

Chapter 2

The Status of Mercury Emission from Coal Combustion Power Station

Abstract Mercury is considered a dangerous heavy metal to both humans and the ecosystem because it is highly toxic to the central nervous system and it tends to bioaccumulate in the human body. Coal-fired power plants are one of the main sources of mercury emission to the environment. During combustion, the mercury in the coal is transformed into three species: particle-bound mercury, vapor-phase elemental mercury, and vapor-phase oxidized mercury. Particle-bound Hg_p is easily removed by dust control equipment such as baghouse filters and electrostatic precipitators (ESPs). Vapor-phase oxidized mercury is water soluble which makes its removal in wet flue-gas desulfurization units (FGD) possible. Vapor-phase elemental mercury is extremely volatile and insoluble. Therefore, the conversion of mercury from one form to another is important for selecting the appropriate mercury removal technology.

Coal-fired power plants have been considered to be the primary anthropogenic source of mercury into the atmosphere. For example, these account for about one-third of all anthropogenic mercury emissions in the USA. In China, mercury emission from nonferrous metals smelting, coal combustion, and miscellaneous activities contributed about 45 %, 38 %, and 17 %, respectively. Mercury contamination is widespread in different ecological compartments such as atmosphere, soil, and water. Mercury is a global pollutant. The research on mercury in America and Europe has been widely conducted. Anthropogenic emissions of mercury still increase in Asia because of increased burning of coal and increased industrialization.

Keywords Mercury emission • Coal-fired power plants • Status • Global

Mercury is considered a dangerous heavy metal to both humans and the ecosystem because it is highly toxic to the central nervous system and it tends to bioaccumulate in the human body. Coal-fired power plants are one of the main sources of mercury emission to the environment. As a consequence, legislative bodies both in Europe and the USA are considering the reduction of mercury emissions from coal-fired power plants an important priority. According to a ruling announced in the USA, mercury emissions from utility boilers must be reduced to a final cap of 15 ton/year by 2018, equivalent to nearly 70 % reduction.

2.1 Flue Gas Mercury Emission and Its Speciation

During combustion, the mercury in the coal is transformed into three species: (1) particle-bound mercury (Hg_p), (2) vapor-phase elemental mercury (Hg^0), and (3) vapor-phase oxidized mercury (Hg^{2+}), primarily in the form of HgCl_2 . For the optimal removal of mercury from flue gas, a high level of oxidation is beneficial since, unlike Hg^0 , HgCl_2 is water soluble which makes its removal in wet flue-gas desulfurization units (FGD) possible. Particle-bound Hg_p is easily removed by dust control equipment such as baghouse filters and electrostatic precipitators (ESPs). Therefore, the conversion of mercury from one form to another is important for selecting the appropriate mercury removal technology.

Hg^0 may be oxidized to Hg^{2+} via homogeneous (gas–gas) or heterogeneous (gas–solid) reactions. Mercury in coal begins to volatilize at temperatures below 200 °C almost regardless of the mode of occurrence of mercury in the coal. At temperatures above 600–700 °C, Hg^0 is the only stable form. At temperatures <400 °C and in the presence of chlorine, part of the Hg^0 vapor is oxidized to $\text{HgCl}_2(\text{g})$ by direct reaction of atomic chlorine Cl with elemental mercury. According to the equilibrium reactions model proposed by Frandsen et al., $\text{HgSO}_4(\text{s})$ and $\text{HgO}(\text{s})$ also become thermodynamically stable species in conventional coal combustion systems at low temperatures (110–320 °C). A theoretical assessment of the equilibrium composition of mercury-containing species over the 100–1,600 °C range in a combustion atmosphere without chlorine indicated that $\text{Hg}(\text{g})$ is the most abundant species in gas phase with the presence of small amount of $\text{HgO}(\text{g})$, the exact proportions of these two species varying with temperature.

The oxidation of mercury depends on the composition of the flue gas and especially on the quantity of HCl, NO_x, and SO₂ present [1]. Furthermore, an increase of mercury oxidation has been observed in systems equipped with selective catalytic reduction (SCR) units for NO_x control.

