

Speciation and mass distribution of mercury in a bituminous coal-fired power plant

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Abstract

Characterization and mass balance of mercury in a coal-fired power plant were carried out in a 500 MW, bituminous coal consuming electric utility boiler. This facility is equipped with a cold-side electrostatic precipitator (ESP) and a wet flue gas desulfurization (FGD) in series as air pollution control devices (APCDs). Mercury sampling points were selected at both the up and down streams of the ESP and outlet of the FGD, which is at stack. Two different types of sampling methods were employed, one is the Ontario Hydro (OH) method (ASTM D6784) and the other is US EPA101A. Various samples were collected from the coal-fired power plant such as fuel coals, fly ash in hopper, lime/lime stone, gypsum, and effluent water from FGD. These samples were analyzed by US EPA 7470A and 7471A to understand the behavior and mass balance of mercury in the process of a coal-fired power plant. There are no significant differences between the two sampling methods, but the OH method seems to have more advantages for Hg sampling from a coal-fired power plant because mercury speciation is quite an important factor to estimate the mercury emission and control efficiency from combustion flue gas. Approximate Hg mass balance could be obtained from various samples in the study; however, a series of long-term and comprehensive study is required to evaluate the reliable Hg mass distribution and behavior in a coal-fired power plant.

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1. Introduction

The major mercury sources from human activity are coal-fired electric utility boilers that have been reported by both the US EPA, and a Position paper from the European Commission (US EPA, 1997, 1999; European Commission, 2001).

Mercury compounds from combustion sources mainly consist of particle-bound mercury (Hg_p), gaseous elemental mercury (Hg^0), and oxidized

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mercury (Hg^{2+}). In the combustion zone, most of the mercury in the fuel coal is evaporated and exists as an elemental form of mercury (Hg^0), which is cooled by passage through the down stream of the flue gas flow and can be oxidized by flue gas components such as HCl , SO_2 , H_2O , and fly ash, finally, formed an oxidized form of mercury (Hg^{2+}) such as HgCl_2 , HgO , etc. (Meij et al., 2002; Lee et al., 2004a; Laudal et al., 2000a, b). These kinds of oxidized mercury can be attached physically or reacts with the particulate in the flue gas and then, forming a Hg_p . Hg^0 is more difficult to control than Hg^{2+} because of its high volatility property and low reactivity with virgin-activated carbons (non-chemically treated). On the other hand, Hg^{2+} is likely to be removed by wet-type scrubber due to its high solubility in water (US EPA, 1997, 1999, 2002a).

Activated carbons, fly ash or dedicated sorbents injection methods at the upstream of particulate control devices were widely carried out in pilot scale or field experiments to demonstrate the mercury removal efficiencies (Durham et al., 2003; Nelson et al., 2003; Pavlish et al., 2003). The results show that specially treated activated carbons and some typical fly ash have affinity for mercury compounds, which can remove mercury or are able to oxidize as an easily removal form of mercury (Hg^{2+}). (US EPA, 1999, 2002a)

However, the total removal efficiency and mercury speciation are quite dependant on various operating conditions such as type of coal, flue gas temperature and components, and air pollution control devices configuration and so on. The change of mercury speciation is important in order to understand the mercury control technologies and the fate and behavior of mercury from coal-fired power plants.

Coal-firing power covers about 30% (1.5 GW) of total electricity generation capacity in Korea (based on 2001 statistics) and most of the coal-fired power plants (about 1.2 GW) are equipped with both cold-side electrostatic precipitator (ESP) and wet flue gas desulfurization (FGD) that are used for particulate and SO_2 removal, respectively. In addition, recently, a selective catalytic reduction (SCR) system has been started to install in order to control nitrogen oxides (NO_x) from coal-fired power plants. Previous studies indicate that NO_x reduction system can affect the mercury speciation that increases the oxidation of Hg^0 , therefore, if coal-fired boilers are equipped with SCR, ESP, and wet FGD the highest mercury removal efficiency could be expected (Richardson et al., 2002; Lee et al., 2004b).

