

Emission behaviour and Hg speciation from waste incinerators

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Abstract

The control of Hg emissions from waste incinerators is very important in Japan, because more than 80% of organic waste is incinerated in this country. The Hg content of the coal used in utility boilers is relatively low in Japan. However, there are currently no regulations governing Hg in incineration gas. In this study, the emission behaviour of Hg speciation in Japan and technology for controlling Hg emissions, such as a Bag filter (BF) with activated carbon (AC) injection, are discussed. The Hg speciation at the inlet to the BF has a very high percentage of Hg^{2+} compared with Hg^0 . The average Hg emission coefficients from three incinerators were 0.3g/t at a municipal solid waste incinerator, 0.2g/t at a sewage sludge incinerator and 2.0g/t at an automobile shredder refuse incinerator, respectively. The emission coefficient from the automobile shredder refuse incinerator was especially high. The BF with AC injection removes over 95% of the Hg^{2+} .

Keywords: waste incinerator, Hg^0 , Hg^{2+} , emission coefficient, control.

1 Introduction

Because of its high volatility, Hg is emitted into the atmosphere from both anthropogenic and natural sources. Therefore, it enters oceans and terrestrial waters directly from the atmosphere or even from its deposits in the surrounding basin without a particular source of Hg [1]. Japan has passed the Basic Law for Environmental Pollution Control, which is used as the basis for environmental standards for public water supplies, and the “Prevention of Water Pollution Act”, which provides standards for effluents, in response to two reported cases of



Minamata disease. These laws prescribe the standards concerning total Hg and alkyl Hg, respectively; however, there are currently no regulations governing Hg in incineration gas.

The control of Hg emissions from solid waste incinerators (municipal and industrial solid waste) is very important, because more than 80% of solid waste is incinerated in Japan. The Hg content of the coal used in utility boilers is relatively low in Japan. The overall goal of this study was to determine the emission coefficients of the speciated Hg emitted from three solid waste incinerators and to verify Hg reduction by a BF with an AC injection.

2 Experimental procedures

2.1 Measurement method [2]

A MS-1/DM-5 (Nippon Instruments Corporation) mercury CEM was used to measure the Hg^0 and Hg^{2+} emissions at two different locations in real time. One is at the inlet to the BF and the other is at the outlet of the BF. The MS-1/DM-5 is a portable Zeeman-modulated cold-vapour atomic adsorption instrument.

The MS-1/DM-5 monitors elemental (Hg^0) and oxidized (Hg^{2+}) mercury in stack gas. This continuous mercury speciation analyzer consists of the MS-1 speciation unit and a DM-5 detector. The DM-5 detector is connected to the MS-1 by a linking cable and a 6-millimeter (mm) (0.21-inch) Teflon tube. To measure Hg^0 , sample gas and potassium chloride solution are mixed in a reaction tube to remove water-soluble Hg^{2+} and water-soluble organic mercury. Then, the gas and solution are separated in a gas/liquid separating tube. After the gas is washed with a potassium hydroxide solution and dehumidified, the mercury is guided to the detector where gaseous Hg^0 is measured. To measure Hg^{2+} , the solution containing Hg^{2+} and water-soluble organic mercury is guided to the lower part of the reaction tube in order to be mixed with a reducing solution of tin chloride. There, Hg^{2+} and the water-soluble organic mercury in the solution are reduced to gaseous Hg^0 . The gas is washed by potassium hydroxide solution and dehumidified in an electronic cooler. The mercury is then guided to the detector for measurement.



Figure 1: MS-1.



Figure 2: DM-5.

2.2 Measurement facilities

In this study, we measured three kinds of incinerator that are burning different types of solid waste.

2.2.1 Municipal solid waste incinerator (MSWI)

The measurement was performed in a city in western Japan at a continuous fluidized-bed type MSWI, which has an incineration capacity of 150t/day of MSW. The MSWI is equipped with a secondary combustion chamber and an off-gas treatment system for cleaning combustion effluent gases. A schematic of the MSWI is shown in Figure 3.

The off-gas treatment system of the MSWI produced $\text{Ca}(\text{OH})_2$ and AC injection, a BF and a selective catalytic reduction (SCR). The stack is equipped with several sample ports for flue gas sampling; the continuous emission monitoring system issued for measuring carbon monoxide (CO), carbon dioxide (CO_2), oxygen (O_2) and nitrogen oxide (NO_x).

