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Mercury emissions and partitioning from Indian coal-fired power plants

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ARTICLE INFO

Article history:
Received 11 April 2020
Revised 29 May 2020
Accepted 29 June 2020
Available online 22 July 2020

Keywords:
Coal combustion
Emission
Mercury
Flue gas
Thermal power plant

ABSTRACT

In India coal combustion is the single largest source of emission of mercury which is a widespread persistent global toxicant, travelling across international borders through air and water. As a party to the Minamata convention, India aims to monitor and reduce Hg emissions and stricter norms are introduced for mercury emissions from power plants (30 μg/Nm^3 for flue gas in stack).

This paper presents the results obtained during the experimental studies performed on mercury emissions at four coal-fired and one lignite-fired power plants in India. The mercury concentration in the feed coal varied between 0.12–0.27 mg/Kg. In the mercury mass balance, significant proportion of feed coal mercury has been found to be associated with fly ash, whereas bottom ash contained very low mercury. 80%–90% of mercury was released to air through stack gas. However, for circulating fluidised bed boiler burning lignite, about 64.8% of feed mercury was found to get captured in the fly ash and only 32.4% was released to air. The mercury emission factor was found to lie in the range of 4.7–15.7 mg/GJ.

Introduction

Mercury has become one of the major global air pollutants arising due to toxicity, bioaccumulation, persistence and long range transport. In atmosphere, Mercury is present in different forms and can travel long distances leading to global contamination (Selin, 2009). In aquatic ecosystems, mercury is converted to potent neurotoxin methylmercury which poses highest risks to human health (Karagas et al., 2012; Mckelvey and Oken, 2012). Due to its immense impact on human health and concern about its global transport, Minamata Convention on Mercury was adopted in October 2013. The Convention aims to regulate mercury and its compounds, with obligations for mining, use, emissions, releases, and disposal (Giang et al., 2015).

As per UNEP Global Mercury Assessment 2018, stationary combustion of coal is the second largest source of global anthropogenic emission of mercury behind Artisanal small scale gold mining (ASGM) (UNEP, 2018). It is estimated that depending on the execution of Best Available Technology (BAT), global mercury emission in 2050 may vary anywhere from −4% to +96% (Streets et al., 2009).

India is heavily dependant on coal to meet the energy demand of the country. Coal combustion is the single largest source of mercury emission in India, and coal fired power plants are major contributor in this sector. Mercury contents in Indian coal, is typically in the range of 0.003–0.34 g/tonne with average concentration of 0.14 g/tonne (UNEP, 2014). However due to a large volume of coal burnt, a significant amount of mercury is released in atmosphere. In India, about 637 MT of raw coal and 37 MT were combusted for generation of electricity in 2018–19 (Energy Statistics 2020). As rural electrification is priority of government of India, it plans to increase total power generation of the country and coal based thermal power plants will be an integral part of this plan (CEA report, 2012).

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https://doi.org/10.1016/j.jes.2020.06.035

Coal when combusted in boilers at high temperature, majority of mercury in coal releases as elemental mercury (Hg^0) in the exhaust gas (Moretti and Jones, 2012). Depending on the chemical composition of flue gas i.e. presence of HCl, SO_2, fly ash etc. a fraction of Hg^0 is oxidised (Hg^{2+}) by some thermochemical process. $Hg^{2+}$is more soluble and also has a tendency to get adsorbed on the fly ash particles which leads to formation of particle bound mercury (Hg^p) (Park et al., 2008). Concentration of mercury emitted through stack largely depend on the mercury contents of coals being burnt as well as installed air pollution control devices like electrostatic precipitator (ESP), wet flue gas desulphurisation (WFGD), fabric filter (FF) etc. (Cao et al., 2008), Whereas, elemental mercury is difficult to remove by air pollution control devices. In Indian scenario, most of the power plants have particulate control devices like ESP and FF. However, in recent times installation of Flue gas desulphurisation (FGD) in power plants has gained pace with MoEF&CC notification for compulsory installation of FGD system in the existing and upcoming thermal power plants to curb SOx and presently many FGD projects are in various stages of implementation (Energy Statistics 2020).

