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Behavior of heavy metals in air pollution control devices of 2,400 kg/h municipal solid waste incinerator

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Abstract−We analyzed the behavior of heavy metals, such as Cd, Cr, Cu, Ni, Pb, Zn, and Hg, in air pollution control devices of a municipal solid waste incinerator. For this study, a municipal solid waste incinerator with a burning capacity of 2,400 kg/h was selected. A semi-dry reactor (SDR), fabric filter, and wet scrubber were installed to serve as air pollution control devices. Flue gas was sampled upstream and downstream of each air pollution control device to determine the heavy metal concentrations therein. Ash was collected from the furnace, boiler, SDR, and fabric filter to determine the heavy metal concentration in the ash produced by each device. Each heavy metal was found to have a different fate in the incinerator and air pollution control devices. Cd and Pb were mostly present in the fabric filter ash, whereas Cr, Cu, and Ni were most prevalent in the bottom ash of the furnace and boiler, and Zn was present in the bottom and fabric filter ash at a ratio of 7 : 3. However, only a few percent of Hg was identified in the ash from the furnace, boiler, SDR, and fabric filter; the majority of Hg passed through the fabric filter and existed in an oxidized form. The wet scrubber exhibited high control efficiency for oxidized mercury, and the injection of commercial activated carbon at a rate of 0.2 g/Sm^3 resulted in 93.2% mercury removal efficiency.

Keywords: Heavy Metals, Municipal Solid Waste Incinerator, Air Pollution Control Devices, Activated Carbon

INTRODUCTION

Heavy metals are emitted during the incineration of municipal solid waste and are hazardous to human health and the environment [1]. Municipal solid waste is a heterogeneous material containing paper, plastic, and organic as well as inorganic materials, such as glass, metals, and dirt, all of which contain heavy metals. Metals contained in this waste may be transferred into fly ash, and bottom ash, as oxides, chlorides, sulfates, nitrates, etc [2]. Furthermore, certain heavy metals may be emitted into the stack [3]. Evan and Williams reported on the range of municipal solid waste heavy metal content as consisting of 10-40 mg/kg cadmium (Cd), 100-450 mg/kg chromium (Cr), 450-2,500 mg/kg copper (Cu), 50-200 mg/kg nickel (Ni), 750-2,500 mg/kg lead (Pb), 900-3,500 mg/kg zinc (Zn), and 2-7 mg/kg mercury (Hg) [4]. Reimann also reported the heavy metal content of the municipal solid waste as 3-15 mg/kg Cd, 250 mg/kg Cr, 200-600 mg/kg Cu, 80 mg/kg Ni, 430-1,200 mg/kg Pb, 1,000-2,000 mg/kg Zn, and 2-5 mg/kg Hg [5]. It has been reported that every heavy metal exhibits a different fate in waste incinerator and air pollution control devices [5,6]. Field test results demonstrated that Cr, Cu, and Ni were found mostly in the bottom ash of the incinerator, while relatively larger amounts of Cd, Pb, and Zn were found in fly ash [7-11]. Nakamura et al. identified some Pb and Cd in the flue gas of the stack, [12] while Kuo et al. found Cr in the flue gas downstream of the filter bag [13].

Among these heavy metals, Hg is highly volatile and present in a gaseous form in the flue gas. In particular, the elemental form of mercury is seldom captured by existing air pollution control devices [14-16]. Approximately 8% of the total anthropogenic mercury is released from municipal waste disposals to the environment worldwide [17]. A field test of eight municipal solid waste incinerators was conducted to measure the concentrations of oxidized mercury (Hg^{2+}) and elemental mercury (Hg^0), respectively, in the flue gas [18]. The results indicated that a larger portion of mercury was present as oxidized forms in the flue gas. This phenomenon was also reported in several other studies on the mercury speciation for municipal waste incinerators [19-21]. Chang et al. determined the mercury removal efficiency of air pollution control devices in two municipal waste incinerators [21]. Dry lime scrubbing with a fabric filter exhibited a higher Hg^0 removal efficiency than that of the combination of an electrostatic precipitator and wet scrubber. It has been suggested that a higher concentration of hydrogen chloride (HCl) in the flue gas results in higher mercury oxidation [22-25]. In addition, mercury removal was correlated with the unburned carbon and chlorine content of fly ash [24,26]. Takaoka et al. demonstrated that the injection of activated carbon upstream of the fab-

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ric filter increased mercury removal efficiency by 20-30% points, compared to that without injection of activated carbon [27]. In another study, activated carbon showed 71.9% mercury removal efficiency across dry/semi-dry scrubber and fabric filter [28].