The Hg emission factor, E_f (into fly ash), can be calculated in the following equations:

$$E_f = \frac{Q_{\text{ash}}}{Q_{\text{total}}} \quad (2.1)$$

$$Q_{\text{ash}} = C_{\text{ash}} M_{\text{coal}} A_f F_f \eta \quad (2.2)$$

$$Q_{\text{total}} = C_{\text{ash}} M_{\text{coal}} \quad (2.3)$$

where E_f is the emission factor of Hg into ash (%), Q_{ash} is the amount of Hg into ash in the combustion process (mg), Q_{total} is the total Hg in coal (mg), C_{ash} is the Hg concentration of ash (mg/kg), C_{coal} is the Hg concentration of coal (mg/kg), M_{coal} is the mass of coal (kg), A_f is ash content of coal (%), F_f is the ratio of the amount of ash into flue gas to the sum of ash, and η is the efficiency of dust remover (%). Substituting Eq. 2.2 and 2.3 into Eq. 2.1 gives

$$E_f = \frac{C_{\text{ash}} A_r F_f \eta}{C_{\text{coal}}} \quad (2.4)$$

According to the conservation of mass, the relationship within the emission factors is as follows:

$$E_f + E_b + E_a = 1 \quad (2.5)$$

where 1 refers to the total emission amount of the element, E_f is the ratio of the element in fly ash, E_b refers to the ratio of Hg in bottom ash, and E_a is the ratio of element that goes to the atmosphere. All the parameters in Eq. 2.4 were measured directly, E_b was measured using a directly similar method, and E_a is calculated by Eq. 2.5.

The Hg emission from coal combustion of a department or a province, including Hg into atmosphere and into ashes, is equal to the product of the consumption of coal, the Hg content of coal, and the emission factor.

A similar method was employed to calculate E_f in ordinary industrial and domestic layer-burning boilers. As for layer-burning boilers, 64 % of total Hg was sent into the atmosphere with 17.7 and 18.3 % sent into bottom ash and fly ash [2], respectively. As for a pulverized coal-boiler, 74.4 % of total Hg was sent into the atmosphere and 25.6 % sent into cinder.

Coal naturally contains mercury and other chemical elements, which are different from the type of coal and place of the origin; typically, mercury content in coal ranges from 0.01 to 0.48 mg/kg. Mercury emission from such combustion systems occurs when mercury vaporizes during combustion and is exhausted through the combustor stack. There are numerous sources of mercury in wastes. These include electric switches and lighting components, paint residues, and thermometers. The same USEPA report described that around 87 % of mercury is emitted from such combustion activities [3] and recently the USA has put a lot of efforts to manage the emission of mercury with research on any proper analytical methods, speciation in a thermal process, and investigation of better control technologies.

Mercury in coal-fired flue gas often presents as elemental mercury (Hg^0), oxidized mercury vapor (Hg^{2+}), and particulate-bound mercury (Hg_p). The distribution of various forms is dependent on coal rank (mainly on chlorine concentration in coal) and the unburned carbon on fly ash. Hg^0 is highly volatile and insoluble in water; once it has entered the atmosphere, it can remain aloft for nearly several months and as a result is likely to cause global mercury pollution through atmospheric transportations [4].

Hg^{2+} is much less volatile and more highly water soluble than Hg^0 ; HgCl_2 , for example, has vapor pressure of 8.9×10^{-3} Pa and water solubility of 66 g l^{-1} at 20°C , while Hg^0 has vapor pressure of 0.180 Pa and water solubility of $49.4 \mu\text{g l}^{-1}$ at 20°C [5]. This physicochemical property of HgCl_2 makes it possible to employ wet flue-gas desulfurization to control mercury emission. Hg_p can be also removed, along with fly ash particles, using conventional air pollution control devices such as an electrostatic precipitator (ESP) or fabric filter. In contrast to Hg^{2+} and Hg_p , Hg^0

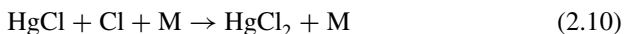
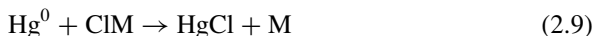
is difficult to capture because it is volatile and insoluble in water. Hence, there are some trials to convert Hg^0 into Hg^{2+} species such as HgO , HgSO_4 , and HgCl_2 , in order to increase the removal efficiency of mercury [6–9].

The chlorination of mercury is the most dominant mechanism during and after coal combustion. The chlorine species HCl and Cl_2 are known to be the most powerful reactants and the major mercury species formed is HgCl_2 . However, Galbreath and Zygarlicke [10] have reported that the addition of HCl at temperatures below 180°C does not increase the formation of HgCl_2 in coal combustion flue gas. Brown et al. [11] have also encountered the similar tendency in their laboratory experiment. These studies indicate that there is little reactivity of HCl toward Hg^0 at lower temperatures. Consequently, it can be expected that Hg^0 oxidation would be reinforced if it works even at low temperature.