In this study, a typical coal-fired power plant in Korea was selected to allow the identification of the present state of mercury emission and to estimate the fate and behavior of mercury from a coal-fired

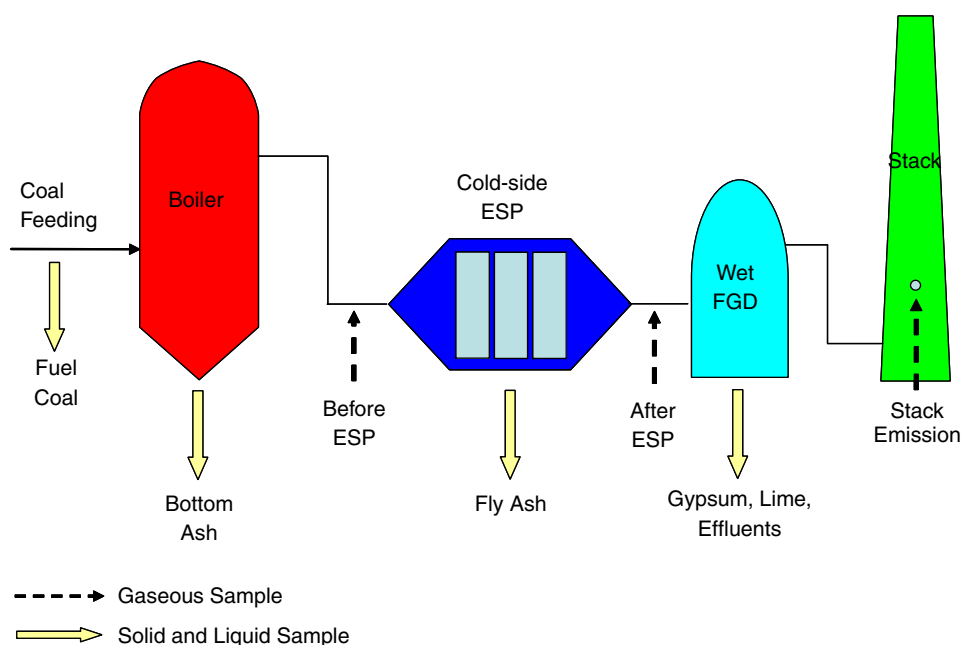


Fig. 1. Schematic diagram of selected coal-fired power plant.

power plant. This study has been carried out for 2 years, twice of tests in each series of experiment every year. In order to estimate the fate and behavior of mercury from the coal-fired power plant, gaseous samples as well as solid and liquid samples such as fuel coal, fly ash from air pollution control device (APCD) hopper, gypsum (by-product from FGD), lime or limestone, and effluents were collected at the coal-fired power plant in each field test year. The gaseous sampling was implemented in accordance with the Ontario Hydro (OH) method (ASTM D6784) and US EPA 101A method. Solid and liquid samples were treated in accordance with US EPA 7470A and 7471A respectively. The assessment of mercury emission and total mercury mass balance from the coal-fired power plant were estimated from the recorded analytical data.

2. Experimental

2.1. Site description and configuration

This coal-fired utility electric boiler is consuming pulverized bituminous coal (imported blend coal), and has 500 MW generating capacity. Also, it has a cold-side ESP for particulate control followed by a wet FGD where lime/limestone slurry is injected to

remove SO_2 into the combustion flue gas; finally, the treated combustion gas is released to atmosphere. This kind of coal-fired utility boiler and APCDs configuration is a typical type of coal-firing power plant as well as about 70% of coal-fired power plant has a plan to install SCR system for NO_x controlling from combustion flue gas in Korea. Fig. 1 shows the schematic diagram of the selected coal-fired utility boiler.