Although a number of studies have been conducted on the emission and control of heavy metals from municipal solid waste incinerators, very few have investigated heavy metal behavior for the different air pollution control devices. Therefore, we selected a municipal solid waste incinerator equipped with a semi-dry reactor (SDR), fabric filter, and wet scrubber for investigation in this study. Flue gas was sampled from several points upstream and downstream of each air pollution control device, and the heavy metal concentrations in the flue gas were determined. Furthermore, the ash produced from each air pollution control device was collected and analyzed to determine its heavy metal concentrations. Using these concentrations, the fate of each type of heavy metal was investigated for all air pollution control devices. Injection of activated carbon upstream of the fabric filter was selected as the mercury control technology, and its mercury removal efficiency was determined.

MATERIALS AND METHODS

1. Plant

A stoker-type incinerator with a burning capacity of 2,400 kg/h was selected. A schematic diagram of the incinerator and air pollution control devices is provided in Fig. 1. The temperature inside the furnace was designed to be maintained at more than 950 °C. Municipal solid waste samples were obtained, the analysis results of which are displayed in Table 1. The heat generated from the furnace was recovered in the boiler system, and the temperature decreased to 250-270 °C upstream of the SDR, which marks the beginning of the air pollution control devices. The SDR mainly controlled sulfur dioxide (SO_2) by means of calcium hydroxide ($Ca(OH)_2$) injection. A fabric filter was installed to capture particulate matter, and the wet scrubber removed acid gases such as hydrogen chloride (HCl). The injection rate of lime slurry into the SDR was 167 kg/h, while the residence time of the flue gas in the SDR, fabric filter, wet scrubber was 9.5 sec, 4.8 sec, and 5.8 sec, respectively. The pH of the solution in the wet scrubber was maintained at pH 6-7 by adding sodium hydroxide (NaOH). Activated carbon was temporarily injected upstream of the fabric filter to examine its mercury removal efficiency. Fig. 1 also shows the concentrations of carbon monoxide (CO), HCl, SO_2, nitrogen oxides (NOx), and total suspended particle (TSP) in the flue gas upstream of the SDR and

Table 1. Waste sample analysis results

Proximate analysis				Elemental analysis	
Moisture (%)	Volatile matter (%)	Fixed carbon (%)	Ash (%)	S (%)	Cl (%)
2.9	57.1	9.6	28.2	0.10	0.38
Heavy metal analysis (mg/kg)					
Hg	Cd	Cr	Cu	Ni	Pb	Zn
0.276	2.3	603	206	133	46	420

