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FEATURE ARTICLE

A review of atmospheric mercury emissions, pollution and control in China

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Abstract Mercury, as a global pollutant, has significant impacts on the environment and human health. The current state of atmospheric mercury emissions, pollution and control in China is comprehensively reviewed in this paper. With about 500–800 t of anthropogenic mercury emissions, China contributes 25%–40% to the global mercury emissions. The dominant mercury emission sources in China are coal combustion, non-ferrous metal smelting, cement production and iron and steel production. The mercury emissions from natural sources in China are equivalent to the anthropogenic mercury emissions. The atmospheric mercury concentration in China is about 2–10 times the background level of North Hemisphere. The mercury deposition fluxes in remote areas in China are usually in the range of 10–50 $\mu g \cdot m^{-2} \cdot yr^{-1}$. To reduce mercury emissions, legislations have been enacted for power plants, non-ferrous metal smelters and waste incinerators. Currently mercury contented in the flue gas is mainly removed through existing air pollution control devices for sulfur dioxide, nitrogen oxides, and particles. Dedicated mercury control technologies are required in the future to further mitigate the mercury emissions in China.

Keywords atmospheric mercury, emissions, pollution, control, China

1 Introduction

Mercury (Hg) is one of the most important environmental contaminants that has caused global concerns because of its toxicity, long range transport, persistence and bioaccumulation in the environment [1]. The most significant releases of Hg emissions are to the air [2]. Mercury in the atmosphere exists mainly in the gaseous form [3]. Atmospheric Hg is usually defined as different species, including total gaseous mercury (TGM) and particle-bound mercury (PBM or PHg) in physical forms. TGM is further divided into gaseous elemental Hg (GEM or Hg^0) and reactive gaseous Hg (RGM or Hg^{2+}) [4,5]. The three Hg species exhibit very different atmospheric behaviors. RGM and to some extent PHg have a high surface reactivity and water solubility and are readily scavenged from atmosphere via wet and dry deposition. However, GEM, the predominant form of atmospheric Hg ($>$ 90% of the total Hg in atmosphere), is fairly stable in the lower atmosphere with a residence time of several months to a year [6–9]. Under normal atmospheric conditions it disperses globally before it is oxidized by atmospheric oxidants (such as Br, OH, O_3, BrO, etc.) to RGM. Therefore, GEM has been identified as a global pollutant. In 2013, 140 countries adopted the first legally binding international treaty aimed at reducing mercury emissions, so called as Minamata Convention on Mercury.

The source and fate of atmospheric Hg are the essential topics in the global Hg cycle. As an economic giant of the world, China has experienced dramatic economic growth over the past three decades, accompanied by an annual growth rate of energy consumption as high as 10%. China has become the world's largest energy consumer. Energy consumption, especially fossil fuel consumption, as well as the production of cement, non-ferrous metals, iron and steel, are the main sources of anthropogenic mercury emissions in China [10]. China plays a key role in both current and future atmospheric mercury emissions and significantly affects the global Hg cycle. It is estimated that atmospheric mercury emissions in China account for 25%–40% of global mercury emissions [1]. To better understand the global Hg cycle, it is necessary to improve the understandings of atmospheric Hg emissions, pollution, and control measures in China. This paper provides an

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overview of current atmospheric Hg studies in China and points out the future research needs.

2 Atmospheric mercury emissions in China

2.1 Mercury emissions from anthropogenic sources

The first complete anthropogenic mercury emission inventory of China was developed by Streets et al. [11]. An inventory of mercury emissions from anthropogenic activities in China was compiled for the year 1999 from official statistical data. China's total mercury emissions in 1999 were estimated to be 536±236 t. The inventory included open biomass burning, but did not include natural sources or re-emissions of previously deposited mercury. The results showed that 45% of the mercury was from non-ferrous metal smelting, 38% from coal combustion, and 17% from miscellaneous activities including battery, fluorescent lamp production and cement production. Pacyna et al. [12] presented an inventory of global emissions of mercury to the atmosphere from anthropogenic sources for the year 2000. Based on their calculations, China, with a total amount of 605 t, contributed about 28% of the global inventory. Stationary combustion and industry production accounted for 78% and 21% in China's mercury emissions, respectively. Wu et al. [10] calculated the historical mercury emissions from all anthropogenic sources in China and found it increased at an average annual rate of 2.9% during the period of 1995–2003, reaching 696±307 t in 2003. Non-ferrous metal smelting and coal combustion contributed 46% and 37% of the total mercury emissions in China in 2003, respectively. Pacyna et al. [13] presented the 2005 global inventory of anthropogenic mercury emissions to the atmosphere as a contribution to the UNEP *Global Atmospheric Mercury Assessment*. With mercury emissions of 825 t, China contributed 43% of the global inventory, and fossil fuel combustion accounted for 47% of the China inventory [14]. Pirrone et al. [15] calculated the global emissions of total mercury from major anthropogenic sources and found China's contribution was 609 t in 2007. Stationary combustion and non-ferrous metal production accounted for 44% and 33% of the whole inventory, respectively. The previous studies on the mercury emissions from anthropogenic sources in China are summarized in Table 1.

There are still large uncertainties in the existing inventories. The uncertainty level of Wu et al.'s inventories [10] were ±78% and ±44% (95% confidence interval) in the estimates of total mercury emissions in 1995 and 2003, respectively. The uncertainties of the inventory were generated from both the activity data and the emission factors. The method for uncertainty analysis in their study was essentially a semiquantitative approach based on uncertainty ranking of each parameter in inventory development. Therefore, the actual uncertainty was probably even larger. Many activities which release large amounts of mercury tend to lie on the fringe of official statistics. Guan et al. [26] indicated that there were large gaps between national and provincial coal consumption

Table 1 Studies on the atmospheric mercury emissions from anthropogenic sources in China

inventory year	source type	amount/t	uncertainty	reference
1999	anthropogenic sources	536	±44%	Streets et al. [11]
2000	anthropogenic sources	605	–	Pacyna et al. [12]
2003[a)]	anthropogenic sources	696	±44%	Wu et al. [10]
2005	anthropogenic sources	825	±40%	Pacyna et al. [13]
2007	anthropogenic sources	609	±30%[b)]	Pirrone et al. [15]
1994	coal combustion	296	–	Feng and Hong [16]
1995	coal combustion	214	–	Wang et al. [17]
2000	coal combustion	162[c)] 220[c)]	–	Jiang et al. [18]
2007[d)]	coal combustion	306	–	Tian et al. [19]
2010	coal combustion	254	(−34%, +44%)	Zhang [20]
2003[e)]	non-coal sources	393	–	Wang et al. [21]
2005	non-ferrous metal smelting	83.2	–	Hylander and Herbert [22]
2006[f)]	zinc smelting	104.2	–	Li et al. [23]
2006	zinc smelting	107.7	–	Yin et al. [24]
2010[g)]	non-ferrous metal smelting	72.5	±85%	Wu et al. [25]

Notes: a) Inventories for 1995–2003 were developed, but only the 2003 inventory is listed; b) the uncertainty was for the global inventory, since the uncertainty for the China inventory was not mentioned; c) using different mercury contents of coal; d) inventories for 1980–2007 were developed, but only the 2007 inventory is listed; e) inventories for 1995–2003 were developed, but only the 2003 inventory is listed; f) inventories for 2002–2006 were developed, but only the 2006 inventory is listed; g) inventories for 2000, 2003, 2005, 2007, 2010 were developed, but only the 2010 inventory is listed

statistics in China. The lack of onsite measurements of mercury release rates, removal efficiencies and species profiles from Chinese boilers and smelters also result the high uncertainties. There are even larger discrepancies in estimates of the typical mercury content of coal and concentrate ore in many provinces. Until these inadequacies are fulfilled, the uncertainty in the mercury emissions estimate will persist.