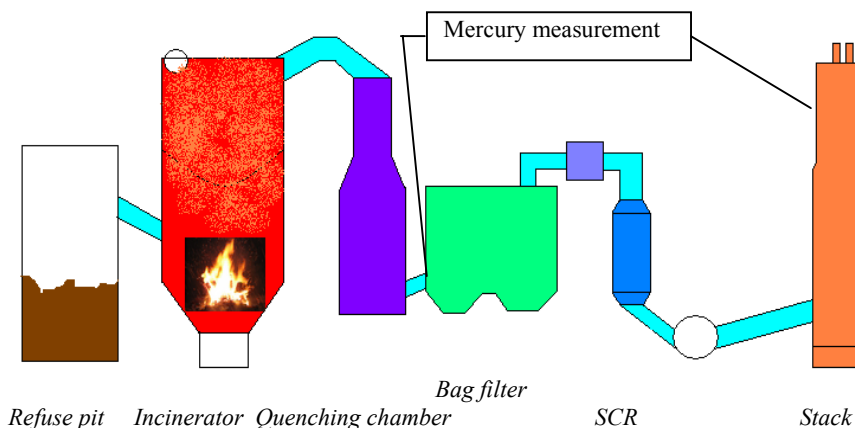


Figure 3: Schematic of the MSWI system.

2.2.2 Sewage sludge incinerator (SSI)

This incinerator is a continuous fluidized-bed type, which has an incineration capacity of 140t/day of industrial waste (sewage sludge and waste plastics). In this facility, an off-gas treatment system for cleaning effluent gas has been installed as shown in Figure 4.

The off-gas treatment system of this incinerator produced urea and $\text{Ca}(\text{OH})_2$, and utilized AC injection and a BF. The device for flue gas sampling at the stack is the same as the MSWI.

2.2.3 Automobile shredder refuse incinerator (ASRI)

The measurement was taken at a continuous fluidized-bed type ASRI, which has an incineration capacity of 75t/day of ASR. The ASRI is equipped with a secondary combustion chamber and off-gas treatment system for cleaning combustion effluent gases. A schematic of the ASRI is shown in Figure 5.

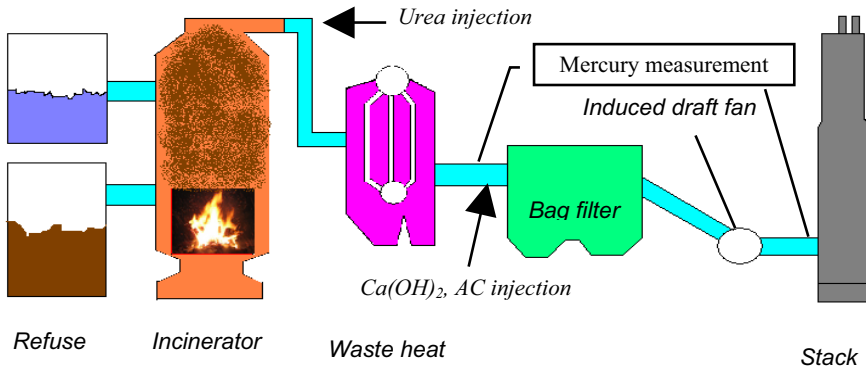


Figure 4: Schematic of the sewage sludge incinerator system.

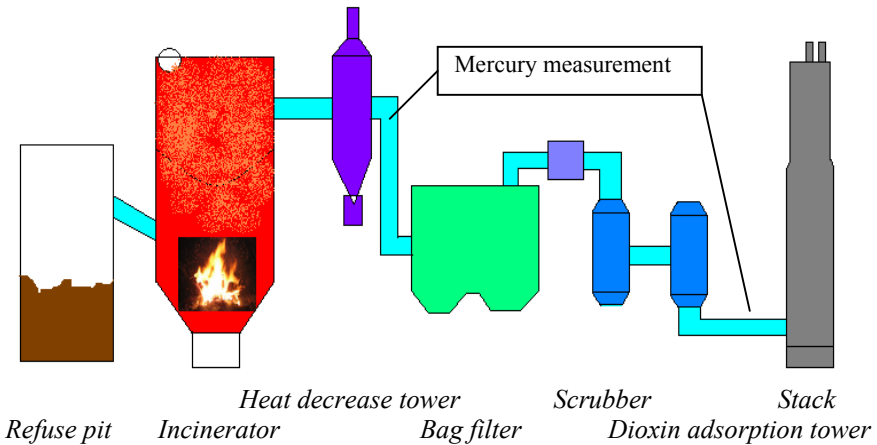


Figure 5: Schematic of the ASRI system.