In the DBD process, the presence of HCl in the gas mixture drives the decomposition of HCl into H and Cl via direct electron impact dissociation of HCl , the collision-induced dissociation of HCl by N_2 , and so on, which are presented in reactions (2.6), (2.7), and (2.8)



It is considered that Cl atoms and Cl_2 molecules, generated by the DBD process, are responsible for the oxidation of Hg^0 through the following reaction channels:



Although there is little effect of H_2O alone on the oxidation of Hg^0 , the oxidation efficiency of Hg^0 is significantly promoted when H_2O and HCl are present simultaneously. This may be due to the additional formation of oxidative species, Cl and HOCl , through reactions of OH and H with HCl and Cl_2 . The increase in

the reaction temperature causes a reduction in the oxidation efficiency of Hg^0 due to the deterioration of the heterogeneous chemical oxidation of Hg^0 into HgCl_2 . The presence of NO inhibits the oxidation of Hg^0 because O and O_3 created by the DBD are rapidly scavenged, before they can react with Hg^0 .

The United States Environmental Protection Agency (US EPA) announced the Clean Air Mercury Rule (CAMR)³ on Hg emission control from coal-fired power generation on March 15, 2005, which requires the reduction of Hg emissions from coal-fired utility boilers of nearly 70 % from 1999 levels by 2018. The Clean Air Interstate Rule (CAIR) calls for intensive investigation of mercury emission control by the combined utilization of flue-gas desulfurization (FGD) and selective catalytic reduction (SCR), which were originally equipped for control of SO_2 and NO_x , respectively.

2.2 The Status of Mercury Emission in the USA

For the past decade, mercury emission has attracted a great concern due to its high toxicity, persistence, bioaccumulation as methyl mercury in the environment, and neurological health impact [12]. Coal-fired power plants have been considered to be the primary anthropogenic source of mercury into the atmosphere. For example, these account for about one-third of all anthropogenic mercury emissions in the USA [13]. In consequence, there have been initiatives to regulate the emission of mercury at a lower level and to develop the mercury emission control technologies.

The interaction of atmospheric Hg with the biogeosphere leads to pools of Hg in vegetation and soil. Simulated percentage change in total deposition of mercury over North America [2] is depicted in Fig. 2.1. When Hg is deposited to the earth, some is taken up (plant specifically) by vegetation; some is collected through fall; most Hg is collected as the litter fall of senesced leaves and needles on the soil surfaces, where the carbonaceous materials decompose and the mercury becomes complexed with reduced sulfur compounds in the organic soil. The rate of deposition is spatially variable. In the USA, the highest wet deposition of Hg occurs in the Southeast, as indicated from the results of the Mercury Deposition Network [14].

A serious deactivation was observed when SCR catalysts were used when burning North Dakota (ND) lignite-fired boilers. The detailed study indicated that the activity loss of SCR catalysts might be attributed to the pores of the SCR catalyst being plugged by alkali oxides (Na and K) with a lower melting point [15, 16]. A high concentration of Na and K particles attaches to the surface of the SCR catalyst, filling its pores. Thus, Se for Texas lignite and Na and K for ND lignite may decrease the surface area of the fly ash. Together with lower LOI content in fly ash from lignite, a higher occurrence of Se or Na and K in lignite may be some of the major reasons for the lower mercury capture capability.