Based on the results from previous studies, coal combustion, non-ferrous metal smelting, cement production and iron and steel production are currently considered as the dominant mercury emission sources in China. The mercury emissions from these sectors are further discussed as follows.

(1) Coal combustion

Feng and Hong [16] estimated of atmospheric mercury emissions from coal combustion in China in 1994 and reported the total mercury emissions to be 296 t. Wang et al. [17] calculated the total mercury emissions from coal combustion in China and found the total emissions to be 214 t in 1995 with an annual growth rate of 4.8% from 1978 to 1995. The difference between these two studies was mainly caused by the average mercury content of coal in China, which was 0.32 $mg \cdot kg^{-1}$ in Feng and Hong's study [16] and 0.22 $mg \cdot kg^{-1}$ in Wang et al.'s [17]. Jiang et al. [18] developed a more detailed mercury emission inventory for the coal combustion sector in China and classified the emission sources into 65 categories by sectors, fuel types, boiler types and pollution control technologies. With two different sets of coal mercury content data the total amounts of mercury released to the atmosphere in 2000 in China were estimated at 162 and 220 t, respectively. Zhang [20] developed a probabilistic emission factor model and estamated the mercury emissions from coal combustion in China in 2010 to be 254 t. Industrial boilers, power plants and residential boilers contributed 47%, 39% and 8%, respectively.

Mercury emission from coal combustion is mainly influenced by two parameters, mercury content of coal and mercury removal efficiencies of air pollution control devices (APCDs). For the mercury content of Chinese raw coal, Wang et al. [27] and Zhang et al. [28] used 0.22 $mg \cdot kg^{-1}$ as a national average. Other researches yielded estimated values of 0.15 $mg \cdot kg^{-1}$ [29] and 0.16 $mg \cdot kg^{-1}$ [30]. United State Geological Survey (USGS) analyzed 305 samples from all provinces in China and obtained an average mercury content of 0.16 $mg \cdot kg^{-1}$ [31]. Based on data from USGS and other studies, Streets et al. [11] presented a complete database of mercury content for China by province. Ren et al. [32] summarized previous results of 619 samples in their book. Zheng et al. [33] summarized 1,699 samples from previous studies and reported the national average to be 0.19 $mg \cdot kg^{-1}$. Tian et al. [19,34–36] summarized previous studies and reported the national average mercury content in the range of 0.18–0.20 $mg \cdot kg^{-1}$. We updated the mercury content database in our recent study [37] and got a national average of 0.17 $mg \cdot kg^{-1}$. Table 2 summarizes the mercury content of raw coal in China by province/autonomous/municipalities (Taiwan, Hong Kong and Macao are not included because lack of data).

There are remarkable differences between the mercury removal efficiencies of different APCDs for coal combustion. Table 3 shows the mercury removal efficiencies of 18 typical APCD combinations for different coal ranks. These data are summarized from 124 onsite measurements [20,38–64]. For the APCD combination of pulverized coal boiler and electrostatic precipitators (PC + ESP), the average mercury removal efficiency for lignite coal is relatively higher, while that for anthracite coal is relatively lower. It can be seen from the database that coal rank is not the most significant impact factor for the removal efficiency, while the influence of coal chlorine content is probably more crucial [20]. The influence of boiler type on the efficiency is not significant except for circulating fluidized bed boilers (CFB). The mercury removal efficiencies for most of the APCD combinations vary within a large range. The average efficiency of wet scrubber (WS) is 23%, and that of electrostatic precipitators (ESP) excluding (CFB + ESP) is 29%. Those of electrostatic precipitators and wet flue gas desulfurization (ESP + WFGD) and fabric filters (FF) are in the range of 60%–70%. Selective catalytic reduction (SCR) can strengthen the mercury removal efficiency of ESP + WFGD by converting Hg^0 to Hg^{2+}, which results the reomoval efficiency of SCR + ESP + WFGD to 69%. Among the most commonly used APCD combinations in China, the combination of FF + WFGD has the highest efficiency, that is, 86%.

(2) Non-ferrous metal smelting

In terms of mercury emissions, non-ferrous metals usually refer to lead, zinc, copper and gold.

Wang et al. [21] estimated the mercury emissions from non-coal sources in China during the period of 1995–2003. The non-coal mercury emissions in China reached 393 t in 2003, of which 51%, 18% and 4% were from zinc, lead and copper smelting, respectively. Hylander and Herbert [22] estimated the mercury emissions from zinc, lead and copper smelting in China to be 83.2 t. Based on the field measurements in five zinc smelters, Li et al. [23] calculated the mercury emissions from Chinese zinc smelters to be 80.7–104.2 t during 2002–2006. Yin et al. [24] used the mass balance method and found the mercury emissions from the zinc smelting sector in China to be 107.7 t in 2006. Wu et al. [25] updated the mercury emission inventory for zinc, lead and copper smelting and found that the atmospheric mercury emissions from non-ferrous metal smelters in 2000, 2003, 2005, 2007 and 2010 were 67.6, 100.1, 86.7, 80.6 and 72.5 t, respectively.

The mercury emitted from non-ferrous metal smelting is

Table 2 Mercury content of raw coal in China by province/autonomous/municipalities (unit: $mg \cdot kg^{-1}$)

province	Zhang et al. [37]	Zheng et al. [33]	Ren et al. [32]	Streets et al. [11]	USGS [31]	Huang & Yang [29]	Wang et al. [27]
Anhui	0.20(9)	0.21	0.46(50)	0.26	0.19(11)	0.26	0.22
Beijing	–	0.34	0.10(1)	0.44	0.55(1)	–	0.34
Chongqing	0.41(5)	–	0.64(12)	–	0.15(7)	–	–
Fujian	–	–	–	0.08	0.07(3)	–	–
Gansu	0.18(2)	–	1.35(1)	0.05	0.05(5)	–	–
Guangdong	–	–	0.10(1)	0.15	0.06(2)	–	–
Guangxi	–	–	–	0.30	0.35(5)	–	–
Guizhou	0.21(30)	1.14	0.70(133)	0.52	0.20(16)	0.52	–
Hainan	–	–	–	0.15	–	–	–
Hebei	0.17(9)	0.46	0.16(33)	0.14	0.14(15)	0.80	0.13
Heilongjiang	0.03(10)	0.13	0.12(14)	0.09	0.06(10)	0.14	0.12
Henan	0.14(10)	0.17	0.14(115)	0.25	0.21(27)	0.17	0.30
Hubei	–	–	0.23(1)	0.16	0.16(3)	–	–
Hunan	–	0.07	0.08(14)	0.10	0.14(10)	0.07	–
Inner Mongolia	0.18(30)	0.16	0.17(14)	0.22	0.16(16)	0.02	0.28
Jiangsu	0.18(5)	0.09	0.18(10)	0.16	0.35(6)	0.09	–
Jiangxi	–	0.16	0.13(4)	0.22	0.27(7)	–	0.16
Jilin	–	0.34	0.34(2)	0.20	0.07(5)	–	0.33
Liaoning	0.10(10)	0.17	0.14(16)	0.17	0.19(9)	0.13	0.20
Ningxia	–	–	0.28(19)	0.20	0.21(4)	–	–
Qinghai	–	–	0.31(4)	0.04	0.04(1)	–	–
Shaanxi	0.25(17)	0.64	0.30(3)	0.11	0.14(11)	0.08	0.16
Shandong	0.16(14)	0.28	0.18(11)	0.18	0.13(19)	0.21	0.17
Shanghai	–	–	–	–	–	–	–
Shanxi	–	0.08	0.17(79)	0.16	0.15(88)	0.20	0.22
Sichuan	0.34(4)	0.18	0.35(14)	0.14	0.09(11)	–	–
Tianjin	–	–	–	–	–	–	0.18
Xinjiang	0.02(12)	0.03	0.09(6)	0.02	0.03(6)	–	0.03
Xizang	–	–	–	–	–	–	–
Yunnan	0.08(10)	0.30	0.32(56)	0.29	0.14(7)	0.34	–
Zhejiang	–	–	0.75(2)	0.35	–	–	–
National	0.17(177)	0.19	0.33(619)	0.19	0.16(305)	0.15	0.22

