Behavior of Radioactive Cesium in Municipal Solid Waste of Thermal Treatment Plants after Fukushima Nuclear Power Plant Accident – 16626

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ABSTRACT
Municipal solid waste (MSW) contaminated by radioactive cesium (r-Cs) has been treated by various thermal treatment facilities in Japan. However, the behavior of r-Cs in their plants is scarcely known. Therefore, we investigated 4 types of the thermal treatment plants to understand the r-Cs behavior during different thermal treatments of the MSW. The distribution of r-Cs between residue discharged from bottom of furnace (bottom ash, incombustibles or slag) and fly ash was affected by furnace type. The leaching rate of r-Cs from the fly ash and the bottom residues were measured. The leaching rate from the former was high and ranged from 40 to 100%. In contrast, the most of r-Cs in the latter did not dissolve into water. Furthermore, we evaluated whether baghouse (an air-pollution control equipment) successfully removed r-Cs compound in flue gas. R-Cs was efficiently removed regardless of thermal treatment type.

INTRODUCTION
A huge amount of radioactive substances was released due to the Fukushima Daiichi Nuclear Power Plant accident on March 11, 2011. The environment in eastern part of Japan was widely contaminated by the fallout of r-Cs, resulting in generation of radioactively contaminated MSW. In Japan, a densely populated country, waste thermal treatments such as incineration and ash-melting process have played a significant role in MSW treatment, because they can reduce mass and volume of the waste by 10% to 20%. After the accident, it was found r-Cs in MSW was highly concentrated into ash from the incinerator. Safety handling of the ash requires understanding the behavior of r-Cs in the MSW thermal treatment plants. Therefore, we investigated the behavior of r-Cs in various types of the plants.

METHOD
In this study, actual thermal treatment plants dealing with contaminated MSW were investigated. The specification for each facility is summarized in TABLE I. Stoker type incinerator discharges bottom ash and fly ash as incineration ash, whereas the others such as fluidized bed type incinerator and melting furnace generate fly ash only. The former and latter discharge incombustibles and slag from individual furnace bottom, respectively. Therefore thermal treatment solid residues can be divided to two resides; 1) residue from bottom of furnace such as bottom ash, incombustibles and slag, 2) fly ash collected by baghouse. Although there are various kinds of radioactive nuclei, we focused on only r-Cs in this research because the amount of r-Cs emission was so much larger. Concentration of r-Cs was expressed as the sum of Cs-134 and Cs-137 activities. We measured the r-Cs concentration of the solid residues as well as flue gas. Using mass balance based on actual plant operation data, the r-Cs

concentration data were converted into the r-Cs distribution among the solid residues and flue gas. In addition, leachability of r-Cs from the solid residues was investigated as follows; first, an amount of the sample was added to pure water in a plastic bottle, where the weight ratio of water to the sample was 10. Second, the solution was stirred or shaken for 6 hours, and then solution was separated by filtration. The leaching rate was determined by measuring the r-Cs concentration of the filtrate.

TABLE I. Overview of each thermal treatment plant.

<table>
<tr><th>Facility</th><th>NO.of date</th><th>Furnace type</th><th>Bottom residue</th><th>Temp. (°C)</th></tr>
<tr><td>A</td><td>1</td><td rowspan="2">Stoker type incinerator</td><td rowspan="2">Bottom ash</td><td rowspan="4">over 800</td></tr>
<tr><td>B</td><td>2</td></tr>
<tr><td>C</td><td>2</td><td rowspan="2">Fluidized bed type incinerator</td><td rowspan="2">Incombustibles</td></tr>
<tr><td>D</td><td>10</td></tr>
<tr><td>E</td><td>2</td><td>Fluidized bed gasification system with ash-melting furnace</td><td>Slag, metal Incombustibles</td><td>1300-1400</td></tr>
<tr><td>F</td><td>2</td><td>Shaft-type melting furnace</td><td>Slag, metal</td><td>1300-1650</td></tr>
</table>

RESULT

Distribution of r-Cs between fly ash and bottom residue is shown in Figure 1. 60 to 99% of R-Cs in MSW moved to fly ash in all type of plants. In the case of the stoker type incinerators, the distribution of r-Cs to bottom ash was relatively high compared with that to bottom residues in the other types. In terms of the r-Cs concentration ratio of fly ash to bottom residue, the ratio for melting furnaces (facility E and F) exceeded 20, while that for all the incinerators (facility A to D) up to 10. There is no significant difference between the two types of incinerators. These results indicate as follows; 1) Stoker type incinerator tends to leave r-Cs in bottom ash to some extent (30 to 40%). 2) In the melting furnaces most of the r-Cs is distributed to fly ash because they basically operate at a much higher temperature than the incinerators. Especially shaft-type melting furnace which sometimes operate over 1600 °C.