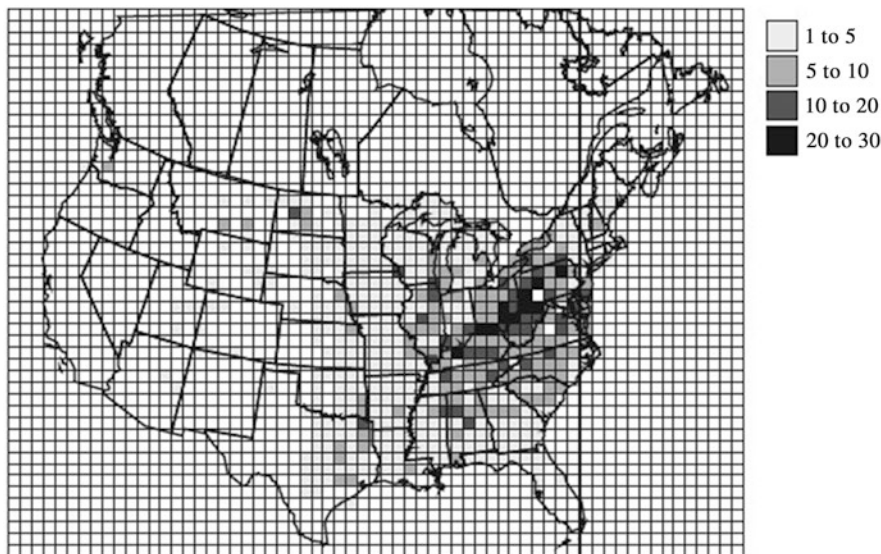


Fig. 2.1 Simulated percentage change in total deposition of mercury from the base case (Reprinted from Ref. [14], Copyright 2004, with permission from Elsevier)

2.3 The Status of Mercury Emission in China

In China, mercury emission from nonferrous metals smelting (especially zinc smelting), coal combustion, and miscellaneous activities (of which battery and fluorescent lamp production and cement production are the largest) contributed about 45 %, 38 %, and 17 %, respectively, to the total Hg emission based on the data of 1999. Mercury contamination is widespread in different ecological compartments such as atmosphere, soil, and water.

It has been estimated that the amount of total Hg emissions was 536 (± 236) t from China in 1999. Mercury emission from nonferrous metals smelting, coal combustion, and miscellaneous activities, of which battery and fluorescent lamp production and cement production are the largest, contributed about 45 %, 38 %, and 17 %, respectively, to the total Hg emission.

China produces and consumes the largest quantity of coal in the world (about 1.25 and 1 billion tons, respectively, in 2000) [17, 18]. Using fuel consumption data and detailed Hg emission factors, the total amount of Hg emission to air from coal combustion was estimated to be 219.5 t in 2000 (based on the average Hg content of 0.20 mg/kg in coal), substantially higher than that in the USA (21.2 t). Using the data of 1999 (and based on the average Hg content of 0.19 mg/kg in coal), a similar amount of 203.7 t Hg emission to air from coal was obtained [19].

The largest three source sectors for Hg emission from coal combustion are industry, power plants, and residential use in China, contributing to 46, 35, and 14 %



Fig. 2.2 Mercury emission from coal combustion of different provinces in China (Reprinted from Ref. [20], Copyright 2007, with permission from Elsevier)

of the total Hg emissions from coal combustion, respectively, and the percentage of elemental (Hg^0), gaseous divalent (Hg^{2+}), and particulate Hg (Hg_p) to the total Hg emissions from coal combustion is 16 %, 61 %, and 23 %, respectively. Figure 2.2 shows the total amount of Hg emissions from coal combustion in different major provinces in China.

China is the largest producer and consumer of coal in the world. As of 2003, China consumed nearly 1,531 Mt of coal, 28 % of the world's total consumption. China's annual coal consumption is also expected to double to 3,037 Mt by 2020. Therefore, China plays an important role in global anthropogenic Hg emissions. The large amount of Hg emitted from China may become a threat to the global environment [20].

Asia, especially China, has been regarded as the world's largest atmospheric Hg emission source. Pacyna estimated that in 1995, Asian countries contributed 56 % of the worldwide THg emissions, compared with 30 % in 1990 [21]. The increase in emissions in Asia was clearly related to the growth of coal combustion in China.

Recently, Streets et al. estimated that 202 t of Hg was emitted from coal combustion in 1999, including 33.1 t of Hg^0 , 124 t of Hg^{2+} , and 45.4 t of Hg_p . Of the Hg emitted from coal combustion in China, 51 % comes from industrial

Table 2.1 Global anthropogenic mercury emission inventory for 2000 (unit: ton)

Continent	Stationary combustion	Cement production	Nonferrous metal production	Pig iron and steel production	Caustic soda production	Mercury production	Gold production	Waste disposal	Other	Total
Africa	205.2	5.3	7.9	0.4	0.3	0.1	177.8		1.4	398.4
Asia (excl. Russia)	878.7	89.9	87.6	11.6	30.7	0.1	47.2	32.6	0.9	1,179.3
Australasia	112.6	0.8	4.4	0.3	0.7		7.7	0.1		126.6
Europe (excl. Russia)	88.8	26.5	10.0	10.6	12.4			11.5	15.3	175.1
Russia	26.5	3.7	6.9	2.7	8.0		3.1	3.5	18.2	72.6
South America	31.0	6.5	25.4	1.4	5.0	22.8				92.1
North America	79.6	7.7	6.4	4.3	8.0	0.1	12.2	18.7	8.8	145.8
Total	1,422.4	140.4	148.6	31.3	65.1	23.1	248.0	66.4	44.6	2,189.9

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use, 33.6 % from power plants, 9.8 % from residential use, and 5.6 % from other uses. Hg emissions from coal combustion in 1999 are shown by province in Fig. 2.1. Hg emissions in central-eastern and southwestern China were obviously most significant. The global background concentration is of 1.5–2.0 ng/m³.