The off-gas treatment system of the ASRI produced Ca(OH)_2 and AC injection, a BF, a scrubber and a dioxin adsorption tower with AC. The stack is equipped with several sample ports for flue gas sampling; continuous emission monitoring system for measuring CO , O_2 and NO_x .

3 Results and discussion

The measurements were performed over a period of 3 days for each facility. The results were as follows below.

3.1 MSWI

Table 1 presents the running conditions of MSWI during the measurements. The Hg concentrations of mercury CEM at the inlet to BF are displayed in Figure 6.



Figure 6 shows that Hg speciation at the inlet to BF has a very high percentage of Hg^{2+} compared with Hg^0 . The Hg speciation results across the BF are shown in Figure 7. Figure 7 demonstrates that the BF with the AC injection removes a high percentage of the Hg^{2+} .

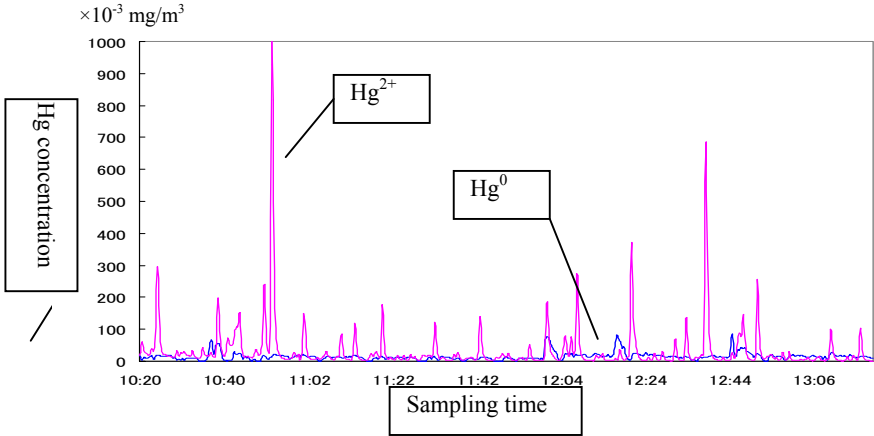


Figure 6: Hg Speciation at the inlet to BF.

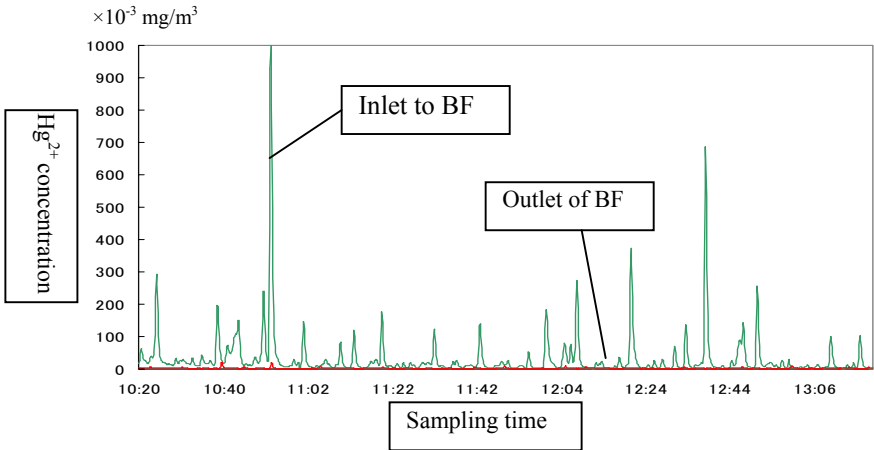


Figure 7: Hg^{2+} concentration across the BF.

Table 1: The running conditions of MSWI during the measurement.

	Amount of incineration (t/h)	Flue gas $\times 10^3(Nm^3/h)$	Combustion temperature ($^{\circ}C$)	Temperature at the inlet to BF ($^{\circ}C$)	Temperature at the inlet to stack ($^{\circ}C$)
Measure	4.0~6.7	29~50	910~1000	135~156	110~130
Average	5.6	41	980	155	120



Table 2: Hg speciation at the inlet to BF.

	Hg ⁰ × 10 ⁻³ mg/m ³	Hg ²⁺ × 10 ⁻³ mg/m ³	Emission (g/h)	Emission coefficient (g/t)
Measure	0.4~23	10~110	0.3~5.1	0.1~0.8
Maximum	23	110	5.1	0.8
Minimum	0.4	10	0.3	0.1
Average	3.6	44	2.0	0.3

Table 3: Hg speciation at the outlet of BF.