Notes: The numbers in brackets are number of samples for each province/autonomous/municipality; Taiwan, Hong Kong and Macao are not included because lack of data

mainly from the non-ferrous metal concentrates. Mercury content of non-ferrous metal concentrates was much higher than that of coal. Nationwide sampling on non-ferrous metal concentrates was carried out and mercury content of different concentrates in China by province/autonomous/municipalities was shown in Table 4 (Taiwan, Hong Kong and Macao are not included because lack of data). Streets et al. [11] pointed out that the mercury concentration in Chinese zinc concentrates varied from less than 1 $mg \cdot kg^{-1}$ to over 1000 $mg \cdot kg^{-1}$. Song et al. [65] analyzed 208 zinc concentrate samples from all over China and got a national average of 9.45 $mg \cdot kg^{-1}$ (geometric mean). Yin et al. [24] found that the mercury concentration in zinc ores depends on the ore types and their geneses. Based on the analysis of 82 samples, the geometric mean of mercury concentration in zinc concentrate in China was 7.34 $mg \cdot kg^{-1}$. Zinc concentrates from the deposits in Shaanxi, Inner Mongolia, Gansu and Guangdong have the highest concentration of

Table 3 Mercury removal efficiencies of air pollution control devices (APCDs) in coal-fired power plants (%)

	bituminous	anthracite	lignite	subbituminous
PC + ESP	29(42)	22(4)	38(6)	27(11)
PC + ESP + WFGD	63(14)	81(1)	65(1)	50(3)
PC + FF	66(8)	–	–	73(2)
PC + WS	12(1)	–	33(1)	–
PC + SCR + ESP + WFGD	69(4)	–	–	–
PC + FF + WFGD	90(2)	–	–	–
PC + SDA + FF	99(1)	–	66(1)	13(1)
PC + SDA + ESP	–	–	–	70(1)
PC + ESP + CFB-FGD + FF	68(1)	–	–	–
PC + SCR + SDA + FF	98(2)	–	–	–
PC + NID + ESP	–	90(1)	–	–
PC + SNCR + ESP	83(1)	–	–	–
CFB + ESP	99(1)	–	61(2)	–
CFB + FF	100(2)	–	59(1)	–
CFB + SNCR + FF	89(1)	–	–	79(1)
SF + WS	16(4)	59(1)	–	–
SF + FF + WFGD	–	77(1)	–	–
BB + WS	–	19(1)	–	–

Notes: PC – pulverized coal boiler; CFB – circulating fluidized bed boiler; SF – stoker fired boiler; BB – boiling bed boiler; WS – wet scrubber; ESP – electrostatic precipitators; FF – fabric filters; FGD – flue gas desulfurization; WFGD – wet FGD; CFB-FGD – circulating fluidized bed FGD; NID – novel integrated desulfurization; SDA – spray dryer absorber; SCR – selective catalytic reduction; SNCR – selective non-catalytic reduction. The numbers in brackets are number of tests

48.2 $mg \cdot kg^{-1}$. According to Wu et al. [25], the geometric mean mercury concentrations of Chinese zinc, lead and copper concentrates were 9.7 $mg \cdot kg^{-1}$, 10.3 $mg \cdot kg^{-1}$ and 2.9 $mg \cdot kg^{-1}$, respectively.

Field experiments have been conducted and the removal efficiencies of APCDs in non-ferrous metal smelters were obtained. Hylander and Herbert [22] reported that the mercury removal efficiency in smelters with double-conversion-double-adsorption (DCDA) acid plant was about 99%, and that for single-conversion-single-adsorption (SCSA) technology was 95%. Wang et al. [66] found that the mercury removal efficiencies of flue gas cleaning, electrostatic demister, mercury reclaiming and acid plant with DCDA technology were 17.4%, 30.3%, 87.9% and 97.4%, respectively. According to the study of Li et al. [23], 3.5%–9.8% of mercury were removed by gas cleaning process. The mercury reclaiming tower had a high removal efficiency of 89.2%–93.5%. Then up to 29.6%–65.3% of mercury was removed by the sulfuric acid plant. Zhang et al. [67] tested six non-ferrous metal smelters and found that the removal efficiencies of electrostatic precipitators, fabric filters, acid plant with SCSA technology and acid plant with DCDA technology were 11.6%, 43.7%, 83.1% and 99.6%, respectively.

In the period of 1995–2003, the atmospheric mercury emissions from gold production decreased from 96 t to 45 t [10]. In 2003, large-scale gold production (LSGP) and artisanal and small-scale gold mining (ASGM) accounted for 36% and 64%, respectively. In the LSGP plant, various technologies are adopted according to the properties of raw materials. Flotation-cyanidation method is commonly used for gold extraction from conventional materials, while roasting-extraction method and bio-oxidation method are two main processes for refractory gold ores. Mercury in the gold ores for flotation-cyanide and bio-oxidation is mainly released to solid waste or waste water, with little mercury emissions to air. Therefore, gold smelters with roasting-extraction method are major mercury emission sources in gold production. The mercury emission factor for LSGP was estimated to be 0.79 $g \cdot g^{-1}$ [11]. Pacyna et al. [12] and Pirrone et al. [15] used a factor of 0.5 $g \cdot g^{-1}$, and Pacyna et al. [13] updated the value to be 0.25 $g \cdot g^{-1}$ in 2010. The mercury content of ore and application percentage of various technologies are also major factors resulting uncertainties. In ASGM plants, gold ores are extracted by amalgamation process, which can lead to severe atmospheric mercury pollution. The mercury emission factor for ASGM plants was as high as 15.0 $g \cdot g^{-1}$ [11], which was much larger than that for LSGP plants. However, the estimation for mercury emissions from ASGM was based on the study of Gunson and Veiga in 2004 [68]. China has forbidden the production of ASGM since 2004 and it is hard to estimate the emissions of such illegal activities.