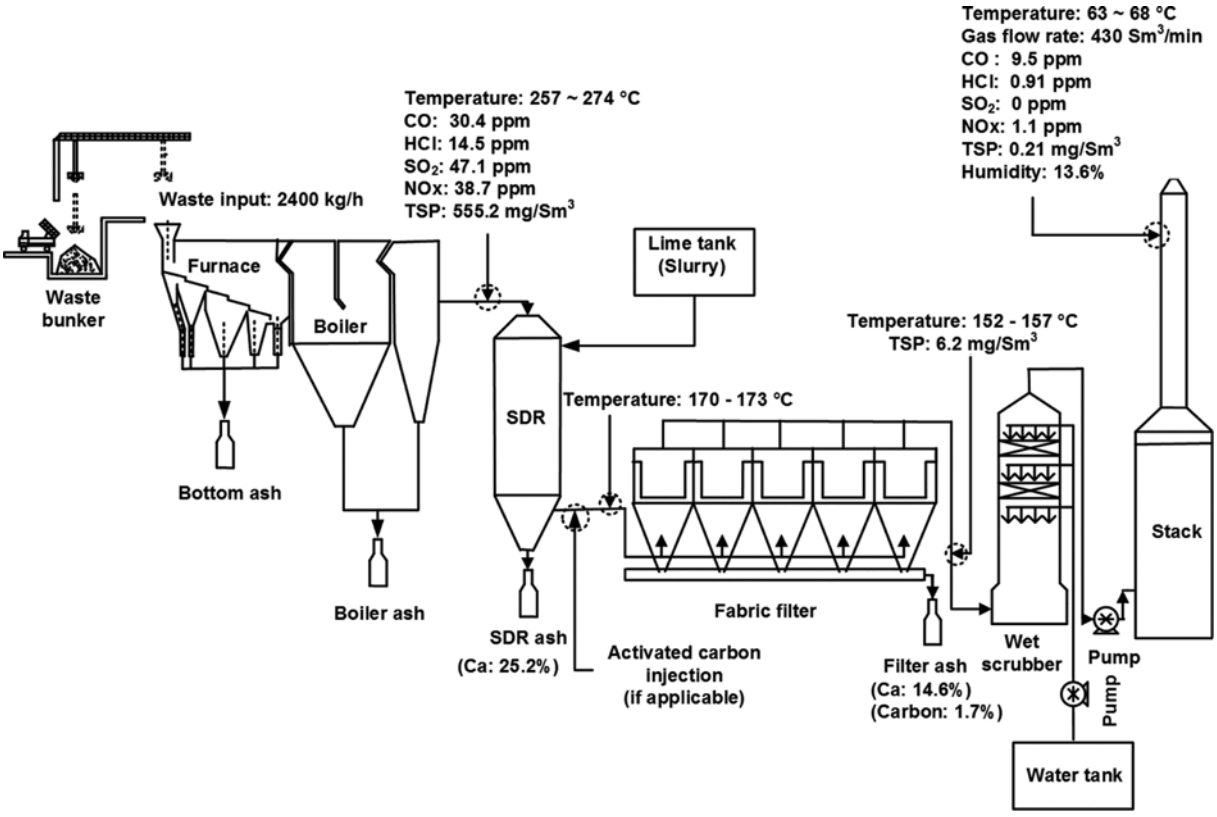

Fig. 1. Schematic diagram of municipal solid waste incinerator.

Table 2. Heavy metal concentrations in flue gas at different sampling points (μg/Sm^3)

	Cd	Cr	Cu	Ni	Pb	Zn	Hg
Upstream of SDR	300	90	2500	40	7600	19800	15.0
Upstream of fabric filter	200	50	1800	20	7500	14500	16.5
Upstream of wet scrubber	ND	2	ND	ND	ND	ND	22.8
Stack	ND	6	ND	ND	ND	ND	6.1
Emission standard	20	300	5000	2000	200	5000	30

ND=Not detected

downstream of the stack, respectively. The HCl concentration was determined using Korea Standard Methods for Examination (ES01305.1). 40 L flue gas upstream of the SDR was drawn using KXC-60 sampler (APEX Instruments, Inc., USA) to determine the HCl concentration. A gas analyzer (GA-21 Plus, Madur, Poland) was used to determine the other gas concentrations. On the other hand, the flue gas concentrations in the stack were measured using the telemonitoring system.

2. Samples Collection and Analysis

Ash was produced at the bottom of the furnace, boiler system, SDR, and fabric filter, and an ash sample was collected and analyzed to determine its heavy metal content. Furthermore, flue gas was sampled to determine its heavy metal concentration. The flue gas was drawn to the heated filter, followed by the impinger train. To conduct mercury speciation, eight impingers were placed in the impinger train. The first three contained a 1 N potassium chloride (KCl) solution, which absorbed oxidized mercury (Hg^{2+}) from the flue gas. The fourth impinger contained a 10% hydrogen peroxide (H_2O_2) and 5% nitric acid (HNO_3) solution, while the fifth, sixth, and seventh impingers contained a 4% w/v potassium permanganate ($KMnO_4$) and 10% v/v sulfuric acid (H_2SO_4) solution for absorbing elemental mercury. Silica gel was applied in the last impinger to absorb moisture, and all impingers were kept in an ice bath during the sampling time. The total time required for a sample was 40 min. All the impingers and connections were rinsed with a 0.1 N HNO_3 solution. The solutions were analyzed by means of a mercury analyzer (RA-3A, Nippon Instrument Corp, Japan) to determine the concentrations of elemental mercury ($[Hg^0]_{flue\ gas}$) and oxidized mercury ($[Hg^{2+}]_{flue\ gas}$) in the flue gas. Using these concentrations, the mercury oxidation percentage was determined according to the following equation.

$$\text{Mercury oxidation}\ (\%) = \frac{[Hg^{2+}]_{Flue\ gas}}{[Hg^0]_{Flue\ gas} + [Hg^{2+}]_{Flue\ gas}} \times 100 \qquad (1)$$