It has been estimated that mercury emissions from coal combustion in China increased from 202 ton in 1995 to 257 ton in 2003, an average annual increase of 3.0 % [22].

Very few studies have been conducted to determine Hg emission factors from different sources in China. As a result, emission factors are typically adopted from studies conducted in Europe and North America with roughly similar sources. Because the processes and pollution control techniques used in China may differ dramatically from those used in developed countries, these adopted Hg emission factors could differ significantly from the actual field conditions in China. Thus, a large uncertainty could exist in China's anthropogenic Hg emissions inventories.

2.4 The Status of Global Mercury Emission

Mercury is a global pollutant. The research on mercury in America and Europe has been widely conducted. Anthropogenic emissions of mercury still increase in Asia because of increased burning of coal and increased industrialization according to Hylander. Seigneur et al. indicated that the natural and marine mercury emissions have the largest average mercury emissions; as for anthropogenic emission, average mercury emissions in Asia are higher than that in other continents. In general, Asia has the largest average emission inventory compared to the other continents in the world. The average mercury inventory in Asia is 1,179.3 tons which occurred about 53.8 % out of the total mercury emission inventory around the world [23] (Table 2.1).

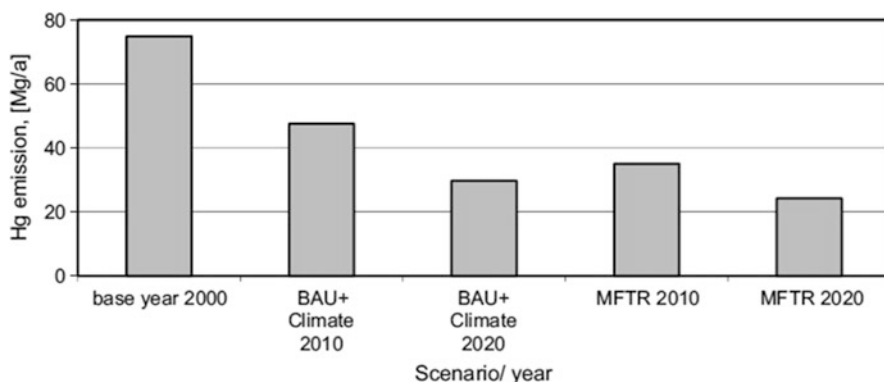


Fig. 2.3 Mercury emission scenarios for power plants in Europe until the 2020 (tons) (Reprinted from Ref. [24], Copyright 2009, with permission from Elsevier)