Mercury speciation in flue gas is an important factor while assessing the environmental impact of Hg present in the atmosphere, as Hg^0, Hg^{2+} and Hg^p have different physiochemical properties and atmospheric lifetime. However, due to variability in the nature of coal and its composition, combustion conditions and use of oxidising additives during combustion, the proportion and quantities of different mercury species present in flue gases vary significantly.

In recent years, lot of studies have been carried out by experts to understand the partitioning of mercury and its removal efficiency by the commercial air pollution control devices in power plants. Shah et al. showed the extent of reduction in mercury emission from five coal power stations in Australia with ESP or FF as particle control technology (Shah et al., 2010). Bilirgen found that 34.5% reduction in mercury emission at stack can be achieved with optimal boiler control and low-NOx system +WFGD (Bilirgen and Romero, 2012). Lei studied the effect of chlorine and ash composition in mercury transformation across six coal based power plants in China (Lei et al., 2007). Zhang et al. investigated the partitioning, removal efficiency in Chinese power plants with different combinations of APCD like cold ESP, FF, flue gas desulfurization (FGD) (Zhang et al., 2008). Zhao studied mercury transformation in an ultralow emission power plant in China (S. Zhao et al., 2017a). Yokoyama et al. reported detail study of mercury emission from Japanese 700 MW power plant having SCR, ESP and WFGD as air pollution control device (Yokohama et al. 2000).

As the nature of coal used in Indian coal fired power generation differs from other countries, it is important to have full scale studies to be carried out to understand the mercury emission pattern. Till now, actual field studies carried out in Indian power plant in context of mercury emission are scanty because of the lack of reliable sampling and analysis data in Indian context (Reddy et al., 2005). Mercury mass balance studies at coal-fired power plants can help to improve our understanding on the impacts of coal quality and APCD configurations on mercury emissions. Estimated average mercury emission factors for Indian power plants with respect to coal sources, combustion technologies and configuration of APCD may be utilised for determining total mercury emissions from the sector and to reduce mercury emission through process optimization.

In this article, comprehensive mercury mass flow has been studied at five selected thermal power plant boiler units of India. amongst them, four are pulverised coal fired and one is circular fluidised bed system burning lignite. In addition to the total mass balance of mercury in the system, mercury speciation was also estimated. The partitioning of mercury in different combustion products was determined and mercury emission factors have been estimated.

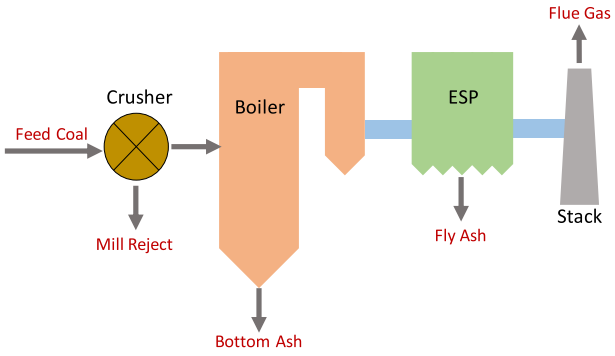

Fig. 1 – Sample collection point from boiler unit.

1. Experimental method

1.1. *Sampling site*

Onsite sampling and tests were carried out in different boiler units of five coal-fired power plants in India, amongst them, four boiler units are pulverised coal fired of 500–660 MW capacity, whereas the other one is circulating fluidised bed (CFB) boiler of 135 MW capacity. All the boiler units have Electrostatic Precipitator (ESP) as only air pollution control device for removing particles from flue gases. The study was carried out in the year 2019. In India majority of the boilers are pulverised coal fired. A couple of lignite fired power plants are based on circulating fluidised bed boiler system. Details of boiler units studied are provided in the Table 1. During the test period, the boiler units were operating under steady full load boiler operation conditions.