Table 4 Mercury content of non-ferrous metal concentrates in China by province/autonomous/municipalities (unit: $mg \cdot kg^{-1}$)

province	zinc			lead	copper
	Wu et al. [25]	Yin et al. [24]	Song et al. [65]	Wu et al. [25]	Wu et al. [25]
Anhui	4.10(1)	4.10(1)	4.10(1)	14.66(2)	0.34(4)
Chongqing	–	–	–	114.91(1)	–
Fujian	0.54(11)	0.54(4)	0.52(10)	12.63(4)	–
Gansu	499.91(9)	132.57(6)	499.91(9)	10.77(3)	2.86(4)
Guangdong	72.16(3)	6.21(4)	85.96(3)	43.75(3)	0.05(1)
Guangxi	9.34(9)	9.09(7)	2.87(4)	10.13(12)	0.62(3)
Guizhou	–	2.13(1)	–		–
Hebei	–	0.39(1)	–		–
Heilongjiang	–	7(1)		25.67(1)	–
Henan	4.96(4)	13.54(1)	7.68(3)	2.25(7)	–
Hubei	–	0.76(1)	–	6.86(1)	0.99(6)
Hunan	4.72(26)	3.17(9)	3.74(12)	1.31(11)	–
Inner Mongolia	2.16(6)	4.22(2)	2.28(5)	62.21(4)	1.84(2)
Jiangsu	13.29(2)	1.64(2)	13.29(2)	18.61(3)	0.06(1)
Jiangxi	1.47(10)	1.55(5)	1.88(9)	19.51(1)	4.66(7)
Jilin	–	10.00(1)	–	55.58(2)	–
Liaoning	–	43.17(4)	–	61.04(6)	–
Qinghai	–	27.79(2)	–	0.6(3)	1.77(1)
Shaanxi	240.77(12)	50.31(2)	233.07(10)	45.14(3)	–
Shandong	–	1.55(1)	–	4.92(1)	1.5(1)
Shanghai	–	–	–		–
Shanxi	–	–	–	52.17(1)	0.14(3)
Sichuan	45.55(10)	15.13(4)	20.71(3)	26.46(5)	2.15(3)
Xinjiang	16.86(3)	4.25(5)	6.86(2)		2.02(7)
Xizang	0.23(1)	–	0.23(1)	0.02(1)	–
Yunnan	10.98(6)	4.51(7)	11.82(5)	21.54(3)	13.68(12)
Zhejiang	0.88(5)	14.51(3)	1.17(3)	20.96(5)	–
National	9.74(118)	7.34(82)	4.35(73)	10.29(83)	2.87(55)

Notes: The numbers in brackets are number of samples for each province/autonomous/municipalities; Taiwan, Hong Kong and Macao are not included because lack of data

(3) Cement production

Cement production is thought to be one of the largest sources of global anthropogenic mercury emissions. Pirrone et al. [15] calculated the mercury emission from this sector was 236 t. China produced more than 2 billion tons of cement in 2011, accounting for more than half of the world production. Thus the mercury emissions from cement plants are very important for China. However, there is a lack of information on emissions of the cement manufacturing process currently, especially for China.

Raw materials used in cement manufacturing process include limestone, coal, clay, slag and other wastes, among which the amount of limestone is the largest. Sikkema et al. [69] summarized the mercury concentrations of raw materials. Based on his result, the Hg concentration of limestone has a wide range of 0.5–2000 $\mu g \cdot kg^{-1}$. The Hg concentration of coal also varies from 5 to 1000 $\mu g \cdot kg^{-1}$. The concentration of clay, shale and sand is in the range of 0.5–400 $\mu g \cdot kg^{-1}$, with a mean value of lower than 100 $\mu g \cdot kg^{-1}$. Other wastes, such as iron ore and fly ash, though having a concentration range of 0.5–600 $\mu g \cdot kg^{-1}$, contribute little to mercury input because of their small usage amount. Won et al. [70] reported that the average concentrations of limestone and fly ash were 19.1 and 144 $\mu g \cdot kg^{-1}$. Mlakar et al. [71] used 7.9 $\mu g \cdot kg^{-1}$ as the mercury concentration of limestone. In China, Li [72] found the mercury concentration of limestone was 11–28 $\mu g \cdot kg^{-1}$, but the result of Zhang [73] was only 2 $\mu g \cdot kg^{-1}$. Obviously, more research is needed on the mercury concentrations of raw materials, especially limestone.

Over 80% of cement plants use precalciner process in China in 2011. In this process, part of the flue gas at the

kiln tail is used to heat the raw materials. Usually fabric filters (FF) are employed at the kiln tail and electrostatic precipitators (ESP) are employed at the kiln head to collect dust. These facilities remove most of the particulate mercury in the flue gas. The collected dust is mixed with raw materials and cycled in the manufacturing process. The mercury concentration in flue gas becomes quite high because of such cycle. The result obtained by Mlakar et al. [71] confirmed this conclusion. The output of mercury is mainly through flue gas, and the clinker only accounts for less than 10% [70,71].

The emission factor obtained by Won et al. [70] was 0.026–0.034 $g \cdot t^{-1}$ clinker. UNEP [74] used 0.1 $g \cdot t^{-1}$ cement for global emission estimate. Li and Zhang reported lower emission factors for shaft kiln, 0.0036 and 0.0069 $g \cdot t^{-1}$ cement, respectively [72,73]. The emission factors of rotary kiln are 0.008 and 0.0138 $g \cdot t^{-1}$ cement.

(4) Iron and steel production

Iron and steel production emits 43 t mercury to air, contributing about 5% to global mercury emissions [15]. Currently there is little information on this source. The production process for iron and steel is quite complicated and has more emission points than other industrial sources. Fukuda et al. [75] tested an iron and steel plant in Japan and found that iron ore was the main input of mercury for iron and steel production, accounting for 48.2% of total mercury input. The mercury concentration of iron ore was 30.8 $\mu g \cdot kg^{-1}$. Coal was also one of the import sources of mercury input, contributing 44.5% of total mercury input, almost equal amount as iron ore. The sinterer in iron and steel production process emits more than half of the mercury to air. Dust collector and desulfurization device are installed in the downstream of flue gas generated in the sinterer, and have high mercury removal efficiency. The emission factor from Fukuda et al.'s study [75] was 0.0488 $g \cdot t^{-1}$ steel, similar to the value used by UNEP (0.04 $g \cdot t^{-1}$) [14]. Currently there is lack of information on iron and steel manufacturing process in China, but the steel production is 0.683 billion tons in 2011, thus efforts are needed to study the mercury emissions from iron and steel production in China.

(5) Municipal solid waste incineration

With the rapid urbanization process in China, Municipal solid waste (MSW) incineration is playing a more and more important role in MSW management. The volume of MSW incineration has amounted to 23.2 million tons by the end of 2010, accounting for 19% of the total MSW treatment quantity. Wu et al. [10] reported the mercury emissions from MSW incineration in China to be 10.4 t in 2003. The UNEP report [14] had an evaluation of 2.05 t in 2005 for MSW incineration in China. The latest study by Tian et al. [76] showed that the total mercury emissions increased from 5.35 t in 2003 to 36.7 t in 2010. The estimates for mercury emissions from MSW in China vary significantly, which implies there is extremely large uncertainty in the evaluation of mercury emissions from this sector. More researches should be conducted in the future to improve the understanding of mercury emission characteristics in MSW incineration.

2.2 Mercury emissions from natural sources

Mercury emissions from natural sources played an important role in the biogeochemical cycling of mercury in the biosphere [77]. Recent results summarized by Pirrone et al. [15] showed that global mercury emissions from natural sources were about 5207 $t \cdot yr^{-1}$, more than two times the emissions from anthropogenic sources (2320 $t \cdot yr^{-1}$). Volcanos, geothermal sources and mercury contaminated soils are main natural sources. Air-surface (soil, water, snowpack and vegetation) exchange of gaseous elemental mercury and re-emission of deposited mercury are other important natural sources which are complex processes related to meteorological conditions, surface type and mercury contents.