The mercury concentration in the solid waste, including raw materials and ash, was determined by using a mercury analyzer (DMA-80, Milestone, Inc., Shelton, CT, USA). The mercury analyzer uses the principle of thermal decomposition, amalgamation and atomic absorption. All the mercury is released from the sample through thermal decomposition and selectively captured through gold amalgamation. The mercury is then released by heating 950 °C. The total mercury is quantitatively measured by atomic absorption spectrophotometry at 253.65 nm. The particulate matters collected on the filter paper during each sampling process were analyzed to determine their heavy metal contents, following the Korea Standard Methods for Examination (ES01400.2). The filter paper was cut into small pieces and kept in aqua regia (HNO_3 and HCl with 1 : 3). The solution was heated to 200 °C and maintained for 20 min. After removing the impurities through filtration, the solution was diluted and analyzed to determine the contents of Cd, Cr, Cu, Ni, Pb, and Zn, using inductively coupled plasma optical emission spectrometry (ICP-OES, Agilent 700 series, USA).

RESULTS AND DISCUSSION

1. Heavy Metals

1-1. Heavy Metal Concentrations in Flue Gas

The concentrations of Cd, Cr, Cu, Ni, Pb, and Zn are displayed in Table 2. The heavy metal concentrations were determined to be 300 μg Cd/Sm^3, 90 μg Cr/Sm^3, 2,500 μg Cu/Sm^3, 40 μg Ni/Sm^3, 7,600 μg Pb/Sm^3, and 19,800 μg Zn/Sm^3 in the flue gas upstream of the SDR. As the flue gas passes through the SDR, the heavy metal concentration decreases slightly. Almost all of the Cd, Cr, Cu, Ni, Pb, and Zn were collected in the fabric filter because these heavy metals exist as particulate matter in the flue gas. A small amount of Cr was present in the flue gas, with concentrations of 2 μg/Sm^3 and 6 μg/Sm^3 upstream of the wet scrubber and in the stack, respectively.

1-2. Heavy Metal Concentrations in Ash

As discussed, almost all of the heavy metals were captured before arriving at the wet scrubber. Ash produced from the furnace, boiler, SDR, and fabric filter located before the wet scrubber was collected and analyzed to determine the heavy metal content. Table 3 displays the heavy metal concentration in the ash produced from each device. Cd and Pb were present in the ash of the fabric filter (fly ash), with higher concentration than in the furnace and boiler ash (bottom ash). However, Cr and Ni were present in the fabric

Table 3. Heavy metal concentrations in ash produced from furnace, boiler, SDR, and fabric filter, respectively (mg/kg)

	Furnace	Boiler	SDR	Fabric filter
Cd	4	45	16.1	526.5
Cr	148	316.8	67.4	32.6
Cu	2274	743.9	171.8	1964
Ni	47.9	133.8	21.8	14.6
Pb	208.8	2733.3	328.7	4883.3
Zn	5480.3	5725.3	1232.2	11953.3
Hg	0.02	0.02	0.16	0.42