2.2. Sampling approach and mercury analysis

Gaseous Hg sampling was conducted in accordance with the OH method (ASTM D6784) and US EPA 101A on which Korean standard method was based for mercury sampling method from combustion sources in Korea (2002). Both sampling methods were tested simultaneously at three sampling points, both up and down streams of ESP and after FGD that is stack. Mercury speciation was implemented by OH method at each sampling points. Liquid and solid samples were collected at each process location from the plant in each field test year and were treated by US EPA 7470A for effluents from FGD and US EPA 7471A for coal, fly ash, limestone, and gypsum from by-products FGD. Cold vapor atomic absorption- (CVAA) type Lab 254 mercury analyzer (Mercury Instruments

Table 1
Properties of coals sampled in each series of tests at the power plant

	Coal #1	Coal #2	Coal #3	Coal #4	Coal #5
<i>Proximate analysis, as received wt%</i>					
Moisture	5.08	2.17	6.14	10.23	10.22
Volatile	29.74	27.47	27.74	41.01	41.05
Fixed carbon	53.16	55.82	56.76	46.00	45.48
Ash	12.02	14.53	9.37	2.76	3.24
<i>Ultimate analysis, dry base wt%</i>					
C	68.96	69.90	74.73	58.23	54.23
H	4.31	4.29	4.30	4.88	4.52
O	10.67	6.59	11.71	19.36	24.23
N	1.54	1.75	1.26	1.20	1.10
S	0.45	0.67	0.50	3.29	2.45
Cl (ppm)	9.2	11.4	18.2	8.3	17.1
Hg ($\mu\text{g kg}^{-1}$)	42.67	65.87	77.07	78.0	58.0
Heating value (kcal kg^{-1})	6072.2	6259.8	6504.9	6237.4	6149.4
Origin/test year	Indonesia/2003	Indonesia/2003	Indonesia + China/2003	Indonesia/2004	Indonesia/2004

GmbH, Germany) was employed to analyze the total mercury concentration in liquid and solid samples. Its detection limit was $0.1 \mu\text{g L}^{-1}$ and 10 wt% SnCl_2 was used for the reduction reagent. All sampling tests and analysis were carried out in triplicate or more and averaged. The sampling point and location for each sample is indicated in Fig. 1.

3. Results and discussion

3.1. Assessment of mercury behavior across APCDs

Table 1 shows the properties of coal sample such as proximate and ultimate analysis, Hg concentration, and heating value and so on, in this study. Table 2 indicates total mercury emission concentrations across APCDs and facility operation conditions like as flue gas components, temperature, and coal type. Fig. 2 shows Hg concentration transition through APCDs. Also, the differences between OH method and US 101A were not significant and the measured values were shown to be within the error range. Mercury concentration was decreased

through APCDs. At the up stream of ESP, the averaged mercury emission ranged from 2.62 to $2.99 \mu\text{g Nm}^{-3}$ and $1.16\text{--}2.50 \mu\text{g Nm}^{-3}$ at the outlet of ESP. Approximately 34% of mercury was reduced across ESP that caused by removal of fly ash (Hg_p) in the flue gas. The efficiency of ESP was about 97–98%. Generally, fly ash has an affinity for mercury compounds which is likely to adsorb on the surface of fly ash because some types of oxidized mercury such as HgCl_2 have sticky properties (Meij et al., 2002); however, it must be noted that the effectiveness of mercury removal can be affected by types of coal, flue gas composition as well as carbon content in fly ash (US EPA, 1997, 1999, 2002a). Final emission to the atmosphere from stack was about $1.03\text{--}2.41 \mu\text{g Nm}^{-3}$. It can be explained that some oxidized mercury could be absorbed by lime-slurry when mixing in the FGD scrubber tower due to its water-soluble property.

Overall mercury removal efficiency with existing APCDs (cold side ESP and wet FGD) was achieved at a rate of about 33–40% that agrees with previous US EPA results that ranged from 29% to 74% according to coal type (US EPA, 1999, 2002a, b).