	Hg ⁰ × 10 ⁻³ mg/m ³	Hg ²⁺ × 10 ⁻³ mg/m ³	Emission (g/t)
Measure	<0.1~0.6	2.2~3.3	<0.1~0.2
Maximum	0.6	3.3	0.2
Minimum	<0.1	2.2	0.1
Average	0.3	2.4	<0.1

As seen in Table 2, the Hg²⁺ concentration at the inlet to the BF ranged from 10 to 110 × 10⁻³ mg/m³, and the arithmetic mean was 44 × 10⁻³ mg/m³. Furthermore, the Hg mission coefficient from MSWI was 0.3 g/t. Because the Hg content in MSW has decreased [3] the Hg emission coefficient has been considerably reduced compared with the earlier concentration. Fluorescent lamps are estimated to be the largest contributor to the Hg content in MSW in Japan [4]. As shown in Table 3, the Hg²⁺ concentration at the outlet of the BF ranged from 2.2 to 3.3 × 10⁻³ mg/m³, and the arithmetic mean was 2.4 × 10⁻³ mg/m³. The BF with AC injection removes over 95 percent of the Hg²⁺. In Japan, AC injection often is used for dioxin removal.

3.2 SSI

Table 4 presents the running conditions of SSI during the measurements. The Hg concentrations of mercury CEM at the inlet to BF are shown in Figure 8. Figure 8 shows that Hg speciation at the inlet to BF is mainly Hg²⁺. The Hg speciation results across the BF are shown in Figure 9. Figure 9 indicates that the BF with AC injection removes a high percentage of the Hg²⁺.

As seen in Table 5, the Hg²⁺ concentration at the inlet to the BF ranged from 0.8 to 52 × 10⁻³ mg/m³, and the arithmetic mean was 10 × 10⁻³ mg/m³. The Hg mission coefficient from SSI was 0.2 g/t. The emission coefficient from SSI is similar to that of MSWI. Because the Hg content in sewage sludge has decreased, the Hg emission coefficient has become less than one-tenth of that in 1982 [5]. As shown in Table 6, the Hg²⁺ concentration at the outlet of the BF ranged from 0.1 to 5.8 × 10⁻³ mg/m³, and the arithmetic mean was 1.2 × 10⁻³ mg/m³. The BF with AC injection removes over 85 percent of the Hg²⁺.



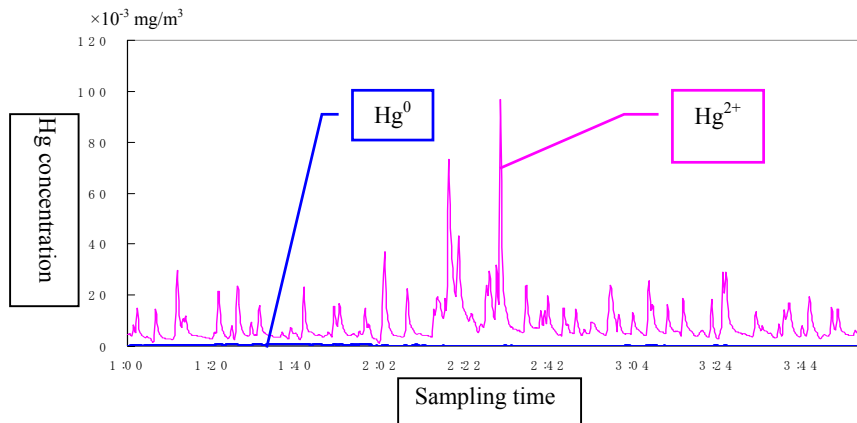


Figure 8: Hg Speciation at the inlet to BF.