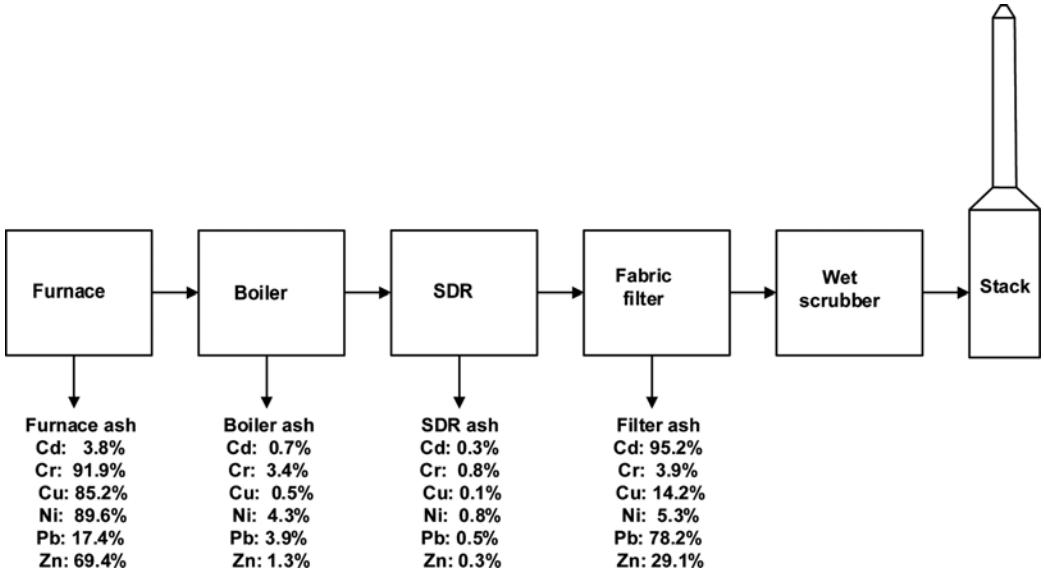

Fig. 2. Fate of heavy metals in incinerator and air pollution control devices.

filter with lower concentration. Cu and Zn were present with similar concentration in the furnace and fabric filter ash samples. These results demonstrate that every heavy metal exhibits a different fate in the incinerator and air pollution control devices.

For the incineration of 2,400 kg of waste, averages of 364, 6.3, 7, and 70 kg of ash were produced in the furnace, boiler, SDR, and fabric filter, respectively. Based on the heavy metal concentration and ash production rate from each device, the fates of the heavy metals in the incinerator and air pollution control devices are presented in Fig. 2. As illustrated in the figure, Cd and Pb are mainly present in fly ash and collected in the fabric filter, while Cr, Cu, and Ni are mostly present in the bottom ash, and Zn is present in the bottom and fly ash at a ratio of 7 : 3. However, the mass balance closures between input and output heavy metal contents were not in a reasonably acceptable range. Further study is necessary on the mass balance of each heavy metal of Cd, Cr, Cu, Ni, Pb, and Zn.

2. Mercury

2-1. Mercury Concentrations in Flue Gas

Flue gas was sampled at different points downstream of the air pollution control devices. The concentration of elemental and oxidized mercury was determined for flue gas upstream of the SDR, fabric filter, wet scrubber, and in the stack, as indicated in Fig. 3. The figure shows that the majority of gaseous mercury is present in an oxidized form in the flue gas upstream of the wet scrubber. The gaseous mercury decreases significantly, from 22.8 to 6.1 μg/m^3, through the wet scrubber. However, the elemental mercury concentration in the stack is higher, while oxidized mercury concentration in the stack is lower than those upstream of the wet scrubber. This indicates a reduction of oxidized mercury to elemental mercury, with a significant amount of oxidized mercury being removed in the wet scrubber. Hg^0 re-emission occurs in a complicated fashion. Many parameters have been found to affect the re-emission, such as pH, temperature, and concentration of sulfites, chloride and bromide ions [29-32]. Among the parameters, increases in the concentration of sulfites, chloride and bromide ions may inhibit a

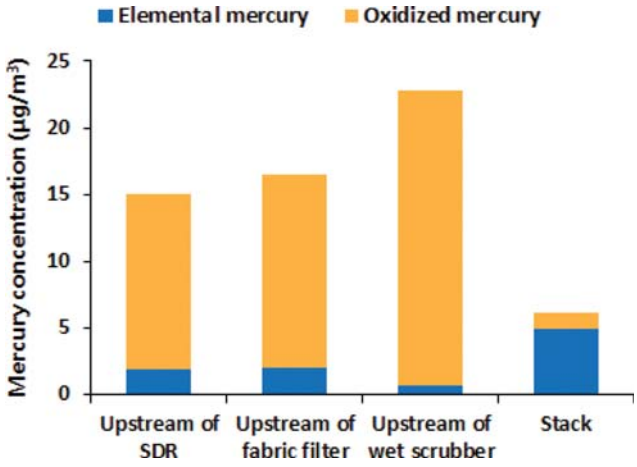

Fig. 3. Elemental and oxidized mercury concentration in flue gas at different sampling points.

reduction of oxidized mercury in the wet scrubber [29,30]. Different results have been reported on the effect of pH on Hg^0 re-emission [30-32]. Therefore, further studies on the parameters are necessary to understand Hg^0 re-emission in the wet scrubber. The higher total mercury concentration upstream of the wet scrubber than the concentrations upstream of the SDR and fabric filter is a result of the errors caused by subsequent sampling. The mercury oxidation percentage was determined by using Eq. (1). Fig. 4. shows that more than 86% of the gaseous mercury is present in an oxidized form in the flue gas upstream of the wet scrubber, while the gaseous mercury emitted to the stack consists of 20% oxidized mercury.