Table 2
Mercury concentrations across APCDs with operating conditions

Sampling points	Sampling method	Total mercury concentration ($\mu\text{g Nm}^{-3}$)		Average ($\mu\text{g Nm}^{-3}$) and standard deviation		Operating condition		
		1st year (2003)	2nd year (2004)	Avg.	SD	Flue gas temp. ($^{\circ}\text{C}$)	Flue gas components	Coal type
Before ESP	Ontario-1	2.88	2.99			123–146	O_2 (%) 2.6–3.3	Imported bit. coal
	Ontario-2	1.77	2.89	2.63	0.58		CO_2 (%) 15.3–16.1	
	101A-1	2.89	2.82				NO_x (ppm) 190.5	
	101A-2	2.82	2.62	2.79	0.12		SO_x (ppm) 154.3	
After ESP (before FGD)	Ontario-1	2.25	1.26			136–140	O_2 (%) 3.01–3.5	Imported bit. coal
	Ontario-2	2.02	1.56	1.77	0.45		CO_2 (%) 15.3–15.6	
	101A-1	2.50	1.16				NO_x (ppm) 156.0	
	101A-2	2.44	1.26	1.84	0.73		SO_x (ppm) 218.0	
Stack (after FGD)	Ontario-1	2.41	1.21			92–96	O_2 (%) 4.1–4.7	Imported bit. coal
	Ontario-2	2.13	1.03	1.70	0.68		CO_2 (%) 14.2–14.7	
	101A-1	2.26	1.12				NO_x (ppm) 165.0	
	101A-2	2.20	1.07	1.66	0.66		SO_x (ppm) 56.0	
Stack (ICR) ^a	Ontario	—	—	2.57	1.80	—	—	Bit. coal-fired plant

^aUS EPA, Information Collection Request, 1999.

3.2. Mercury speciation

Mercury speciation results are shown in Table 3 and Fig. 3, which were calculated from the results of OH method. With those results, a clear transition of Hg speciation along with APCDs could be obtained.

Higher portions of Hg_p were measured at the inlet of ESP, which was about 24–59%, on the other hand, at the outlet of the ESP the portion of Hg_p was only 0.9–3.8% in the tested conditions at the plant. Large amounts of Hg_p were removed by the use of the ESP. When passing through ESP a little portion of Hg^{2+} increased, this effect could be

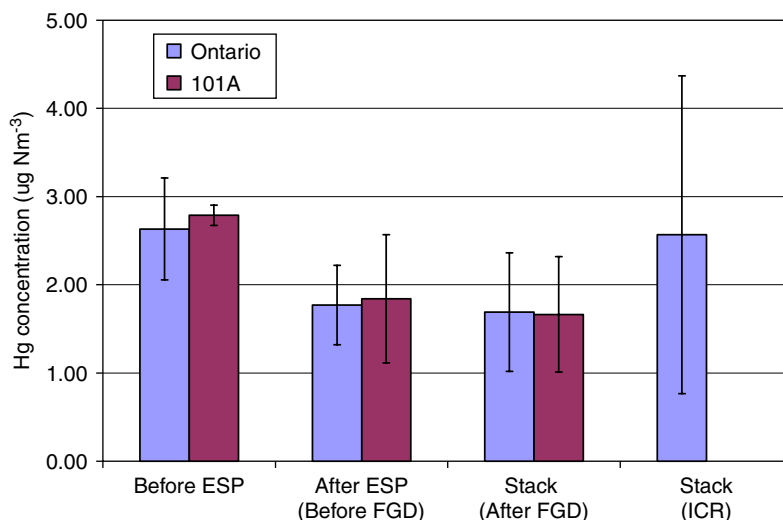


Fig. 2. Hg concentration across APCDs and comparison of two sampling methods—between OH and US EPA 101A method (ICR; Information Collection Request, US EPA, 1999).

