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Article Title: Emission of particulate matter 2.5 (PM2.5) from sewage sludge incinerators in Japan

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ABSTRACT

Because fine particulate matter ≤2.5 μm in diameter (PM2.5) causes health problems, PM2.5

- emissions are of concern. However, little research on stationary sources has been conducted. To
- determine the concentration and filtration behaviour of PM2.5, dust was collected from five fluid
- bed sewage sludge incinerators (SSIs) sorted by particle size using cascade impactors. The
- 6 average PM2.5 concentration was $0.00014-4.8$ mg/Nm³. The total estimated amount of PM2.5
- emissions from the SSIs for all plants in Japan was 0.96–8.9 tons/year. Since the SSIs with dry
- Electrostatic Precipitators (EP) contributed 75–99% of the total emissions, replacing dry EPs with
- Bag Filters would significantly reduce the PM2.5 emissions from SSI.
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KEYWORDS

PM2.5, sewage sludge incinerator, bag filter, electrostatic precipitator, emission

INTRODUCTION

15 In recent years, fine particulate matter ≤ 2.5 µm in diameter (PM2.5) has attracted increasing attention 16 because of its health risk and the high PM2.5 concentrations in some rapidly industrialising areas.^[1,2] Epidemiological studies have demonstrated a relationship between mortality and long-term exposure to 18 PM2.5 in ambient air.^[3,4] Many international organisations have assessed the health risks of exposure to PM, and the European Union, United States, and World Health Organisation (WHO) established quality 20 standards for ambient air in 2005^[5] and 2006.^[6,7] In Japan, PM2.5 was added to the Japanese environmental

- 21 standards in September 2009. These hold that the annual average PM2.5 concentration should be less than
- 22 15 μ g/m³ and the daily average less than 35 μ g/m³.

 Recently, the multinational European EBoDE-project estimated that particulate matter (PM) was associated with the highest disease burden (6 000–10 000 DALYs per million people), followed in order by second-hand smoke, noise, and radon. Disability-adjusted life years (DALYs) are a summary measure 26 of population health combining mortality and morbidity.^[8] The International Agency for Research on 27 Cancer (IARC) also classified PM as carcinogenic to humans.^[9]

 As primary particles, sources of PM2.5 include both anthropogenic sources such as combustion plants and vehicle emissions, and natural sources such as yellow sand, etc. It is also notable that secondary particles produced from gases affect atmospheric PM2.5 levels. However, few studies have determined PM2.5 levels from sources other than ambient air and most of the studies of PM2.5 sources have examined 32 mobile sources, with few direct measurements of PM2.5 in flue gases.^[10-12] To prevent PM2.5 emissions, it is necessary to examine the present state of PM2.5 emissions from stationary sources. We have 34 investigated some stationary sources of PM2.5 in recent years.^[13,14]

 In Japan, sewage sludge represents one of the largest sources of industrial waste, with amounts produced increasing with the increase in the population using sewage treatment systems. The overall treatment 37 system produces ~2.2 million tons of dry sewage sludge (Dried Sludge; DS) annually.^[15] Incineration techniques are widely used for disposal of sewage sludge, this reduces the volume of sludge and transforms waste into solid incineration ash, which facilitates easy waste handling. Due to the small land area available

- in Japan, it is difficult to dispose of large quantities of sludge without intermediate treatment. Therefore,
- ~68% (1.5 million tons-DS) of sludge was incinerated in sewage sludge incinerators (SSIs) in 2008. [15]
- 12 Incineration emits toxic substances such as dioxin $[16]$ and mercury $[17]$ as well as large quantities of dust
- and PM2.5. However, no recent study has measured PM2.5 emissions from SSIs.
- This study focused on the emission of PM2.5 from SSIs. We collected dust samples from five SSI plants according to particle size using an Andersen stack sampler to evaluate the concentration, removal ability, emission factor, and emission mass of PM2.5 from SSIs. This paper assumes that PM2.5 from SSI is the primary particle in the SSI flue gases.
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MATERIALS AND METHODS