Few field tests were done to measure mercury emissions from surfaces in China. Feng et al. [78] measured TGM evasion from Baihua reservoir with a surface area of 14.5 km^2 in Guiyang. Significant correlation between Hg flux and solar radiation was observed. The annual emissions of GEM from the reservoir were 752 g (about 5.9 $ng \cdot m^{-2} \cdot h^{-1}$), constituting 3% of all mercury in the reservoir. Emission from Hongfeng reservoir was also measured during cloudy weather conditions [79]. The mean TGM evasion varied from 3.1 to 8.3 $ng \cdot m^{-2} \cdot h^{-1}$ during the sampling time and significantly positive correlation between Hg flux and humidity was observed. The behavior of TGM over water surface during cloudy days was different from sunny days. Mercury emission from soil surface at four sites in Guiyang which was located in the Circum-Pacific global mercuriferous belt was measured from 21 May to 16 June in 2003 [80]. The net emission fluxes at the four sites were 44.0, 15.0, 0.4 and 32.8 $ng \cdot m^{-2} \cdot h^{-1}$, respectively. The annual mercury emission from Guiyang soil was estimated to be 408 kg, while emission from coal-combustion was 634 $kg \cdot yr^{-1}$ at the same period.

Models are useful tools to estimate mercury emissions from natural sources. Shetty et al. [81] estimated the mercury emissions from soil, vegetation and waters based on the empirical models derived from field measurements and meteorological fields calculated from MM5 model. The total GEM emissions from China were 462 $t \cdot yr^{-1}$ in 2001, which were comparable to the anthropogenic emissions (575 ± 261 $t \cdot yr^{-1}$) [82]. Pan et al. [83] estimated a total GEM emission of 1140 $t \cdot yr^{-1}$ from China using the inverse model, of which about 565 $t \cdot yr^{-1}$ was from natural sources. Strode et al. [84] used observed GEM/CO ratio at Okinawa, Japan and Mount Bachelor, Oregon as constraints to estimate mercury emissions from Asia. They reported the total GEM emissions from Asia were 1260–1470 $t \cdot yr^{-1}$, which was consistent with GEOS-Chem

model. The gridded natural emissions from the Asian domain are shown in Fig. 1.

The amounts of mercury emissions from natural and anthropogenic sources in China are estimated to be at same level, but anthropogenic sources are concentrated in East China. The emission intensities of anthropogenic sources in East China are much higher than that of natural sources. Our current understanding of mercury natural sources is quite limited and the uncertainties of all these estimates are much larger than anthropogenic sources. For bottom-up estimates, the empirical models of mercury emission from surfaces were all simple regression models. Taken soil mercury emission as an example, temperature, solar radiation and soil content are considered in the regression analysis and the field test data are from the United States [85]. The applicability of these models should be investigated as key factors such as China's soil type, meteorology, mercury content and chemical composition in soil are different from the United States. For top-down methods, the model uncertainty will significantly influence the results. For example, if we increase the boundary conditions in Pan et al.'s study [83] from 1.2 to 1.5 ng·m^{-3}, the GEM emission will decrease from 1140 to 718 t·yr^{-1}.

3 Atmospheric mercury pollution and its environmental impacts in China

3.1 Current status of atmospheric mercury pollution

One of the earliest ambient air mercury measurements in China was conducted at an urban site in Guiyang, a seriously polluted city [86]. Fu et al. [87] found the average TGM, PBM and RGM concentrations in Guiyang to be 9.7, 368 and 35.7 pg·m^{-3}, respectively. The average TGM concentration at the monitoring site was 8.4 ng·m^{-3}. Fang et al. monitored TGM and PBM at two sites in Changchun, a city with large amount of coal combustion, and found that the average TGM concentrations at the urban and suburban sites were 18.4 ng·m^{-3} and 11.7 ng·m^{-3}, respectively [88]. Wang et al. [89] reported the GEM concentration in atmosphere at a few sites around China. Their study showed that the average GEM concentrations in Mt. Waliguan (China Global Atmosphere Watch Baseline Observatory) were 1.7 ng·m^{-3} in summer and 0.6 ng·m^{-3} in winter. The average GEM concentration in Yangtze Delta regional site was 5.4 ng·m^{-3}. In Beijing urban area the average GEM concentrations were 8.3 ng·m^{-3} in winter, 6.5 ng·m^{-3} in spring, 4.9 ng·m^{-3} in summer, and 6.7 ng·m^{-3} in autumn, respectively. The atmospheric mercury concentration reached 13.5 ng·m^{-3} in Guangzhou city. The average TGM concentrations in the urban area of Chongqing, Shanghai, Ningbo and Nanjing were 6.7, 2.7, 3.8 and 7.9 ng·m^{-3}, respectively [90–93]. Based on the studies of Fu et al. [94,95], the average TGM, PBM and RGM concentrations in Mt. Gongga on the south-eastern fringe of the Tibetan plateau were 4.0 ng·m^{-3}, 31 pg·m^{-3} and 6.2 pg·m^{-3}, respectively, and a seasonal distribution pattern of TGM in ambient air was observed on the descending order of winter, autumn, spring, and summer. Wan et al. [96,97] reported that the average TGM concentration in ambient air in Changbai Mountain area was 3.6 ng·m^{-3}, while the average concentrations of RGM and TPM were 65 and 77 pg·m^{-3}, respectively. However, Fu et al. [98] monitored at a site in the vicinity of Wan et al.'s site in Changbai Mountain area, and found the

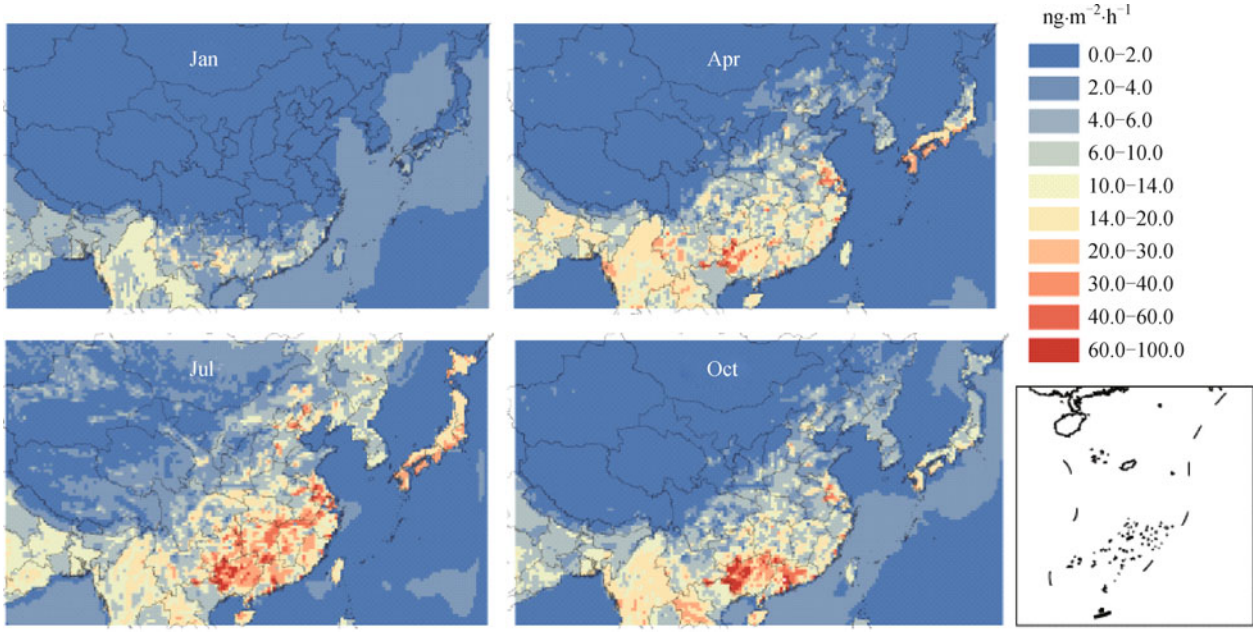

Fig. 1 Gridded natural emissions from the Asian domain in January, April, July and October, 2005