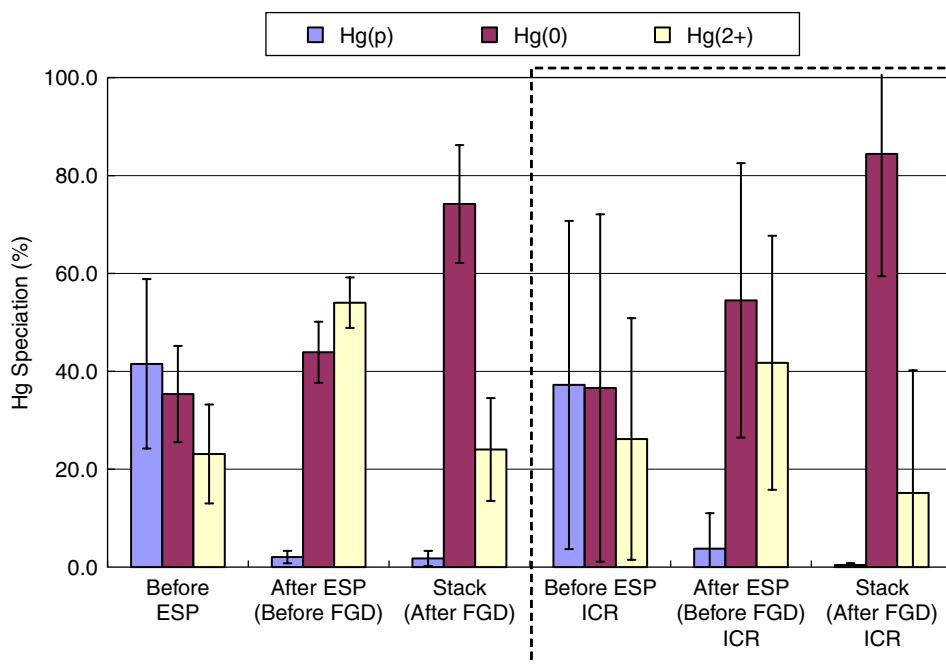


Fig. 3. Variation of Hg speciation across APCDs (in dotted line, ICR; Information Collection Request, US EPA, 1999).

explained by the fact that Hg compounds could be oxidized through the ESP because of increasing the contact time in the flue gas and fly ash or/and acidic gas components are able to enhance the mercury oxidation rate (Meij et al., 2002; Lee et al., 2004a; Laudal et al., 2000a, b; Senior et al., 2000), however, to clarify the mechanism of Hg oxidation in the flue gas more comprehensive and continuous studies are required.

After wet FGD (stack) a large portion of Hg^0 was measured about 74.2% and 24.0% of Hg^{2+} was emitted to the atmosphere, but the portion of Hg^0 at the upstream of FGD was 44.9% only. The change of Hg speciation through FGD was due to the absorption of Hg^{2+} in the scrubber solution because Hg^{2+} has soluble properties in water. In addition, many researchers have studied the role of wet FGD for mercury removal and change of Hg speciation

and the results indicate that some oxidized mercury could be absorbed in wet slurry as well as being reduced to Hg^0 by through chemical reaction with sulfite ion (SO_3^{2-}) in the scrubber solution and then, into the wet FGD outlet flow (Constantinou et al., 1995). Therefore, the total mercury concentration and portion of Hg^0 in the flue gas were occasionally higher than those of the inlet (Richardson et al., 2003; Chang and Ghorishi, 2003). Fig. 3 shows the results of Hg speciation changes in range from this study and ICR data (US EPA, 1999). The results inside of dotted line were selected and averaged from ICR data, which is the same APCD's configuration (cold-ESP and wet FGD equipped facilities) with this study. It is difficult to compare directly with this study and previous ICR results, but the transition of Hg speciation across APCDs is relatively agreed with together.