Sewage Sludge Incineration plants

 Dust sampling was carried out at five continuous fluidized-bed-type SSI plants with different dust collectors. Two of the five plants had dry electrostatic precipitators (EP; plants EP1 and EP2), and the remaining three plants had both EP and wet EP (plant EP+WEP), bag filter (BF; plant BF), and ceramic filter (CF; plant CF), respectively. Generally, EP removes dust from flue gases using an electrostatic force. The removal efficiency for 10 μm particles is more than 99.5%. WEP is an EP that includes cleaning equipment that rinses gases with water, which results in removal efficiency better than that provided by EP alone. Fabric filters such as BF and CF remove dust from flue gases by filtration. This type of filter has much higher dust removal efficiency than EP has, particularly for fine particles. The temperature resistance 59 of CF is better than that of BF due to the materials used in their manufacture.^[18] All of the incinerators had 60 fluidised beds, a popular type of incinerator for sewage sludge in Japan.^[19] The fluidized beds are heated to an annual average temperature between 820 and 854°C, and sewage sludge burns continuously at a rate that depends on the incineration capacity of each plant. The sewage sludge in cake form is partially dewatered by a dewatering system such as a belt filter press (plant EP1, EP2, and EP+WEP), centrifuge (plant BF), screw press and indirect heating (plant CF). The sludge cakes contained 74.6 to 84% moisture when wet and 70.6 to 86.0% organic matter content when dried; the composition of the sludge tended to 66 be similar to each other and in good agreement with previous reports. $[20-24]$ Among these plants, operational conditions are similar with respect to the incinerator, coagulant, combustion temperature, and organic content in sludge cake. Figure 1 and Table 1 show a summary of the plant flow and an outline of technical data, respectively, for each SSI plant.

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- FIG. 1. Plant flow and sampling points
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- 74 TABLE 1 Technical data for five SSI plants^[25]
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- **Sampling**

 Sampling was carried out at five SSI plants: plants EP1, EP2, EP+WEP, BF, and plant CF, which were sampled during on May 12-14, 2010, November 19-21, 2012, May 26-28, 2010, July 11-13, 2012, and November 17-19, 2010, respectively. In all plants, samples were collected from the dust collector and stack inlets to evaluate the proportion removed by the air pollution control devices (APCDs). Fig. 1 shows the sampling points. Dust samples in SSI flue gas were collected isokinetically in nine particle size fractions (from submicron to approximately 10 µm) using Andersen stack samplers (AS-500, Tokyo Dylec, Japan) inserted into the flue gas ducts of five SSI plants. The sampling methods were based on Japanese Industrial 85 Standards (JIS) Z8808 and K0302.^[26,27] The representative particle size at each stage of the cascade impactor is defined as the aerodynamic particle size at which 50% separation is achieved and was obtained using the following equation:

$$
f_{\rm{max}}
$$

 $D_{\text{p50n}} = -1.26 \times \lambda + \sqrt{1.58 \times \lambda^2 + \frac{1.08 \times \pi \times N \times \Psi_{50} \times D_{on}^2 \times (172 + 0.4 \times \theta_{s}) \times 10^{-1}}{4 \times q_{s}}}$ (1)

- 90 *D_{p50n}*: 50% separation particle size at the n^{th} stage (um)
- 91 *N*: number of jet nozzles at the nth stage
- 92 D_{cn} : bore of the jet nozzles at the n^{th} stage (mm)
- *Ψ50*: inertia parameter for 50% separation by particle size
- 94 θ_s : flue gas temperature (°C)
- *qs*: suction flow rate at the suction nozzle (L/min)
- *λ*: mean free path of a gas molecule (μm)
-
- 98 In this formula, the mean free path of a gas molecule (λ) was obtained using equation (2).

99 $\lambda = 2.10 \times 10^{-5} \times (172 + 0.4 \times \theta_s) \times \sqrt{(273 + \theta_s)}$ (2)

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- A period of at least 5 min was required for sample collection at the dust collector inlets and 44 h at the 102 stack inlets. Two impingers (one without solution and the other with 5% H_2O_2) were placed behind the samplers to remove moisture and to absorb any gaseous substances in flue gas that had removed dust. 104 Samplings was carried out under stable conditions. Table 1 and FIG. 2. summarize the sampling conditions and sampling design, respectively.
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- TABLE 2 Sampling conditions
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- FIG. 2. Design for sampling dust in flue gas
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- **Analysis**
- 114 The quartz filters used for sampling were heated at 250°C for 2 h and cooled to room temperature before
- sampling to remove volatile substances. Before and after sampling, each filter was dried and weighed in a
- clean room kept at 21.5ºC (upper: 23ºC , lower: 20ºC) and 35% relative humidity (upper: 40%, lower: 30%), using microbalances (M5P-F, Sartorius, USA, or XP26, METTLER TOLEDO, USA) at a sensitivity of 1 μg. To determine the PM2.5 fraction exactly, the fraction up to a diameter of 2.5 μm was divided linearly according to the upper and lower diameters defining the fraction.
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RESULTS AND DISCUSSION