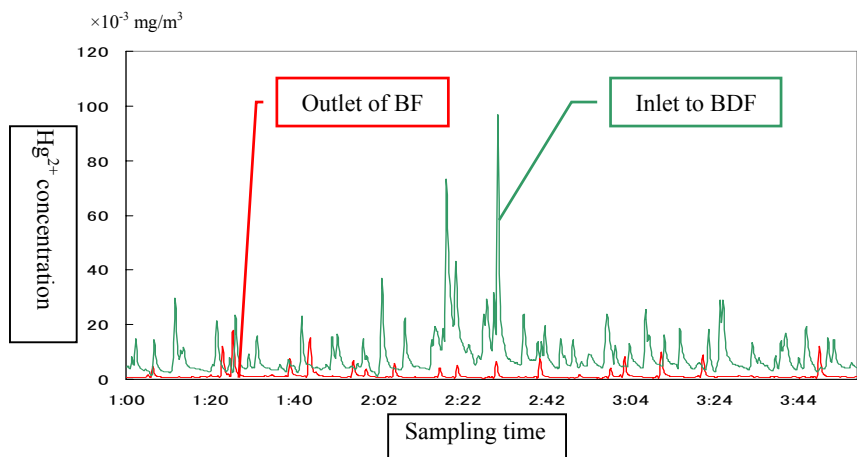


Figure 9: Hg²⁺ concentration across the BF.

Table 4: The running conditions of SSI during the measurement.

	Amount of incineration (t/h)	Flue gas $\times 10^3(\text{Nm}^3/\text{h})$	Combustion temperature ($^{\circ}\text{C}$)	Temperature at the inlet to BF ($^{\circ}\text{C}$)	Temperature at the inlet to stack ($^{\circ}\text{C}$)
Measure	3.2~7.3	44~55	800~1000	182~188	183~188
Average	3.8	47	960	184	185



Table 5: Hg speciation at the inlet to BF.

	Hg ⁰ ×10 ⁻³ mg/m ³	Hg ²⁺ ×10 ⁻³ mg/m ³	Emission (g/h)	Emission coefficient (g/t)
Measure	<0.1~13	0.8~52	<0.1~2.8	<0.1~0.7
Maximum	13	52	2.8	0.7
Minimum	<0.1	0.8	<0.1	<0.1
Average	1.0	10	0.6	0.2

Table 6: Hg speciation at the outlet of BF.

	Hg ⁰ ×10 ⁻³ mg/m ³	Hg ²⁺ ×10 ⁻³ mg/m ³	Emission (g/t)
Measure	<0.1~5.1	0.1~5.8	<0.1~0.3
Maximum	5.1	5.8	0.3
Minimum	<0.1	0.1	<0.1
Average	0.1	1.2	<0.1

Table 7: The running conditions of ASRI during the measurement.

	Amount of incineration (t/h)	Flue gas ×10 ³ (Nm ³ /h)	Combustion temperature (°C)	Temperature at the inlet to BF (°C)	Temperature at the inlet to stack (°C)
Measure	2.9~3.3	24~38	780~940	173~189	75~79
Average	3.1	33	890	183	77

Table 8: Hg speciation at the inlet to BF.

	Hg ⁰ ×10 ⁻³ mg/m ³	Hg ²⁺ ×10 ⁻³ mg/m ³	Emission (g/h)	Emission coefficient (g/t)
Measure	<0.1~1.0	110~420	3.5~15	1.2~4.5
Maximum	1.0	420	15	4.5
Minimum	<0.1	110	3.5	1.2
Average	0.1	180	6.4	2.0

3.3 ASRI

Table 7 presents the running conditions of ASRI during the measurements. The Hg concentrations of mercury CEM at the inlet to BF are demonstrated in Figure 10. Figure 10 shows that Hg speciation at the inlet to BF has a very high percentage of Hg²⁺ compared with Hg⁰. The Hg speciation results across the BF are shown in Figure 11. Figure 11 indicates that the BF with AC injection removes almost all of the Hg²⁺.

As seen in Table 8, the Hg²⁺ concentration at the inlet of the BF ranged from 110 to 420×10⁻³mg/m³, and the arithmetic mean was 180×10⁻³mg/m³. The Hg mission coefficient from ASRI was 2.0 g/t. The emission coefficient from ASRI was especially high. Mercury switches are estimated to be the largest contributor to the Hg content in ASR [6]. As shown in Table 9, the Hg²⁺ concentration at the outlet of the BF ranged from 0.2 to 1.7 ×10⁻³mg/m³, and the arithmetic mean was 0.6×10⁻³mg/m³. The BF with AC injection, a scrubber and a dioxin adsorption tower with AC remove over 99 percent of the Hg²⁺.

