, Pb, Cd, As, Se among measured elements. Cr is in “Cr(VI)” in the standard), the contribution ratio of “Vinyl and Plastic” was the highest in B, Cr, Pb, especially Pb accounts for 68%. Among the source of Cd content, “Wood, Bamboo and Straw” (38%) and “Rubber and Leather” (30%) accounted for the majority. For the source of As content, “Small Particles” contributed 44% of total inflow, followed by “Vinyl and Plastic” in 27%. There was no category whose contribution ratio exceeded 30% for Se. For valuable metals, 74% of Cu originated from “Paper and Cloth”. Zn of 43% and 20% was derived from “Vinyl and Plastic” and “Rubber and Leather”, respectively. The largest source of precious metals such as Pd, Ag, Pt, and Au was “Vinyl and Plastic” among the categories. However, we should note that these results are based on combustibles and small particles only, and incombustible fractions are excluded. It is expected that, for example, not only “Paper and Cloth” but some metals (incombustibles) will contribute origin of Cu.
When focusing on each classification, the highest contribution of “Kitchen waste” was P (37%), followed by K, Cl, Na, Se, Rh, Mo, each contributing 17% or more. “Paper and Cloths” contributed 74% of Cu followed by Ga (54%) and Al (53%), and also contributed Mg, Nd, Sc, Be and Ce in 39 to 42%. For “Wood, Bambo and Straw”, Cd (38%), Mn and Sn (around 30%), and Au, Eu and Y (22 to 24%) are higher contribution ratio than the weight composition value of this category (11.2%). The contribution ratio of many elements in “Vinyl and Plastic” exceeded the weight composition value of this category (36%). Which means the contribution of many elements was high compared to the weight ratio of input waste. Nb, Ti, Ta and Sb accounted for 80 to 97%, Pb, Zr, Hf, B, Cr, Ni, Sr account for 50% or more, and Pt, Ca, Li, Ti, Mo, Mg etc. contributed more than 36%. For “Rubber and Leather”, Zn (20%), Cd (30%), and Pt (17%) are remarkably high with respect to the weight composition value of 1.3% of this category. For “Small Particles” most elements accounted for more than 3.9% by weight composition, with W being 82%, Si being 46% and As being 44%.

**Figure 3.** Contribution ratio of each waste category to elements in incineration residues.
From these results, it is possible to lead various recommendations to reduce harmful elements and recovering valuable metals, in and from the incineration residues respectively. For example, to reduce the content of Pb, more attention to “Vinyl and Plastic” should be payed. Elaborate analyses should be promoted to advice policy to avoid the use of lead in specific vinyl and plastics.

4. Conclusion
In this paper, the origin of toxic/beneficial metals in incineration residues were estimated by igniting and digesting the original MSW samples separated into waste categories such as kitchen waste, paper and cloth, wood, vinyl and plastics, rubber and leather and so on. As a conclusion, source of Pb proved to be vinyl and plastic accounting for 68% of total Pb amount in combustible MSW. As for Cd, woods accounted for 38% and rubber and leather accounted for 29%, respectively. These information will help reduce the total content of toxic element in incineration residues by excluding such type of waste from MSW or by reducing elemental content of such toxic elements in the waste to be incinerated.

Acknowledgements
This study was funded in part by the Environment Research and Technology Development Fund (3K-14007) and (3-1804) of the Ministry of the Environment, Japan.

References
[1] Ministry of the Environment Japan, Survey results of actual condition of municipal solid waste treatment, 2016. http://www.env.go.jp/recycle/waste_tech/ippan/stats.html (accessed May 15, 2018).
[2] H. Sakanakura, H. Kuramochi, K. Sato, T. Fujikawa, S. Mizutani, H. Mongi, T. Naruoka, Construction of a total scheme for recycling of municipal solid waste incineration residues focused on useful and harmful metal behaviour (in Japanese), 2016. http://www.erca.go.jp/suishinhi/seika/pdf/seika_1_h29/3K143007.pdf.
[3] T. Astrup, C. Riber, A.J. Pedersen, Incinerator performance: effects of changes in waste input and furnace operation on air emissions and residues, Waste Manag. Res. 29 (2011) S57–S68.
[4] H. Belevi, H. Moench, Factors Determining the Element Behavior in Municipal Solid Waste Incinerators. 1. Field Studies, Environ. Sci. Technol. 34 (2000) 2501–2506.
[5] R. Bunge, Recovery of metals from waste incinerator bottom ash, Hochschule Für Tech. Rapperswil. Rapperswil, 2016.
[6] L.S. Morf, R. Gloor, O. Haug, M. Haupt, S. Skutan, F. Di Lorenzo, D. Böni, Precious metals and rare earth elements in municipal solid waste – Sources and fate in a Swiss incineration plant, Waste Manag. 33 (2013) 634–644.
[7] S.L. Law, G.E. Gordon, Sources of metals in municipal incinerator emissions, Environ. Sci. Technol. 13 (1979) 432–438.
[8] A.A. Olajire, E.T. Ayodele, Heavy Metals Analysis of Solid Municipal Wastes in the Western Part of Nigeria, Water. Air. Soil Pollut. 103 (1998) 219–228.
[9] H. Zhang, P.J. He, L.M. Shao, Flow Analysis of Heavy Metals in MSW Incinerators for Investigating Contamination of Hazardous Components, Environ. Sci. Technol. 42 (2008) 6211–6217.
[10] Y.Y. Long, D.S. Shen, H.T. Wang, W.J. Lu, Y. Zhao, Heavy metal source analysis in municipal solid waste (MSW): Case study on Cu and Zn, J. Hazard. Mater. 186 (2011) 1082–1087.
[11] H. Sakanakura, K. Yui, H. Kuramochi, T. Naruoka, H. Mongi, Concentration and distribution of 56 elements in residues from stoker-type municipal solid waste incineration, in: 9th International Conference on Combustion, Incineration/Pyrolysis, Emission and Climate Change (9th i-CIPEC), Kyoto, Japan, 2016.