average TGM concentration was only 1.6 ng·m^{-3}. Fu et al. [99–101] monitored the atmospheric mercury at another three remote sites in China, Mt. Leigong, South China Sea and Mt. Waliguan, and obtained the average TGM concentrations at 2.8, 2.6 and 2.0 ng·m^{-3}, respectively. Ci et al. [102,103] conducted observation at two remote sites, Chengshantou and Yellow Sea, and the average TGM concentrations were 2.3 and 2.6 ng·m^{-3}, respectively. Another two remote sites were located in Pearl River Delta and Shangri-La, whose average TGM concentrations were found to be 2.9 and 2.6 ng·m^{-3}, respectively [104,105]. Tsinghua University built two monitoring sites, Miyun in Beijing and Chongming Island in Shanghai, which were considered as backgrounds for two typical regions in China. The average GEM, TPM and RGM concentrations at Miyun station were 3.2 ng·m^{-3}, 98 pg·m^{-3} and 8.9 pg·m^{-3}, respectively [106], and those at Chongming station were 2.7 ng·m^{-3}, 22 pg·m^{-3} and 8.0 pg·m^{-3}, respectively [107]. Atmospheric mercury monitoring studies in China are summarized in Table 5. The atmospheric mercury concentrations in Chinese cities are usually at the range of 6.7–18.4 ng·m^{-3}, about 2–7 times of that in the cities of Europe, United States or Japan. The atmospheric mercury concentrations in rural and remote area of China are about 2.7–11.7 ng·m^{-3} and 1.6–2.9 ng·m^{-3}, respectively, which are also much higher than the background concentration in North Hemisphere. Generally, the mercury concentrations in mountains are lower than that of surface observations.

3.2 Atmospheric mercury depositions

Atmospheric mercury deposition can be divided into dry deposition and wet deposition. Precipitation is a major form of wet deposition, while throughfall includes wet deposition and part of dry deposition. Litterfall is a dominant pathway for Hg dry deposition. Precipitation, throughfall and litterfall were all measured, while dry deposition was usually calculated in existing studies.

Fang et al. [88] estimated the wet and dry depositions at a urban site and a rural site in Changchun city. The precipitation fluxes for urban and rural Changchun area were 152 and 64 μg·m^{-2}·yr^{-1}, respectively. The dry deposition fluxes were 166 and 98 μg·m^{-2}·yr^{-1}, respectively. Wang et al. [108] collected precipitation, throughfall and litterfall samples at three forest sites including Tieshanping, Luchongguan and Mt. Leigong. The throughfall fluxes at these sites were calculated to be 140, 51 and

Table 5 Summary of atmospheric mercury monitoring studies in China

location	type	TGM /(ng·m^{-3})	PBM /(pg·m^{-3})	RGM /(pg·m^{-3})	reference
Guiyang	urban	8.4	–	–	Feng et al. [86]
Changchun	urban	18.4	276	–	Fang et al. [88]
Beijing	urban	7.9	1180	–	Wang et al. [89]
Guangzhou	urban	13.5	368	–	Wang et al. [89]
Chongqing	urban	6.7	–	–	Yang et al. [90]
Guiyang	urban	9.7	368	35.7	Fu et al. [87]
Shanghai	urban	2.7	–	–	Friedli et al. [91]
Ningbo	urban	3.8	–	–	Nguyen et al. [92]
Nanjing	urban	7.9	–	–	Zhu et al. [93]
Changchun	rural	11.7	109	–	Fang et al. [88]
Yangtze River Delta	rural	5.4	–	–	Wang et al. [89]
Miyun, Beijing	rural	3.2	98	8.9	Zhang et al. [106]
Chongming, Shanghai	rural	2.7	22	8.0	Dou [107]
Mt. Changbai	remote	3.6	77	65	Wan et al. [96,97]
Mt. Changbai	remote	1.6	–	–	Fu et al. [98]
Mt. Gongga	remote	4.0	31	6.2	Fu et al. [94,95]
Mt. Leigong	remote	2.8	–	–	Fu et al. [99]
South China Sea	remote	2.6	–	–	Fu et al. [100]
Mt. Waliguan	remote	2.0	19	7.4	Fu et al. [101]
Chengshantou, Weihai	remote	2.3	–	–	Ci et al. [102]
Yellow Sea	remote	2.6	–	–	Ci et al. [103]
Pearl River Delta	remote	2.9	–	–	Li et al. [104]
Shangri-La	remote	2.6	44	8.2	Zhang [105]

46 $\mu g \cdot m^{-2} \cdot yr^{-1}$, respectively. Wan et al. [97] and Fu et al. [99,109] reported the precipitation flux in Mt. Changbai, Mt. Leigong and Mt. Gongga to be 8.4, 6.1 and 26.1 $\mu g \cdot m^{-2} \cdot yr^{-1}$, respectively. Precipitation samples collected at a remote high elevation site, Nam Co Station, in the southern Tibetan Plateau were analyzed and a flux of 1.75 $\mu g \cdot m^{-2} \cdot yr^{-1}$ were found [110]. Dai et al. [111] evaluated the wet and dry deposition fluxes in Wanshan mercury mining area in Guizhou. The mercury concentrations in the precipitation samples at Shenchong, Dashuixi and Supeng sites were 503, 814 and 7490 $ng \cdot L^{-1}$, respectively. The dry deposition fluxes were as high as 379, 2614 and 6178 $\mu g \cdot m^{-2} \cdot yr^{-1}$ for these three sites respectively. Studies of atmospheric mercury deposition in China were summarized in Table 6. To make a comparison, the mercury wet deposition is about 1.5–20 $\mu g \cdot m^{-2} \cdot yr^{-1}$ in Northern Hemisphere background areas [113–115], and dry deposition is of the same magnitude as wet deposition, about 5.9 $\mu g \cdot m^{-2} \cdot yr^{-1}$ in in arid south central New Mexico [116] and 4.9 $\mu g \cdot m^{-2} \cdot yr^{-1}$ in the Florida Everglades [117].

3.3 Environmental impacts from atmospheric mercury emission

Environment around non-ferrous metal smelters was heavily polluted by the atmospheric emission. Surface water samples collected from the site located at upper reaches of the artisanal zinc smelting areas in Guizhou reached 12–30 $ng \cdot L^{-1}$, which was mainly from dry deposition of atmospheric mercury [118]. Topsoil and corns also suffered from severe contamination. The average mercury concentrations in the topsoil (0–5 cm) and the leaf of corn in these polluted areas reached 0.38 $mg \cdot kg^{-1}$ and 0.32 $mg \cdot kg^{-1}$, respectively. The cities of Huludao and Zhuzhou were also heavily contaminated of mercury caused by zinc smelting. Mercury concentrations