1.2. *Sample collection method*

Samples are collected by adopting standard sampling protocol. All the input and output materials were collected. Inputs included feed coal, whereas outputs are mill reject (MR), bottom ash (BA), fly Ash (FA) and flue gas. Flue gas samples were collected from stacks of boiler units at a good elevation where streamline gas flow could be obtained. Sample collection points are shown in Fig. 1. The onsite mercury determination in stack gases were performed by adopting EPA method 30B for total mercury and speciation of flue gas mercury (USEPA Method 30B). At the start of each run, average gas velocities and flue gas temperatures were measured using pre-calibrated pitot tube and temperature probes respectively. Requisite volume stack gas samples have been collected from the flue gas stream by inserting gas lined stainless steel probe through the sampling port of the stack. Gas volume was measured in the dry gas meter and vapour phase mercury in the gas was adsorbed in two different sorbent traps through the sampling port of the stack. Gas volume was measured in the dry gas meter and vapour phase mercury in the gas was adsorbed in two different sorbent traps. For total Hg concentration, activated carbon trap was used whereas for Hg speciation activated carbon along with KCl traps were utilised. The temperature of the probe was maintained at 125 °C to avoid condensation of mercury vapour before they

Table 1 – Description of boiler.

Boiler unit	Capacity (MW)	Boiler type	Air pollution control device	Coal type	Location
Boiler A	500 MW	Sub-Critical PC-Boiler	Electrostatic Precipitator	Sub-Bituminous	West Bengal
Boiler B	500 MW	Sub-Critical PC-Boiler	Electrostatic Precipitator	Sub-Bituminous	West Bengal
Boiler C	660 MW	Super-Critical PC-Boiler	Electrostatic Precipitator	Sub-Bituminous	Madhya Pradesh
Boiler D	660 MW	Super-Critical PC-Boiler	Electrostatic Precipitator	Sub-Bituminous	Madhya Pradesh
Boiler E	135 MW	CFB boiler	Electrostatic Precipitator	Lignite	Rajasthan

PC-Boiler: Pulverised coal; CFB: Circulating Fluidised Bed.

Table 2 – Feed coal properties.

Unit	Ash%	Moisture %	Volatile Matter%	Gross calorific Value Kcal/Kg	C%	H%	N%	S%	Chlorine (mg/Kg)	Mercury mg/Kg
A	37.8	6.1	15.9	4201	45.48	3.92	1.57	0.36	1300	0.172 ± 0.004
B	42.2	3.3	22.3	4175	45.19	1.25	3.37	0.35	500	0.128 ± 0.005
C	39.7	9.6	21.9	4057	39.33	3.77	2.03	0.40	500	0.270 ± 0.010
D	35.2	9.8	21.7	3629	43.90	4.02	1.21	0.41	1300	0.243 ± 0.005
E	18.9	36.1	38.3	2937	32.88	0.57	6.96	0.18	500	0.163 ± 0.006

Table 3 – Partitioning of mercury in solid samples.

Mercury contents of solid samples (mg/Kg)				
Unit	Feed coal	Fly ash	Bottom ash	Mill reject
A	0.172 ± 0.004	0.106 ± 0.008	0.014 ± 0.003	0.261 ± 0.017
B	0.128 ± 0.005	0.064 ± 0.001	0.008 ± 0.004	0.640 ± 0.006
C	0.270 ± 0.010	0.065 ± 0.001	0.034 ± 0.002	0.031 ± 0.001
D	0.243 ± 0.005	0.085 ± 0.003	0.032 ± 0.001	3.535 ± 0.056
E	0.163 0.006	0.492 ± 0.011	0.012 ± 0.002	–

are captured in the sorbent traps. The average value of mercury concentration of triplicate runs was considered for calculation. The following solid samples were collected simultaneously during each run of stack gas sampling -crushed coal from mill feeder, mill reject from bawl mills, bottom ash from boiler ash discharge point, fly ash from ESP hoppers.

1.3. *Analysis method*

The solid samples were properly crushed, pulverised, thoroughly homogenised and preserved in polythene bottles. Mercury concentrations of all the samples had been determined with the instruments Tri-cell DMA-80 (EPA Method 7473, Milestone, Italy) or RA915M with PYRO 915+analyser (Lumex, Russia). Determination of mercury combines the techniques of thermal decomposition, catalytic conversion, amalgamation, and atomic absorption spectrophotometry (ASTM D6722–01).

2. Results and discussion

2.1. *Feed coal properties*

The proximate analysis, Chlorine, Sulphur and mercury contents of collected coal samples were analysed and shown in the Table 2.