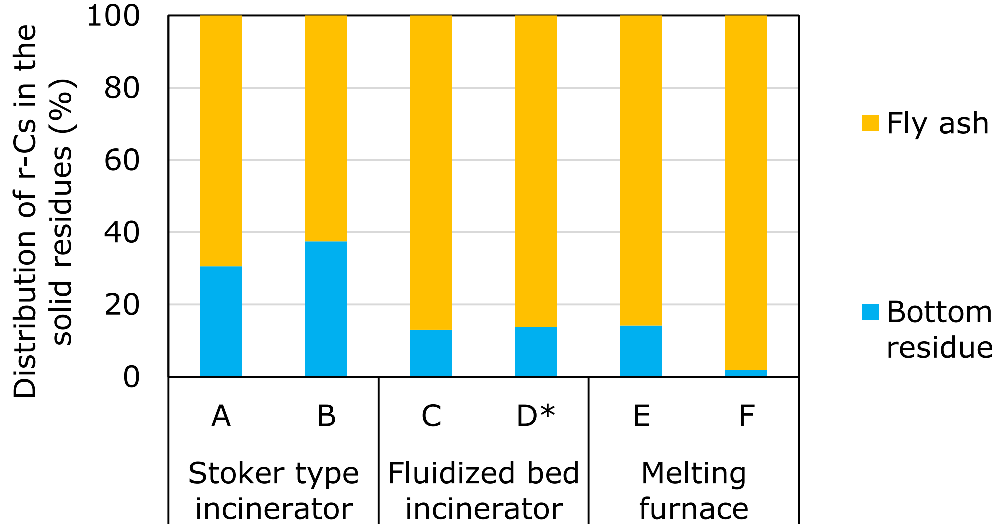

Fig. 1. Distribution of the amount of r-Cs between two solid residues in thermal treatment plants. **[1] Harada et al., 2014*

Figure 2 shows the result of leaching test. The leaching rate of r-Cs from fly ash ranged from 40% to 100%. Unlike fly ash, most of r-Cs did not leach from bottom residue. These results were in good agreement with our previous study on the leaching test for other incineration ashes [2]. It is important to pay attention to the storage and disposal of r-Cs-containing fly ash regardless of thermal treatment type. We try to predict the chemical form of thermal treatment residue from the leaching property and chemical composition. R-Cs form in the residue discharged from furnace bottom is considered to be an insoluble mineral form such as pollucite or a slag phase. On the other hand, r-Cs in fly ash exists as a water-soluble chemical such as cesium chloride because the chlorine (Cl) content in fly ash is much higher than that in bottom ash by one order of magnitude.

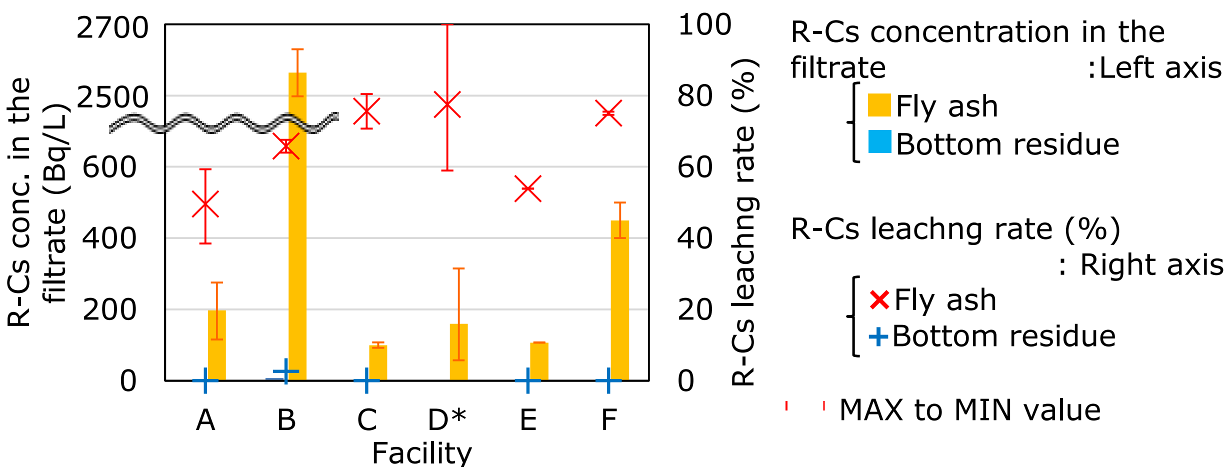

Fig.2 The result of leaching test from two solid residues. **[1] Harada et al., 2014*