Table 3

Results of the transition of mercury speciation across APCDs

Year	Test no.	Mercury speciation	Before ESP		After ESP (before FGD)		Stack (after FGD)		
			µg N m ⁻³	(%)	µg N m ⁻³	(%)	µg N m ⁻³	(%)	
2003	1	Hg _p	0.84	29.2	0.02	0.9	0.01	0.4	
		Hg ₀	1.38	47.9	1.08	48.0	2.03	84.2	
		Hg ²⁺	0.66	22.9	1.15	51.1	0.37	15.4	
		Total Hg	2.88	100.0	2.25	100.0	2.41	100.0	
	2	Hg _p	0.43	24.3	0.04	2.0	0.01	0.5	
		Hg ₀	0.68	38.4	0.83	41.1	1.81	84.9	
		Hg ²⁺	0.66	37.3	1.15	56.9	0.31	14.6	
		Total Hg	1.77	100.0	2.02	100.0	2.13	100.0	
2004	1	Hg _p	1.61	53.8	0.02	1.6	0.04	3.3	
		Hg ₀	0.84	28.1	0.63	50.0	0.78	64.5	
		Hg ²⁺	0.54	18.1	0.61	48.4	0.39	32.2	
		Total Hg	2.99	100.0	1.26	100.0	1.21	100.0	
	2	Hg _p	1.70	58.8	0.06	3.8	0.03	2.9	
		Hg ₀	0.78	27.0	0.57	36.6	0.65	63.1	
		Hg ²⁺	0.41	14.2	0.93	59.6	0.35	34.0	
		Total Hg	2.89	100.0	1.56	100.0	1.03	100.0	
Hg speciation percentage (%) and standard deviation (SD)				(%)	SD	(%)	SD	(%)	SD
Averaged value (this study)		Hg _p		41.5	17.3	2.1	1.3	1.8	1.5
		Hg ₀		35.4	9.8	43.9	6.2	74.2	12.0
		Hg ²⁺		23.1	10.1	54.0	5.2	24.0	10.5
				100.0	—	100.0	—	100.0	—
ICR study results ^a		Hg _p		37.2	33.5	3.8	7.3	0.4	0.4
		Hg ₀		36.6	35.5	54.5	28.0	84.4	25.1
		Hg ²⁺		26.2	24.7	41.7	25.9	15.2	25.1
				100.0	—	100.0	—	100.0	—

^aData selected and averaged from ICR data, which is the same APCDs configuration (cold-ESP and wet FGD equipped facilities) with this study.

For getting better statistical Hg behavior data from coal-fired power plant in Korea, continuous and comprehensive field test have been planed and carrying out now.

3.3. Mercury concentration in solid/liquid sample

Table 4 shows the Hg concentrations of solid and liquid sample collected in the field and the treatment method. Hg concentration in fuel coal ranged from 42.67 to 78.00 $\mu\text{g kg}^{-1}$. The concentration of Hg in fly ash from the ESP hopper ranged from 243.75 to 379.25 $\mu\text{g kg}^{-1}$, on the other hand, bottom ash contained low Hg concentration of about 47.25 $\mu\text{g kg}^{-1}$. In addition, Hg in gypsum (by-product from the wet FGD) ranged from 131.25 to 304.25 $\mu\text{g kg}^{-1}$. Previous study (US EPA, 2002b) of Hg in coal combustion residues (CCRs) is also indicated in Table 4 to allow comparison with this study. All of the collected data have been used for the calculation of mercury mass balance in the coal-fired power plant.

3.4. Hg mass balance in a coal-fired power plant

Hg mass balance was estimated based on the results of Hg analysis in above session and Table 5 shows results of two series of test in different periods with details of calculation. The operation parameters such as coal/lime feeding rate, fly ash (in hopper) and gypsum generation rate were provided from power plant operational records. The averaged production rate of fly ash and gypsum were estimated by the total amounts of fly ash and gypsum from coal-fired power plant complex, because total six coal-fired boilers were operating and all of them were managed by combined operating control systems. Other minor output materials such as effluents and sludge from wet FGD as well as bottom ash were considered when calculating the Hg mass balance in this study.