Coal samples: The analysis shows that the ash contents of coal samples vary between 35.2%–42.2%; moisture contents lie in the range of 6.0%–9.8%. The heat values of the coal samples are moderate to low and the gross calorific values of feed coal samples is in the range of 3600–4200 Kcal/Kg.

Lignite samples: The characteristics of the lignite samples are presented under Unit E in Table 2. Due to low rank, the air dried moisture is found to be high. The sample is having moderate ash content and heat values.

The total sulphur contents of the studied coal and lignite samples are in the range of 0.18–0.41%. Total chlorine content varied between 500 and 1300 mg/Kg. The mercury content of feed coal and lignite samples varies from 0.163–0.270 mg/kg. The values are comparable with average mercury concentration of Indian coals (UNEP 2014).

2.2. *Partitioning of mercury in combustion products*

During combustion, traces of mercury present in coal or lignite completely volatilises at high temperature as elemental mercury, due to its low boiling point. Under boiler ambient conditions, mercury vapours undergo thermo-chemical transformations. A part of the mercury may get oxidized under the influence of excess oxygen, halides and sulphur oxides present in flue gases. Catalytic role of particulates in oxidizing mercury in the vapour phase have also been reported. In the colder region of boiler, mercury may re-condense and get adsorbed on fine ash particles and consequently captured by ESP. Due to milling and combustion at high temperature, feed coal mer-

Table 4 – Mercury concentrations and speciation in flue gas.

Unit	Flue gas Hg (μg/Nm^3)	Oxidised Hg%	Elemental Hg%
A	18.5 ± 1.4	49	51
B	9.8 ± 0.35	45	55
C	29.4 ± 0.5	38	62
D	20.8 ± 1.8	42	58
E	9.2 ± 0.37	51	49

Table 5 – Mercury emission factors.

Unit	G (ton/hr)	Q (LHV) (kJ/kg)	V (Nm^3/hr)	C (μg/Nm^3)	MEF1 (mg/GJ)	MEF2 (mg Hg/ton of coal)
A	218	16,544.86	1,617,962	18.5	8.2	137.7
B	191	17,094.49	2,114,951	9.8	6.3	108.9
C	209	15,890.18	1,775,503	29.4	15.7	249.7
D	197	14,041.03	2,051,975	20.8	15.4	217.2
E	68.5	11,265.02	405,859	9.2	4.8	54.4

cury is partitioned in different products like fly ash, bottom ash, mill rejects, and flue gas.

Mercury content in feed coal varies largely was found in the range of 0.163 mg/Kg to 0.270 mg/Kg. Mercury concentrations of solid residues of power plants are presented in Table 3. Mercury contents of fly ash for pulverised coal fired boilers (A-D) found to be in the range of 0.064–0.106 mg/Kg. In the lignite fired circulating fluidised bed boiler (E), significant enrichment of mercury content in fly ash has been noticed. In all the boilers, mercury contents of bottom ash samples have been found to be much lower as compared to fly ash mercury values.

2.3. *Relative enrichment factor (REF)*

The relative enrichment factors (REF) were calculated to understand the partitioning of mercury after coal combustion process. The REF relates the mercury concentrations in the fly ash and the bottom ash to the concentration of mercury in the feed coal (Meij et al., 2002). Relative enrichment factor for mercury in fly ash and bottom ash were calculated as Eq. (1).

$$REF = \frac{C_i * C_{ash}}{C_{coal}} \quad (1)$$

where, REF is relative enrichment factor; C_i is the Hg concentration in fly ash or bottom ash, C_{ash} is the ash yield of coal, C_{coal} is the Hg concentration in coal.

The REF for Hg in fly ash and bottom ash are low (<0.23) for pulverised coal fired boilers which indicates volatile nature of mercury (Bhanagare et al., 2011). However, for circulating fluidised bed boiler, REF for fly ash is quite high (REF = 0.57) which indicates higher enrichment of Hg in fly ash in CFB boiler. The high mercury enrichment in fly ash for circulating fluidised bed boiler may be attributed to higher retention time of ash particles inside the boiler, lower boiler temperature and high unburnt carbon in the fly ash (Lei et al., 2007; Zhang et al., 2016; Zheng et al., 2017). The Loss on ignition of fly ash of circulating fluidised bed boiler was found to be 7.93%, whereas for pulverised coal fired boilers, the values range from 0.37–0.75%.