2-2. Mercury Concentrations in Ash

To understand the fate of mercury in the incinerator and air pollution control devices, the ash produced from each air pollution control device was collected and analyzed to determine the mercury concentration. Table 3 also displays the mercury concentration in each ash sample, where the SDR and fabric filter ash

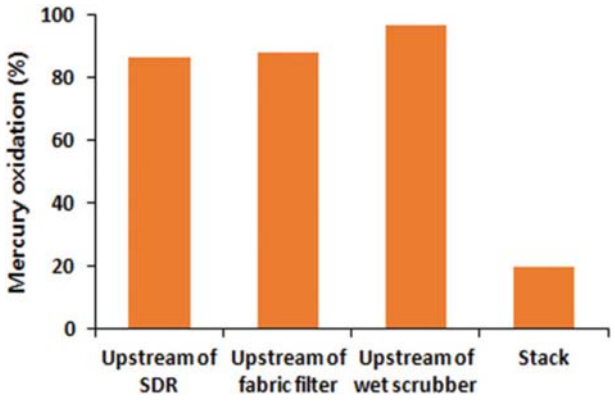

Fig. 4. Mercury oxidation percentage at different sampling points.

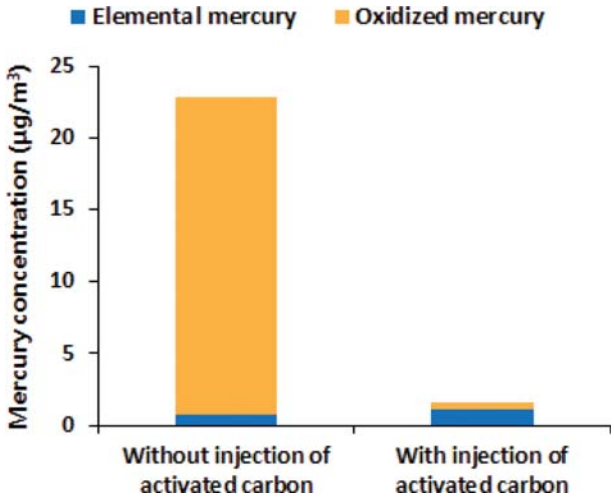

Fig. 6. Elemental and oxidized mercury concentrations in flue gas upstream of wet scrubber, without and with activated carbon injection.

samples exhibit higher mercury concentration than the furnace and boiler ash samples. This indicates that some of the gaseous mercury in the flue gas is removed in the SDR and fabric filter. The fate of mercury in the incinerator and air pollution control devices is presented based on mass balance calculations for mercury concentration of the inlet waste (0.276 mg/kg) in Fig. 5. Although the fabric filter exhibits the highest mercury removal efficiency among the devices, with the exception of the wet scrubber, only 4.45% of the mercury entering the furnace is removed in the fabric filter. The mercury removal efficiency of the wet scrubber was determined to be 74.86% based on the mass balance calculations. This result is consistent with the 73.2% determined from the difference in mercury concentration upstream and downstream of the wet scrubber, as shown in Fig. 3. It is therefore suggested that wet scrubber or mercury control technology may be required for efficient control of mercury emissions from municipal solid waste incinerators.

3. Injection of Activated Carbon for Mercury Control

In this study, we selected injection of commercial activated carbon upstream of the fabric filter to serve as mercury control technology. The commercial activated carbon (HOK-super, RWE, Germany) has a BET surface area of 400 m^2/g, and 85% of its particle size is less than 45 μm. Because the majority of gaseous mercury is present in an oxidized form in municipal waste-fired flue gas, the activated carbon was not chemically promoted. It was injected at a constant rate of 0.2 g carbon/Sm^3 flue gas and captured in the fabric filter with fly ash. To determine the mercury adsorption efficiency of the activated carbon injection, the flue gas was sampled downstream of the fabric filter, which was in turn upstream of the wet scrubber. Fig. 6. illustrates the concentration of elemental and oxidized mercury in the flue gas upstream of the wet scrubber with activated carbon injection, compared to those without. As shown in Fig. 6, the total gaseous mercury concentration decreased significantly from 22.8 to 1.56 $\mu g/m^3$ with injection of activated carbon, which demonstrates 93.2% mercury removal efficiency. The