Table 6 Summary of atmospheric mercury deposition fluxes in China

location	location type	deposition type	concentration /($ng \cdot L^{-1}$)	deposition flux /($\mu g \cdot m^{-2} \cdot yr^{-1}$)	reference
Shenchong, Guizhou	polluted	precipitation	503	29.1	Dai et al. [111]
Shenchong, Guizhou	polluted	dry deposition	–	379	Dai et al. [111]
Dashuixi, Guizhou	polluted	precipitation	814	68.8	Dai et al. [111]
Dashuixi, Guizhou	polluted	dry deposition	–	2614	Dai et al. [111]
Supeng, Guizhou	polluted	precipitation	7490	593	Dai et al. [111]
Supeng, Guizhou	polluted	dry deposition	–	6178	Dai et al. [111]
Changchun, Jilin	urban	precipitation	345	152	Fang et al. [88]
Changchun, Jilin	urban	dry deposition	–	166	Fang et al. [88]
Changchun, Jilin	rural	precipitation	139	63.7	Fang et al. [88]
Changchun, Jilin	rural	dry deposition	–	98.1	Fang et al. [88]
Wujiang, Guizhou	rural	precipitation	36.0	34.7	Guo et al. [112]
Tieshanping, Chongqing	rural	precipitation	55.3	67.3	Wang et al. [108]
Tieshanping, Chongqing	rural	throughfall	98.9	140	Wang et al. [108]
Tieshanping, Chongqing	rural	litterfall	105	221	Wang et al. [108]
Luchongguan, Guizhou	rural	throughfall	83.6	51.1	Wang et al. [108]
Mt. Gongga	remote	precipitation	9.9	9.1	Fu et al. [95]
Mt. Changbai	remote	precipitation	13.4	8.4	Wan et al. [97]
Mt. Leigong	remote	precipitation	19.5	22.4	Wang et al. [108]
Mt. Leigong	remote	throughfall	54.2	46.4	Wang et al. [108]
Mt. Leigong	remote	litterfall	135	78.0	Wang et al. [108]
Mt. Leigong	remote	precipitation	4.0	6.1	Fu et al. [99]
Mt. Leigong	remote	throughfall	8.9	10.5	Fu et al. [99]
Mt. Leigong	remote	litterfall	91	39.5	Fu et al. [99]
Mt. Gongga	remote	precipitation	14.3	26.1	Fu et al. [109]
Mt. Gongga	remote	throughfall	40.4	57.0	Fu et al. [109]
Mt. Gongga	remote	litterfall	35.7	35.5	Fu et al. [109]
Nam Co, Tibet	remote	precipitation	4.8	1.75	Huang et al. [110]

Notes: The unit of mercury concentration in litterfall is $ng \cdot g^{-1}$

in the soils were 1.28–2.89 mg·kg^{-1} and 0.13–28.18 mg·kg^{-1}, respectively [119–121]. Study of the topsoil in vicinity of secondary copper smelters indicated annual mercury emission of 17.3–27.2 kg to topsoil [122].

As a global pollutant, mercury emission from China can be transported long distances and its influence to mercury concentrations and deposition in China, North America, the Arctic and other regions has been discussed. Many model results showed high deposition in East Asia caused by high emission density [123–126], and the amount of mercury export from Asia continent was very large. Lin et al. [125] estimated mercury deposition in East Asia to be 821 t·yr^{-1} and most of the deposition (75%) was caused by anthropogenic sources for the modeling year 2005. The outflow of mercury caused by emissions in East Asia was in the range of 1396–1671 t·yr^{-1}, of which 40%–50% was caused by anthropogenic sources. Pan et al. [126] estimated that the dry deposition and wet deposition in East Asia were in the range of 590–735 t and 482–696 t in 2001 and mercury outflow from East Asia caused by anthropogenic sources was about 681–714 t·yr^{-1}, constituting 70% of total emissions.

Seigneur et al. [127] used a global CTM model and a nested continental model, TEAM to estimate the contributions of global sources to mercury deposition in the United States. Results showed that Asia anthropogenic emissions contributed 21% of total deposition to the contiguous United States and anthropogenic emissions from North America accounted for 30%. Strode et al. [84] reported that anthropogenic sources from Asia contributed 14% of mercury deposition in United States while North American anthropogenic emissions contributed 16%. Although various studies got different values, all results indicated that the contribution of Asia anthropogenic sources was equivalent to that of North America. Durnford et al. [128] studied the transport of mercury to the Arctic using Environment Canada's Global/Regional Atmospheric Heavy Metals model (GRAHM). They found that Asia was the dominant sources at all stations and all seasons. Although Asia had low transport efficiency compared to Russia and Europe, it still generated most long-range-transport pollution events at the monitoring sites considered.

4 Mercury emission control in China

4.1 Mercury emission control legislations

Currently China has enacted the mercury emission standards for power plants, non-ferrous metal smelters, and waste incinerators.

In 2010, the Ministry of Environmental Protection of China initiated a pilot mercury emission control project involving 16 power plants. In 2012, China's new state-of-the-art national Emission Standard of Air Pollutants for Thermal Power Plants [129] went into effect, replacing the standards that had been in effect since 2003. This standard not only tightened the standards for particles, SO_2, and NO_x substantially, but also included the Hg concentration limits for the first time, starting from January 1, 2015. This standard brings Chinese power plant regulation generally in line with developed world standards in important respects (see Table 7).

For lead and zinc industry, emission standard of pollutants was issued in 2010 (GB 25466-2010). The Hg emission limits for existing lead and zinc smelters are 1.0 mg·m^{-3} before December 31, 2011 and 0.05 mg·m^{-3} after January 1, 2012. For new lead and zinc smelters, Hg emissions to air shall be less than 0.05 mg·m^{-3} after October 1, 2010. The emission standard of pollutants for copper, nickel, cobalt industry issued in 2010 (GB 25467-2010) has more strengthened Hg emission limits, 0.012 mg·m^{-3} for existing smelters after January 1, 2011 and for new smelters after October 1, 2010. These regulations have promoted the Hg emission control in China.

China's Hg emission limits for municipal waste incinerators and medical waste incinerators are 0.2 and 0.1 mg·m^{-3}, respectively. These standards are in line with the standard in Europe but much looser than that of the United States, which requires the Hg emissions from existing and new municipal waste incinerators less than 0.0054 and 0.00016 mg·m^{-3}, respectively.

Mercury is also one of the five pollutants targeted in the "12th Five-Year Plan for Heavy Metals Pollution Prevention and Control." By the end of 2015, the Hg emissions shall be reduced by 15% of 2007 levels in the key areas. Mercury emissions in other areas shall be kept in the 2007 levels by 2015. The Minamata Convention on Mercury will control emissions of mercury and mercury compounds into the atmosphere from coal-fired power plants, coal-fired industrial boilers, smelting and roasting processes used in the production of non-ferrous metals (lead, zinc, copper and industrial gold), waste-incineration facilities and cement clinker production facilities. New sources are required to employ "best available techniques" and "best environmental practices" (BAT/BEP) to reduce their mercury emissions to the air. For existing facilities, countries that are parties to this Convention will have to develop a National Plan in which they will employ one or more of five measures, including the BAT to reduce overtime the emissions from these existing facilities. Therefore, to further reduce Hg emissions and comply with the new global mercury Convention, more efforts shall be made to control emissions from coal-fired industrial boilers, lead, zinc, copper and industrial gold smelting and roasting processes, waste-incinerators and cement clinkers.