Dust concentration

 Figure 3a shows mass distribution versus aerodynamic diameter at the inlet of the stack. In particular, more than 90% of dust collected from the stacks of EP1 and EP2 consisted of particles under 3 μm. Although 55-90% of the stack dusts from WEP and CF also tended to consist of particles under 3 μm, 126 some stack dust fractions from WEP and CF and most from BF were collected in only small amounts, and their weight was negligible. Figure 3b shows mass distribution versus aerodynamic diameter at the inlet of the dust collector. Of the dusts collected from the collectors of EP1, EP2, and WEP, 80-90% consisted of particles under 10 μm. In contrast, 40-70% of dusts collected from the dust collectors of BF and CF tended to consist of particles over 5 μm. Pretreatment of flue gases with a cyclone in some plants can account for 131 this differences in size distribution at the dust collector.^[18]

 FIG. 3. Mass distribution of dust. (a) at inlet of stack, (b) at inlet of dust collector. Dp: aerodynamic diameter.

 The total dust and PM2.5 concentrations at the dust collector and stack inlets at each SSI plant are shown 138 in Figure 4. The total dust and PM2.5 concentrations were $\leq 0.32-5,000 \pm 670$ and $\leq 0.14-4,800 \pm 730$ μ g/Nm³ (mean \pm mean deviation), respectively, in the flue gas in the stacks. Ehrlich et al. and Tirler et al. reported that the PM2.5 concentrations in German and Italian waste incineration plants were 752 and 32.9 141 μ g/Nm³, respectively ^[10,12]. Our results for plants WEP, BF, and CF were within the same or smaller range, 142 while the results for plants EP1 and EP2 were considerably larger. More than 55% of total dust in the stack 143 consisted of PM2.5. This result is similar to those reported by Ehrlich et al.^[10] and Bounanno et al.^[11] The PM2.5 concentration at the stack inlet of plant BF was less than the annual average Japanese environmental 145 standard (15 μ g/m³). Plant BF operated a state-of-the-art APCD, and the PM2.5 concentrations remained 146 less than $0.14 \mu g/Nm^3$. In contrast, the PM2.5 and total dust levels at plants EP1 and EP2 were considerably higher than those at the other plants. Similar results were reported previously for municipal solid waste 148 incinerators [13,14]. The respective total dust and PM2.5 concentrations in the dust collector inlet were $1.5 \pm$ 149 0.5 and 0.43 ± 0.13 g/Nm³ in plant EP1, 0.45 ± 0.8 and 0.32 ± 0.7 g/Nm³ in plant EP2, 4.4 ± 0.3 and 1.9 ± 1.6 150 0.1 g/Nm³ in plant EP+WEP, 5.6 \pm 0.3 and 0.51 \pm 0.09 g/Nm³ in plant BF, and 3.9 \pm 1.0 and 0.86 \pm 0.09 151 g/Nm³ in plant CF. The BF, WEP, and CF removed PM2.5 efficiently, but not the EP.

FIG. 4. Total dust and PM2.5 concentrations in the dust collector inlet

and the stack inlets at SSI plants ; error bar is mean deviation.

Removal of PM2.5 by APCD

 The removal efficiency of PM2.5 and dust from the plants was investigated to determine the relationship of PM2.5 and the dust concentration in the stack inlet for different dust collectors, where the removal efficiency was calculated as follows:

$$
\eta = (1 - \frac{C_{oN} Q_{oN}}{C_{iN} Q_{iN}}) \times 100
$$
\n
$$
(3)
$$

η: removal efficiency (%)

- 163 *C_{iN}*: dust or PM2.5 concentrations at the dust collector inlet of each plant (g/Nm³)
- 164 C_{oN} : dust or PM2.5 concentrations at the stack inlet of each plant (g/Nm³)
- 165 Q_{iN} : flow rate of flue gas at the dust collector inlet of each plant (Nm³/h)
- 166 $Q_{\text{o}N}$: flow rate of flue gas at the stack inlet of each plant (Nm³/h)
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 The results are shown in Table 3. Plants EP+WEP, BF, and CF removed more than 99.99% of the PM2.5 and total dust, demonstrating that those dust collectors are effective for particle emission control. Most importantly, BF was the best dust collector in terms of both the dust concentration in the stack inlet and removal efficiency. The EP at plants EP1 and EP2 were the least efficient collectors. These findings are 172 similar to the results of studies of municipal solid waste incinerators [13,14], in which the PM2.5 removal efficiency of BF was higher than that of EP. The European Environment Agency (EEA) reported that the PM2.5 removal efficiency was 77–98% for industrial waste incineration, including hazardous waste and 175 sewage sludge with some APCDs.^[28] In 1996, a U.S. Environmental Protection Agency (US EPA) report ^[29] suggested that advances in incineration and APCD technology have improved removal efficiency. Replacing dry EP with BF would improve the PM2.5 removal in SSIs.