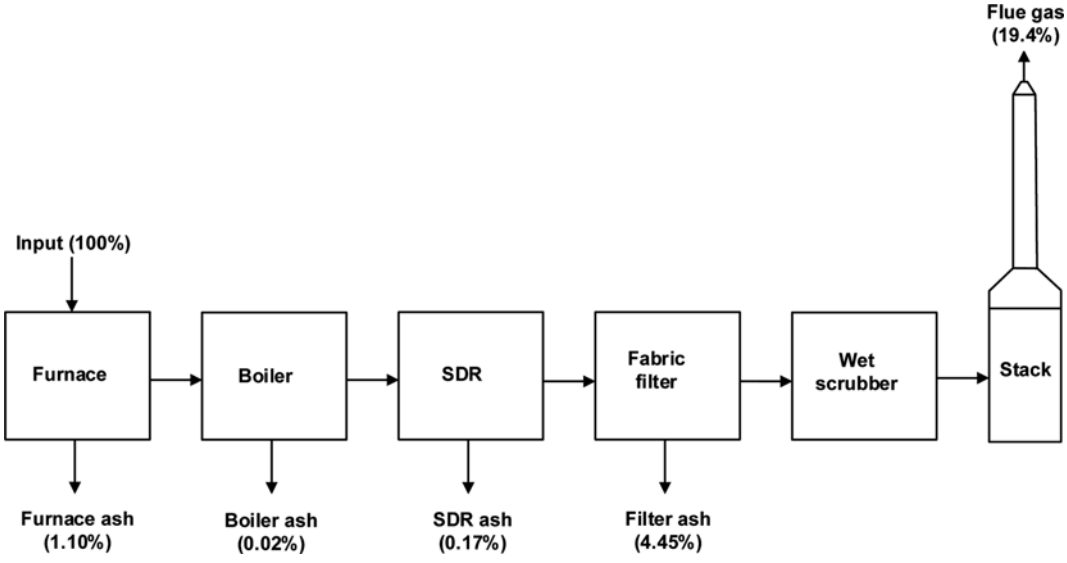

Fig. 5. Fate of mercury in incinerator and air pollution control devices.

fact that elemental mercury concentrations in the flue gas are very similar with and without activated carbon injection indicates that the activated carbon mainly captured oxidized mercury. This also suggests that injection of non-chemically promoted activated carbon upstream of the fabric filter is a highly effective method for mercury control in municipal waste-fired flue gas, which includes most of the gaseous mercury in an oxidized form.

CONCLUSION

This study was conducted to gain an understanding of the fate of heavy metals, such as Cd, Cr, Cu, Ni, Pb, Zn, and Hg, in municipal solid waste incinerators and air pollution control devices. Heavy metal concentrations were determined in the flue gas upstream and downstream of the SDR, fabric filter, and wet scrubber. Almost all of the Cd, Cr, Cu, Ni, Pb, and Zn were captured in the fabric filter, which indicates that its filtering efficiency determines the removal efficiency for these heavy metals. However, most of the Hg was emitted from the fabric filter, and downstream of the fabric filter, the majority of gaseous mercury was present in an oxidized form. The heavy metal concentrations were determined for the ash produced from the furnace, boiler, and SDR located upstream of the fabric filter. Cd and Pb were found to be present mostly in the fly ash, while Cr, Cu, and Ni were mostly present in the bottom ash, and Zn was present in the bottom and fly ash at a ratio of 7 : 3. The mass balance calculation result indicates that 94.26% of inlet Hg was present in the flue gas downstream of the fabric filter, while 19.4% of the inlet Hg left the stack, showing that 74.86% of the inlet Hg was removed from the wet scrubber. The high temperature range of 152-157 °C downstream of the fabric filter may be attributed to the low Hg removal. Because the wet scrubber was found to be efficient for removing oxidized mercury, its use is suggested for the efficient control of mercury emissions from municipal solid waste incinerators. The reduction of oxidized mercury to elemental mercury was also found in the wet scrubber, which requires further investigation. With the injection of commercial activated carbon at a rate of 0.2 g/Sm^3, 93.2% mercury removal efficiency was obtained from the combination of activated carbon and bag filter. The presence of HCl in the flue gas and unburned carbon in the ash of the fabric filter may contribute to efficient mercury removal of the activated carbon. This suggests the use of efficient particulate matter control for emissions of Cd, Cr, Cu, Ni, Pb, and Zn, as well as wet scrubber or mercury-specific control technology for the emission of Hg from municipal solid waste incinerators.

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