2.4. *Mercury in flue gas and speciation*

Mercury being very volatile, it vaporises during combustion. Some mercury condenses on the fly ash and gets separated by ESP, and the rest emitted to the air. Table 4 shows the flue gas mercury concentrations and per cent elemental and oxidized mercury species present in flue gases. The total mercury concentration per unit volume in the flue gas samples studied have been found to lie in the range of 9–29 μg/Nm^3 (Table 4). The values are within the threshold limit of mercury emission norm of 30 μg/Nm^3 for Indian thermal power plants. It is observed that with increasing mercury contents of fuels, flue gas mercury concentration has also increased. However, other factors like boiler design, operational parameters, temperature profile inside the boiler and flue gas line, dust load in the flue gas stream, ESP efficiency etc. influence the flue gas mercury concentration. Low mercury emission level has been observed in circulating fluidised bed boiler flue gas (Table 4).

The speciation of elemental and oxidised mercury was measured with help of special sorbent traps having KCl and activated carbon as adsorbents. It is observed that oxidised mercury varies from 38% to 51% of total mercury emitted. Oxidised mercury concentration highly depends on interaction of elemental mercury in flue gas with different species like Cl_2, HCl, O_2, NOx, etc. present in flue gas (Wang et al., 2009). Oxidised mercury being more water soluble and reactive compared to elemental mercury can be easily removed by wet gas desulphurisation, whereas elemental mercury is inert in nature and is difficult to remove by air pollution control devices.

2.5. *Mercury mass balance*

The mercury mass balance has been worked out using the results of above analysis for the selected five boiler units. It indicates the distribution of the mercury within the system which allows us to understand the major mercury release pathways for better mercury management. For material balance exercise, material flow and other necessary data like coal feed rates, production of fly ash, bottom ash, mill rejects, plant load factor, ambient temperature and pressure, flue gas composition, stack dimension etc. have been collected from plant op-

Mass Balance of boiler units

Unit	Hg Output/Hg Input (%)	Flue gas %	Fly ash %	Bottom Ash %	Mill Reject %
A	100.2	79.8	19.6	0.80	0.03
B	103.2	84.6	17.9	0.4	0.08
C	102.1	92.5	8.5	1.12	0.002
D	101.7	89.4	11.0	1.0	0.29
E	97.8	32.4	64.8	0.5	--

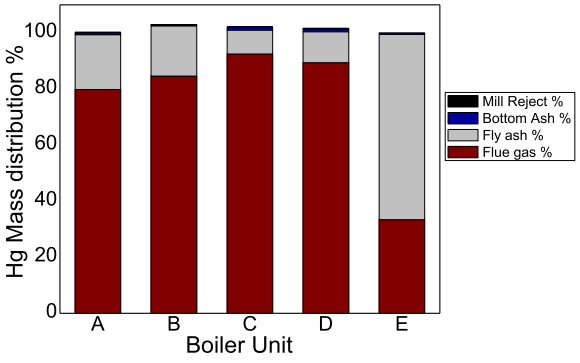

Fig. 2 – Distribution of mercury in combustion products.

erators. To perform mercury mass balance calculation, it is assumed feed coal mercury is the only mercury input source and the feed mercury is partitioned in different outputs like fly ash, bottom ash, mill rejects and flue gas. It is also assumed that for pulverised coal fired boilers 80% of total ash is fly ash, 20% is bottom ash, whereas for circulating fluidised bed boiler 75% of total ash generated is fly ash and 25% is bed ash. In case of CFBC boiler, lime is added for SO_2 control. Lime contributes small mercury input into the system. Mercury content of lime is found to be 0.096 ± 0.003 mg/Kg and the value has been taken into account for overall mercury mass balance.