Table 3 Removal efficiency of total dust and PM2.5 by APCDs

Calculating the PM2.5 Emission Factor from SSIs

 To evaluate PM2.5 emissions from SSIs in Japan, we calculated the emission factor for PM2.5 from our results although there were insufficient samples taken at each SSI plant. We surveyed the number and incineration capacity of SSIs with each type of dust collector, and calculated the PM2.5 emissions. These methods are described in detail below. Dust removal efficiency is generally known to be higher in 187 WEP, BF, and CF than in EP.^[18] Temperature resistance differes between BF and CF. Because these dust collectors have different characteristics, we calculated the emission factor for each dust collector separately (EP, WEP, BF, and CF). The factor for EP was the average of those calculated for plants EP1 and EP2. In this study, the emission factor is defined as the weight of PM2.5 per ton of sewage sludge incineration capacity. The emission factor is defined by the following equation:

$$
Ef = \frac{24 \times Q \times A}{10^6 \times B}
$$
 (4)
193 *Ef*: emission factor, weight of PM2.5 per ton of the incineration capacity (g/ton-sludge cake)
2: flow rate of flue gas at the stack inlet in the plant (Nm³/hour)
3.4: concentration of PM2.5 at the stack inlet (µg/Nm³)

B: incineration capacity of the plant (ton-sludge cake/day)

 The resulting emission factors are shown in Table 4. The PM2.5 emission factor for the SSI with EP was 8.7 g/ton-sludge cake, whereas those for wet EP, BF or CF were less than 0.26 g/ton-sludge cake and BF had the lowest value. The US EPA has set 0.18 kg/Mg-DS as the PM2.5 emission factor for a fluidised bed incinerator for SSI with a scrubber; this equals 180 g/ton-DS or approximately 36 g/ton-sludge cake 203 based on an 80% water content.^[30] The EEA set 0.004 kg/Mg-waste (4 g/ton-waste) as the default PM2.5 204 emission factor for industrial waste incineration, including hazardous waste and sewage sludge.^[28] Although the units and materials burned in these other studies differed, the results were similar to the values set by the US EPA and EEA, and the values for WEP, BF, and CF were considerably smaller. Therefore, advances in technology could improve the removal efficiency.

Table 4 Emission factors of PM2.5

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 The results of the survey of the number and incineration capacity of SSI plants with each type of dust collector (EP, WEP, BF, and CF) are shown in Table 5. This was based on a survey of SSI plants, to which 85 plants responded. The total incineration capacity was 9,950 ton/day, which is 48% of the total number 215 of plants and 39.3% of total plant capacity, for all of the SSI plants in Japan.^[31]

Table 5 Results of survey on SSI plants and calculated PM2.5 emission values

The PM2.5 emissions are defined by the following equation:

222
$$
E = \sum_{i=1}^{4} \frac{Ef \times C}{10^6} \times \frac{Xi}{100}
$$
 (5)

Ef: PM2.5 emission factor from the SSIs (g/ton-sludge cake)

C: amount of sewage sludge burnt in SSIs annually (ton-DS/year)

Xi: relative incineration capacity of SSI plants with each type of dust collector (%)