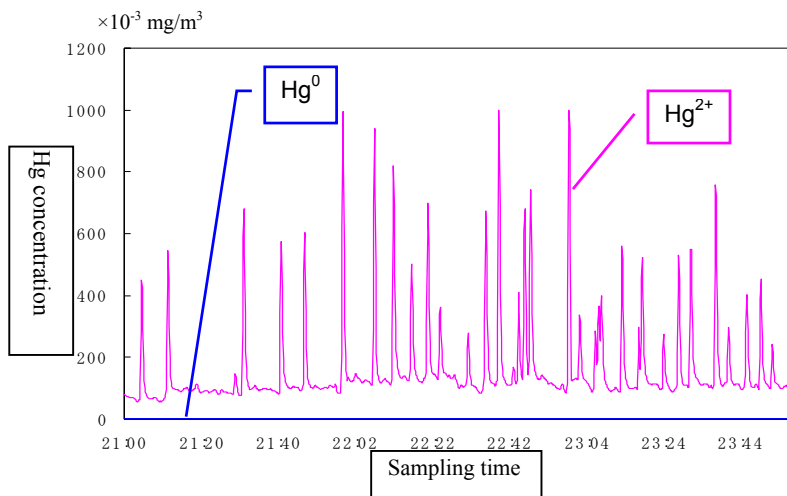


Figure 10: Hg speciation at the inlet to BF.

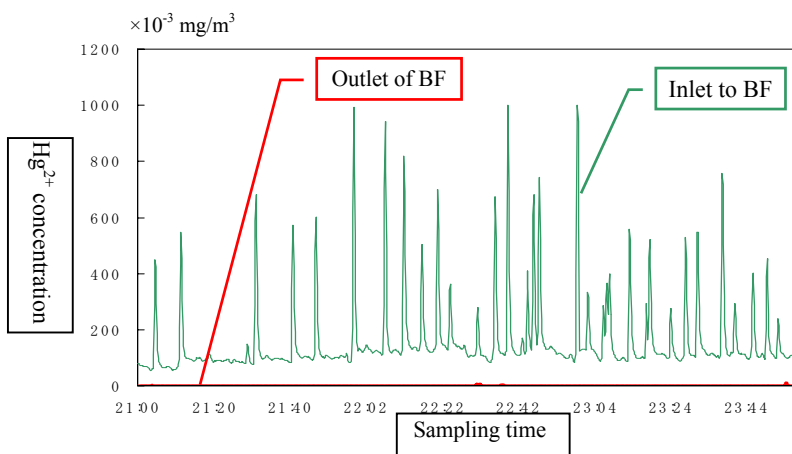


Figure 11: Hg²⁺ concentration across the BF.

Table 9: Hg speciation at the outlet of BF.

	Hg ⁰ × 10 ⁻³ mg/m ³	Hg ²⁺ × 10 ⁻³ mg/m ³	Emission (g/t)
Measure	<0.1~1.7	0.2~1.7	<0.1
Maximum	1.7	1.7	<0.1
Minimum	<0.1	0.2	<0.1
Average	0.1	0.6	<0.1



4 Conclusions

In this study, the emission behaviour of Hg speciation in Japan as well as technology of controlling Hg emission such as a Bag filter with AC injection, are considered. The major results are as follows.

(1) The average concentrations of Hg speciation from each incinerator are Hg^0 0.0036 mg/m^3 , Hg^{2+} 0.044 mg/m^3 in MSWI, Hg^0 0.001 mg/m^3 , Hg^{2+} 0.01 mg/m^3 in SSI, Hg^0 0.0001 mg/m^3 and Hg^{2+} 0.18 mg/m^3 in ASRI, respectively. For the most part of Hg comes in the form Hg^{2+} .

(2) The average Hg emission coefficients from the three incinerators were 0.3g/t at MSWI, 0.2g/t at SSI and 2.0g/t at ASRI, respectively. The emission coefficient from the ASRI was especially high.

(3) The data collected by waste incinerators measurement indicate that mercury removal for plants equipped with BF including AC injection reaches over 95% of the total mercury.

References

- [1] Fitzgerald, W.E., Engstrom, D.R., Mason, R.P. and Nater, F.A.; *Environ. Sci. Technol.* **32**, pp.1–7, 1998.
- [2] *Environmental Technology Verification Report*; Nippon Instruments Corporation MS-1/DM-5 Mercury Continuous Emission Monitor, U.S. EPA, September 2003.
- [3] *Report Concerning MSWI*; Kyoto Municipal Government: Kyoto Japan, 1999.
- [4] Japan Electro Lamp Manufactures Association; *Statistical information 2001–2005*.
- [5] Yasuda, K., Ootsuka, Y. and Kaneko, M.; *J. Japan Soc. Air Pollut.*, **18**, pp.286–290, 1983.
- [6] Wisconsin Mercury Sourcebook: *The Automotive Sector, 2000*.