Above 80% of in/out Hg mass balance was obtained despite the small number of data points (two times, duplicated tests each time). The recovery (in/out mass balance) rates showed 82% and 99%,

Table 4
Hg concentrations measured solid and liquid samples from the coal-fired power process

Sample	Hg concentration ($\mu\text{g kg}^{-1}$)	Collection year and treatment method	Reference data ^a (ppm)
Coal #1	65.87	2003	0.095
Coal #2	42.67	2003	
Coal #3	77.07	2003	
Coal #4	78.00	2004	
Coal #5	58.00	2004	
Avg.	64.3 \pm 14.7	US EPA 7471A	
Fly Ash #1	243.75	2003	0.46 ^b
Fly Ash #2	375.00	2003	
Fly Ash #3	379.25	2004	
Fly Ash #4	283.00	2004	
Avg.	320.3 \pm 67.0	US EPA 7471A	
Bottom ash	47.25	2004 US EPA 7471A	0.058
Gypsum #1	131.25	2003	0.32
Gypsum #2	304.25	2003	
Gypsum #3	155.50	2004	
Avg.	131.25 \pm 93.7	US EPA 7471A	
Lime/lime stone	18.75	2004 US EPA 7471A	—
Effluent	0.01 ($\mu\text{g L}^{-1}$)	2004 US EPA 7470A	—

^aUS EPA, characterization and management of residues from coal-fired power plants, 2002.

^bFly ash in cold-side ESP, bituminous coal firing plant.

Table 5

Hg mass balance in a Korean coal-fired power plant and the comparison with previous studies

Hg source		Hg mass flow rate and balance—this study				Reference mass balance studies			
		Averaged value (2003 and 2004)		Mass balance		Dutch type ^a	MRY-A ^b	MRY-B ^b	CC ^b
		g h ⁻¹	%	2003 %	2004 %				
Hg IN	Fuel coal	11.52 ^c	99.7	99.6	99.7	99.5	20.76 (99.8)	20.76 (99.7)	24.05 (99.7)
	Lime/limestone	0.03 ^d	0.3	0.3	0.3	0.2	NA	<0.03	<0.03
	Process water	—	—	—	—	0.2	<0.03	<0.03	<0.03
	Sub total	11.55	—	—	—	—	—	—	—
Hg OUT	Fly ash (in ESP hopper)	6.80 ^e	58.9	59.1	57.6	49.0	NA	0.90 (4.3)	<0.03 (0.1)
	Flue gas (in stack)	2.80 ^f	24.2	33.7	15.2	25.0	17.65 ^g (84.9)	16.86 ^g (81.0)	21.49 ^g (89.1)
	Fly ash (in flue gas)	0.05 ^h	0.43	0.62	0.28	0.07	—	—	—
	Gypsum	0.75 ⁱ	6.5	4.5	7.2	17.0	1.83 ^j (8.8)	0.74 ^j (3.6)	2.1 ^j (8.7)
	Bottom ash	0.10 ^k	0.87	0.91	0.83	0.8	—	—	—
	Sludge	0.07 ^l	0.61	0.5	0.7	9.0	—	—	—
	Effluent	—	—	—	—	0.04	—	—	—
	Out/in balance (%)	10.57	(91.5)	(99.3)	(81.8)	(100.9)	(93.7)	(88.9)	(96.9)

^aMeij et al. (2002)—bituminous coal.^bLaudal et al. (2000b)—lignite coal.^cAvg. coal feed rate 4300 ton day⁻¹.^dCalculated based on gypsum generation rate and including process water.^eAvg. Hg in fly ash 0.32 mg kg⁻¹.^fEstimated flue flow rate 1,680,000 N m³ h⁻¹.^gHg exiting with stack.^hAvg. fly ash content is 1.8% of total emission in stack.ⁱEstimated gypsum generation 91.7 ton day⁻¹.^jHg in scrubber blowdown.^kEstimation base on coal ash contents.^lIncluding sludge and effluent from wet FGD.