Removal efficiency of baghouse (BH) for r-Cs was evaluated by measurement of concentration of r-Cs in flue gas before and after BH. In all the facilities, the presence of r-Cs in flue gas after BH was not detected in this investigation. The detection limits are much lower than the concentration limit in the air by two or three orders of magnitude. Therefore, this demonstrates that the flue gas after BH is safe in any case. The r-Cs removal efficiency was calculated with the following equitation.

$$\text{R-Cs removal efficiency (\%)} = \left(1 - \frac{\text{amount of r-Cs in flue gas after BH (Bq/h)}}{\text{amount of r-Cs in flue gas before BH (Bq/h)}}\right) \times 100 \qquad \text{(Eq.1)}$$

In the calculation, we assumed that the r-Cs concentration after BH was equal to the detection limit. The results are shown in TABLE II. The removal efficiency often achieved over 99.9%. Although the efficiency values in some cases are below 99.9%, it is due to the low concentration in the flue gas before BH and high value of detection limit after BH. It should be noted that eq. 1 cannot fairly evaluate the r-Cs removal efficiency when the low concentration in the flue gas before BH and high value of detection limit after BH. For example, "98.36< %" does not stand for "below 99%". Therefore, we consider that BH can remove r-Cs in flue gas efficiently independent of the thermal treatment type because r-Cs removal rate were the similar level in all facilities. From another point of view, r-Cs gas generated during combustion of contaminated MSW is considered to be solidified in surface of fly ash since BH operated below 200°C to avoid dioxin resynthesis. Hence, the use of a BH treatment for fly ash can prevent the emission of r-Cs to the environment.

TABLE II. R-Cs removal efficiency of baghouse (BH) and r-Cs concentration in flue gas.

Facility	**R-Cs concentration (Bq/Nm^3-dry)**				**R-Cs removal efficiency [b] (%)**	
	Before BH		**After BH [a]**			
	Cs-134	**Cs-137**	**Cs-134**	**Cs-137**	**Cs-134**	**Cs-137**
A	16 [c]	22 [c]	<0.015	<0.017	99.86<	99.88<
B	52 - 68	84 -118	<0.015	<0.013-<0.014	99.97<-99.98<	99.98<-99.99<
C	1.8-4.7	3.7 -9.4	<0.012-<0.020	<0.011-<0.020	98.83<-99.76<	99.43<-99.89<
D	13 - 50 [d]		<0.104 -<0.36 [d]		98.36< -99.61< [e]	
E	No analysis		<0.012-<0.013 [f]	<0.011-<0.012 [f]	No analysis	
F	7.1 -9.8	12 - 17	<0.010	<0.010	99.87<-99.90<	99.92<-99.94<

[a] *"<" means value of detection limit*, [b] *"<" means the value thought to be minimum*, [c] *measured before cooling tower*, [d] *[1] Harada et al., 2014*, [e] *calculated from concentration of r-Cs based on the hypothesis that flow rate before and after BH were the same*, [f] *measured at stack.*

CONCLUSION

We investigated the distribution of r-Cs between fly ash and bottom residue for several thermal treatment facilities dealing with contaminated MSW. The r-Cs distribution and r-Cs concentration in the solid residues depended on type of treatment. Fly ash has a high r-Cs leaching rate whereas bottom residue showed quite a low leaching property. From measurements of r-Cs concentration in flue gas before and after baghouse for all the plants, r-Cs in flue gas can be removed efficiently, and thus there is no release of r-Cs through the thermal treatment plants with the conventional air pollution control system such as baghouse. These results will be useful for the operation and handling the highly contaminated residues at the post-treatment site. The knowledge presented here will not only apply for incineration of decontamination waste in Fukushima, but also provide useful countermeasures for unexpected widespread diffusion of radioactive or chemical substance.

REFERENCES

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