The partitioning of mercury in the products is presented in Fig. 2. The Table shows that major portion of feed mercury is emitted through stack. For the pulverised coal fired boilers (A-D) about 80–90% of total mercury, released to the atmosphere and rest portion remained associated with fly ash. Mercury concentration in fly ash was higher than that in bottom ash. Bottom ash and mill reject contained only a very small fraction of feed coal mercury. The proportion of feed coal mercury distributed in the combustion products and mill rejects is depicted graphically in Fig. 2.

In case of Circulating Fluidised Bed boiler, mercury distribution pattern in the products have been found to be quite different. It is observed that major portion of mercury is associated with fly ash (64.8%). As previously discussed, in CFBC boiler due to high retention time of the ash particle in the boiler, high unburnt carbon and low boiler temperature, majority of the mercury gets associated with the fly ash and captured by the ESP. Only a small portion are emitted through flue gas (32.4%). Bed ash contained an insignificant percentage of input mercury (0.5%).

2.6. *Mercury emission factor (MEF)*

Mercury emission factor (MEF1), a relationship between mercury emission into the atmosphere and heat value of the consumed coal in thermal power stations, has been derived for all the studied plants using following equations.

$$\text{MEF1} = \frac{m}{G \times Q}$$

$$m = V \text{ x } C$$

where, MEF1 is mercury emission from flue gas per unit of Lower heating value (LHV) of coal, mg/GJ; m (mg/hr) is emission of mercury from flue gas; G (ton/hr) is coal consumption; Q (kJ/kg) is Lower heating value (LHV) of feed coal; V (Nm^3/hr) is flow rate of flue gas; C (ug/Nm^3) is concentration of mercury in flue gas.

The estimated MEF for five power plants are shown in Table 5. The MEF1 varied in the range of 4.8–15.7 mg/GJ. MEF1 values indicate that for boiler units' B and E, MEF1 values are much lower than the other units. This is corroborated by the findings that flue gas mercury concentrations are also lower (Table 4) as compared to other three boiler units. The MEF values for nine power plants in US varied in the 0.82–9.46 mg/GJ whereas for Chinese power plants it varied between 0.68–4.70 mg/GJ. The derived MEF1 for the plants under the present study are in the higher side compared to the US and Chinese coal power plants (US Department of energy, 1996; Gao et al., 2014; Wang et al., 2017; S. Zhao et al., 2017b; Wu et al., 2010). Similarly, Mercury emission factor (MEF2) in terms of the amount of mercury released in the atmosphere per tonne of coal combusted were also estimated. The estimated MEF2 values as shown in Table 5, ranges between 54 and 249 mg Hg/tonne of coal. The estimated MEF2 are slightly higher than reported for Chinese power plant (Wang et al. 2010). The MEFs depend on various factors like mercury content in the feed coal, coal rank, efficiencies of air pollution control devices installed. Higher MEFs obtained for these plants may be attributed to the fact that the plants have only ESP as pollution control device; whereas coal fired power plants in China and US have additional air pollution control devices like FF, FGD, and SCR etc.

3. Conclusions

Comprehensive mercury mass balance studies have been performed for five selected Indian coal and lignite fired power plants. Mercury contents of feed coals, mercury emissions concentrations, partitioning of mercury in various combustion products, mercury speciation in flue gas and mercury emission factors have been derived. Relative enrichment factor for fly ash and bottom ash were also estimated. The mercury contents of feed coal and lignite samples varied within 0.163–0.270 mg/Kg. The REF values show that there is significant enrichment of mercury in fly ash, particularly for circular fluidized bed boiler. The mercury concentration of flue gases varied in the range of 9–29 μg/Nm^3. It may be noted that mercury concentrations of flue gas are always below the threshold limit of 30 μg/Nm^3 prescribed for Indian coal based power plants. The speciation results indicate that 50–60% emitted mercury are in elemental form. The mercury emission factor for the tested plants varied in the range of 4.8–15.7 mg/GJ. The

mercury emission from Indian thermal power plants could be significantly reduced by modification and optimization of process parameters of ESP; installation of other air pollution control devices like FF, FGD, SCR and use of oxidizing additives during combustion.

Acknowledgments

H. A. and T. B. D acknowledge UNDP India for financial support. Authors thankfully acknowledge Director, CSIR-Central Institute of Mining and Fuel Research, Dhanbad for his constant encouragement and support.

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