respectively, and other previous investigations available from literatures (Meij et al., 2002; Laudal et al., 2000b) showed in relatively similar ranges of recovery. Fig. 4 is the summarized diagram with mass distribution of mercury from the first attempted investigation for a Korean bituminous coal-fired power plant. About 57–59% of Hg is collected in the ESP fly ash hopper and 15–34% of Hg is released to atmosphere, 5–7% of Hg remains in the gypsum. When comparing to a previous study (Meij et al., 2002) at typical Dutch coal-fired power plants, which has a similar APCDs configuration with Korean plants, the results of this study agree with them showing same partitioning trend. However, another available data (Laudal et al., 2000b) from two North Dakota lignite coal-fired plants in

US showed that major portion of Hg distributed in flue gas at stack due to different fuel coal and limited APCDs configuration, mainly consisted of a scrubber. To get better representative results, more data collection from coal-fired power plants are now going on and comprehensive analyses have to be conducted in future.

4. Conclusions

Studies on emission and behavior of Hg from a coal-fired power plant equipped with cold side ESP and wet FGD have been carried out and obtained the following results.

There were no significant differences between the Ontario hydro (OH) method and the US EPA

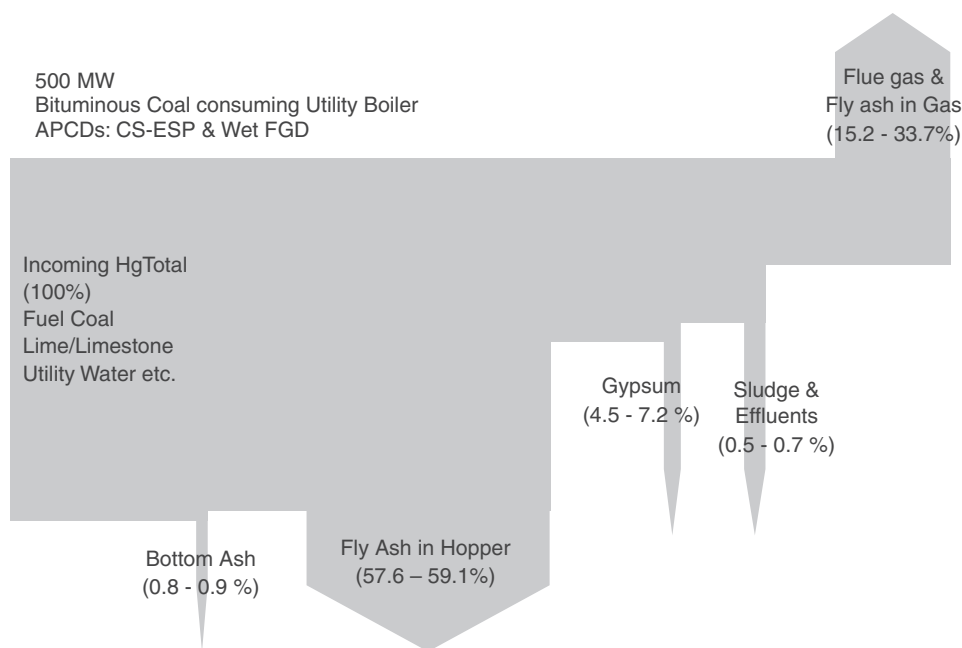


Fig. 4. Mass distribution of mercury in range at a bituminous coal-fired plant.

101A; however, to understand the transformation of Hg in the coal combustion flue gas, Hg speciation possible method has a lots of advantages. Approximately 93% of in/out Hg balance was obtained with this study and major Hg outgoing was fly ash in hopper and flue gas in stack. Large amount of Hg_p , above 50% of Hg could be removed by cold-side ESP and Hg compounds seemed to be oxidized to Hg^{2+} through ESP, which resulted in increase of Hg^{2+} . Then wet FGD unit could remove Hg^{2+} with lime-slurry scrubber solution, which covers less than 10% of Hg, and about a quarter of mercury emits to the atmosphere through stack. Such results are similar to the available data for Dutch bituminous coal-fired power plants. Hg speciation changes with increasing oxidized portion through ESP and reduction of absorbed Hg to Hg^0 in the wet FGD scrubber solution are reported from other investigations, however, need more comprehensive study with field experiments.

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