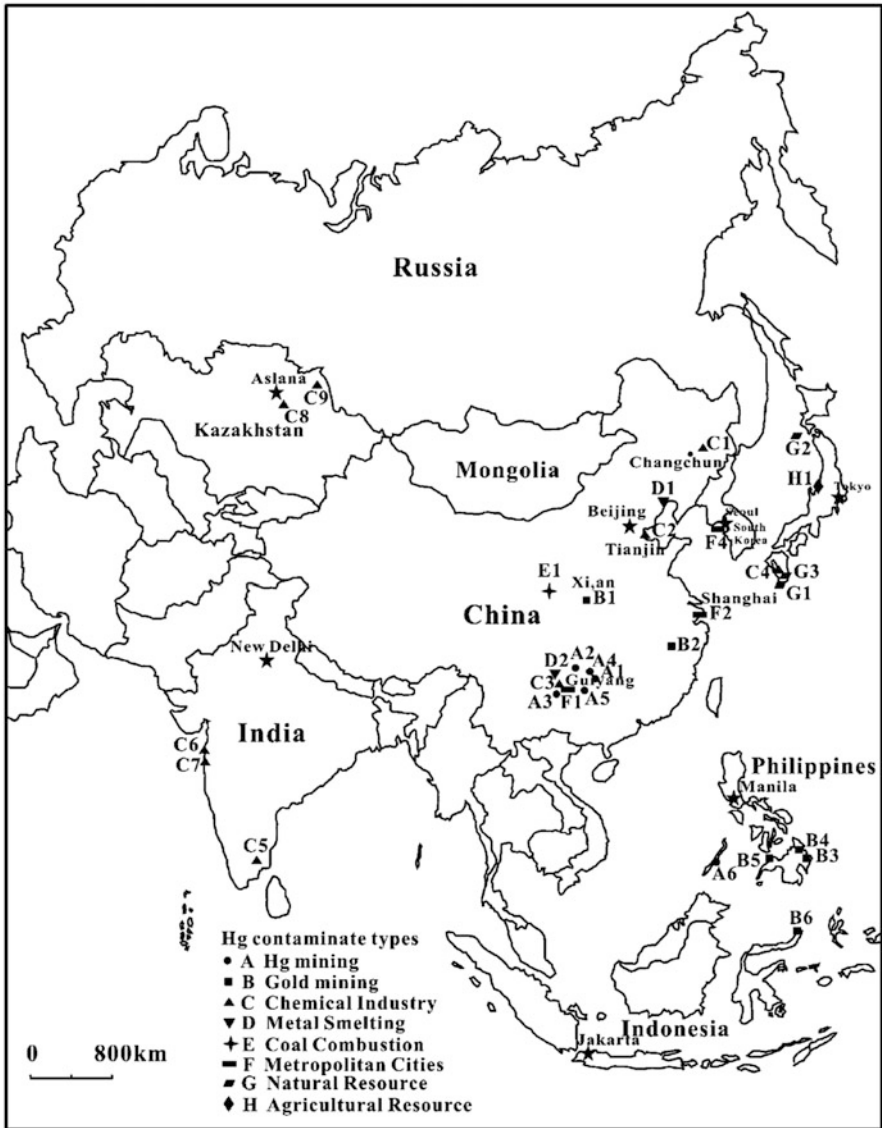


Fig. 2.4 Mercury-contaminated sites in Asia (Reprinted from Ref. [21], Copyright 2009, with permission from Elsevier)

Mercury (Hg) is one of the most important environmental contaminants emitted to the atmosphere, water, and land. Global atmospheric emission of mercury from all anthropogenic sources in 2005 was estimated to be 1,930 tons. Coal combustion processes are found to be the main source of anthropogenic mercury emission to the atmosphere. These processes are accounting for about 45 % of the total global anthropogenic mercury emissions.

It is predicted that mercury emission in the power plant in Europe until 2020 will decrease [24] (in Fig. 2.3), as a result of mercury emission control.

Global oceanic emission is estimated to be 800–2,600 tons/a and global natural terrestrial emission is estimated to be 1,000–3,200 tons/a [25]. These give a global natural mercury emission of 1,800–5,800 tons/a. Such a significant yet uncertain emission quantity can considerably influence model results of atmospheric mercury.

The global anthropogenic Hg emission to the atmosphere is estimated to be 2,190 tons in 2000. The largest emissions occur from combustion of fossil fuels, mainly coal in utility, industrial, and residential boilers. As much as two-thirds of the total emission of ca. 2,190 ton of Hg came from combustion of fossil fuels. And Asian countries contributed about 54 % (1,179 tons) to the global Hg emission from all anthropogenic sources worldwide in 2000 [26]. The mercury-contaminated sites are shown in Fig. 2.4. And the major emissions of Hg to the global atmosphere still occur from combustion of fossil fuels (879 tons). China heads the list of the 10 countries with highest Hg emissions from anthropogenic activities. With more than 600 tons of Hg, China contributes about 28 % to the global emissions of mercury. There are also four other Asian countries (India, Japan, Kazakhstan, Korea Democratic Republic) on the list. Due to long-range transport, mercury emissions from Asia are thought to significantly influence mercury deposition over North America [21, 27].

Current literature suggests that South Africa may be the second highest emitter of Hg in the world, after China [28]. This contention has largely been attributed to gold mining and stationary coal combustion. South Africa is the third largest coal producer in the world, and coal accounts for 64 % of South Africa's primary energy supply (DME 2005). Electricity generation accounts for 61 % of the total coal consumption in South Africa, and more than 90 % of the country's electricity requirements are provided for by coal-fired power plants [29].

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Coal Fired Flue Gas Mercury Emission Controls

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