 The emission results are shown in Table 6. These calculations are outlined in detail below. There were 230 92 plants with no available information on dust collector type. We assumed that all of these 92 plants were equipped with EP, WEP, BF, or CF, representing cases I, II, III, and IV, respectively (in Table 5). The amount of sewage sludge burnt annually in SSIs is 1.5 million tons-DS/year, or 68% of the 2.2 million 233 tons-DS/year produced annually; these amounts are on a dry weight basis.^[15] It is possible that the PM2.5 emissions are underestimated. The total estimated emissions of PM2.5 from SSIs were 0.96–8.9 tons/year, of which more than 75% was from SSIs equipped with EP. Sugiyama et al. estimated that the total PM2.5 236 emissions in 2000 in Japan were 252 kton, of which 49% was from mobile emission sources.^[32] Kannari et al. estimated that anthropogenic PM2.5 emissions in 2000 in Japan equalled 147 Gg, of which 12% was 238 due to waste incineration and field burning.^[33] The PM2.5 emissions from SSIs in our study was less than 0.0035% of the total estimated by Sugiyama et al. and less than 0.006% that of Kannari et al., so the contribution of SSIs to the total PM2.5 emissions is negligible. The emission results in our study could be underestimated because of the difference in units between Ef (g/ton-sludge cake) and C (ton-DS/year), the difference in the real amount of burnt sewage sludge, and incineration capacity. Because most of the PM2.5 emissions might originate from plants with EP, replacing dry EP with BF could significantly reduce the PM2.5 emissions from SSIs.

- Table 6 Calculated PM2.5 emission values
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CONCLUSIONS

 This study measured the concentrations and removal efficiency of PM2.5 and total dust in flue gas from five SSI plants. The average PM2.5 concentration in the stack inlet of the plant with BF was lower than 253 the Japanese environmental standard for ambient air (15 μ g/m³ annually). In the plants with WEP or CF, 254 the PM2.5 concentrations in the stack inlet were also close to the environmental standard (35 μ g/m³ daily). The removal efficiencies of the plants with these dust collectors exceeded 99.99%. The emission of PM2.5 as primary particles from these plants was very low. By contrast, the plants with EP not only had the 257 highest PM2.5 concentrations in the stack inlets but also the lowest PM2.5 removal. The PM2.5 emission factor for SSIs using EP was 8.7 g/ton-sludge cake, whereas that with wet EP, BF, or CF was less than 0.26 g/ton-sludge cake. The total estimated emission of PM2.5 from SSI in Japan was 0.96–8.9 ton/year, which was less than 0.0035% of the total PM2.5 emissions and 0.006% of the anthropogenic PM2.5 emissions estimated for Japan for 2000, although these emissions could underestimates. Since the SSIs with dry EP contributed 75–99% to the total emissions, replacing a dry EP with BF would significantly reduce the PM2.5 from SSIs.

- The results suggest that PM2.5 emissions as primary particles from some SSI plants were very low.
- 265 Future work should determine the contribution of secondary particles from gaseous substances such as $SO₂$,
- HCl, and NO in ambient air.
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374 FIG. 3. Mass distribution of dust. (a) at inlet of stack, (b) at inlet of dust collector. Dp: aerodynamic 375 diameter.

382

384 TABLE 1

385 Technical data for five SSI plants

EP: Electrostatic Precipitator , WEP: Wet Electrostatic Precipitator , BF: Bag Filter, CF: Ceramic Filter

(a): Sum amount contained other lines in the plant

389 TABLE 2

390 Sampling conditions

391 EP: Electrostatic Precipitator , WEP: Wet Electrostatic Precipitator , BF: Bag Filter, CF: Ceramic Filter

403 TABLE 5

- 404 Result of survey on SSI plants and calculated PM2.5 emission value; caseI-IV are assumed cases
- 405 because there were 92 plants with no available information on dust collector type. In case I, II, III, and
-

406 IV all of the 92 plants are assumed to be equipped with EP, WEP, BF, or CF, respectively

Dust collector type	Number of plants	Sum of incineration capacity	Ratio of incineration capacity (%)			Use of emission factor	
		(ton-sludge cake/day)	Case I	Case II	Case III	Case IV	(g/ton-sludge cake)
Cyclone, EP	24	1750	67.6	6.9	6.9	6.9	8.7
Cyclone, EP, WEP	14	2200	8.7	69.4	8.7	8.7	0.26
BF	34	3500	13.8	13.8	74.5	13.8	$< 5.9 E-7$
CF	13	2500	9.9	9.9	9.9	70.6	0.17
No information	92	15383	all of EP	all of $EP+WEP$	all of BF	all of CF	$\overline{}$
Whole of Japan	177	25333	100	100	100	100	$\overline{}$

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EP: Electrostatic Precipitator , WEP: Wet Electrostatic Precipitator , BF: Bag Filter, CF: Ceramic Filter

408

EP: Electrostatic Precipitator , WEP: Wet Electrostatic Precipitator

BF: Bag Filter, CF: Ceramic Filter

414 415

416

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- speakers of English. For a certificate, please see:
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