4.2 Application of mercury emission control technologies

Mercury emission control technologies include fuel/raw

Table 7 China, EU, and US Coal-fired power plant standards

		China	EU	US
Hg	new and existing plants	0.03	0.03 (A German standard only)	new: 0.001 (bituminous, gangue), 0.005 (lignite)
				existing: 0.002 (bituminous, gangue), 0.006 (lignite)
Particle	new and existing plants	30	50, with an exception of 100 for low quality coal (eg lignite)	22.5
SO_2	new plants	100	200	160 (built after 2005)
	existing plants (28 provinces)	200	400	160 (built between 1997 and 2005)
	existing plants (4 provinces with high sulfur coal)	400		640 (built between 1978 and 1996)
NO_x	new plants	100	500 until 12/31/2015, then 200	117
	existing Plants (defined in China as built 1/1/04-12/3/11) (defined in US as built after 2/28/05)	100	500 until 12/31/2015, then 200	117
	existing Plants (defined in China as built before 1/1/04) (defined in US as built before 2/28/05)	200	500 until 12/31/2015, then 200	160 (built between 1997 and 2005)

material pretreatment, technique improvement, co-benefit mercury control, dedicated mercury control, and multi-pollutant control. Control technologies for atmospheric mercury emissions from the main emission sectors were presented as follows:

(1) Coal combustion

Coal treatment technologies consist of conventional coal washing, coal beneficiation for mercury content, coal blending/switching, and coal additives. Conventional coal washing methods will remove some of the mercury associated with the incombustible mineral materials. US EPA quoted the test data for 21 bituminous coal samples which had mercury reductions ranging from 3% to 64% with an average reduction rate of 30% on a mass basis [130]. Advanced coal cleaning techniques, such as the ones using naturally occurring microbes and mild chemical processing, were investigated in the past in order to augment mercury removal [131]. Coal beneficiation is capable of improving coal properties beyond what can be achieved with coal washing alone. An example of coal beneficiation may be the K-Fuel process. The process may also be described as a pre-combustion multi-pollutant control process [132]. The K-fuel process delivered a mercury emission reduction of up to 70% [133]. Bituminous coal typically produces higher fraction of Hg^{2+} in flue gas than subbituminous coal, since Hg^{2+} is water-soluble and more readily captured in WFGD systems [134]. As a result, coal blending has the potential of increasing the mercury capture by about 80% [135]. Vosteen and Lindau [136] tested the performance of bromine-based and chlorine-based additives on mercury removal. The results showed that, for any amount of halogen addition, bromine was more effective in oxidizing mercury than chlorine. Mercury oxidation of 80% could be achieved by adding less than 200 ppm of bromine-based additive.

Co-benefit mercury removal by non-mercury air pollution control equipment can be accomplished in two fundamental modes: removal of Hg^{2+} in WFGD and removal of particulate-bound mercury (Hg_p) in PM control device (ESP or FF). Accordingly, the amount of the co-benefit removal may be augmented by the increase of the Hg^{2+} fraction in the total mercury flue gas concentration or by the improvement of PM control effectiveness [137]. Under certain conditions, SCR catalysts have shown to change mercury speciation by promoting the oxidation of Hg^0 to Hg^{2+}, particularly for bituminous coal. By increasing the amount of Hg^{2+} in the upstream of the WFGD, the SCR could improve the mercury capture in WFGD systems, resulting in the co-benefit removal of mercury [138]. The extent of oxidation of Hg^0 by SCR catalyst and subsequent removal of Hg^{2+} in WFGD may be affected by the chlorine content of coal, the amount of catalyst used to treat the gas stream, the temperature of SCR reaction, the concentration of NH_3 and its distribution in the flue gas, and the age of the catalyst [139]. The mercury removal efficiencies of typical APCD combinations have been discussed in Section 2.1.

Injection of sorbents into the flue gas of coal-fired boilers for mercury control is a typical dedicated mercury control technology and has been demonstrated in the United States on several full-scale systems [140]. Typically, the powdered sorbent is injected upstream of the existing PM control device. Alternatively, sorbent may be injected downstream of an existing ESP and a retrofit FF is then added, which is called the toxic emission control process (TOXECON). The third demonstrated configuration for sorbent injection is TOXECON II in which sorbent is injected into the downstream fields of the existing ESP. Some of the factors that impact the performance of any particular sorbent with regard to mercury capture include the physical and chemical properties of the sorbent, the

injection rate of the sorbent, the flue gas parameters such as temperature, concentrations of HCl, HBr and SO_3, and the existing air pollution control configuration [141]. Multi-pollutant control technologies promise the cost advantage of delivering a system capable of controlling several pollutants simultaneously rather than installing a separate system to address each pollutant separately. The multi-pollutant control technologies that are currently under development include EnviroScrub/Pahlman process, electro-catalytic oxidation (ECO) process, low temperature oxidation (LoTOx) process, and plasma-enhanced ESP (PEESP) process [142–145].

In 2010, 93% of the coal power units were equipped with ESP and the rest 7% with FF. In terms of the SO_2 control measures in the 11th five-year plan, the installation rate of FGD reached 86% in 2010, among which WFGD took up over 95%. NO_x control measures started to be implemented in coal-fired power plants in China, and 14% of the units were equipped with NO_x control devices. Over 95% of them were SCR, and the rest were SNCR. The application rate of coal washing was still low in 2010. The rates for power plants, industrial boilers and domestic boilers were 2.1%, 11.5% and 2.7%, respectively [146–148].

(2) Non-ferrous metal smelting

Mercury emission control technologies for flue gas in non-ferrous metal smelters consist of dust collector (DC), flue gas scrubber (FGS), electrostatic demister (ESD), mercury reclaiming tower (MRT) and conversion and absorption process [25]. Hg_p was mainly removed in DC including cyclone, electrostatic precipitators (ESP) and fabric filters (FF). Most of Hg^{2+} was flushed into waste water in FGS and ESD. A small amount of Hg^0 was converted to Hg^{2+} and further removed, but most of them passed through FGS and ESD. Specific mercury control technology is required in smelters using metal concentrate with high mercury concentration. Boliden-Norzink mercury reclaiming technology was applied in a Chinese smelter. However, high patent fee and disposal installation cost limited its further application. The conversion and absorption process, which consists of single conversion single absorption (SCSA) and double conversion double absorption (DCDA), converted a large amount of Hg^0 to Hg^{2+} and thus had high mercury removal efficiency. The application rates of different APCD combinations were listed in the study of Wu et al. [25]. DC + FGS + ESD + DCDA is the most common type of APCD combinations applied in China, the zinc, lead and copper production percent from plants with which reached 76.31%, 61.58% and 93.15% in 2010, respectively. The type of DC + FGS + ESD + MRT + DCDA was only adopted in zinc smelter. Production percent from zinc plants with MRT reached about 10.09% of total zinc production in 2010. The percentage of metal production from plants without APCDs was 5.47%, 26.65% and 3.36%, respectively, for zinc, lead and copper smelters.

(3) Cement production

More than 85% of cement plants use precalciner process till 2011. Other processes, such as shaft kiln and rotary kiln, only account for small parts of cement production, about 10% and 5%, respectively. For cement plants using precalciner process, the output of mercury consists of flue gas at the kiln tail and head, coal mill and dust collector and clinker. The flue gas is thought to be the main output of mercury. FF is widely used in precalciner cement production, usually at the kiln tail. ESP is installed at the head of kiln. Though only accounting for a very small part of cement production, the shaft kiln and rotary kiln also have dust collector at the emission points because China has increased standard for cement plants on PM since 2000. There is currently few desulfurization devices installed in cement plants, so does SCR/SNCR.

The application percentage of different dust collectors is given by Lei et al. [149]. FF and ESP accounted for about 90% in 2008, among which FF was over 40%. Other dust collector including wet scrubber and cyclone were about 10%. Wet scrubber is thought to be more efficient when oxidized mercury is the main species in flue gas. However, more studies are needed on the accurate percentage of dust collector because of the difference in mercury removal efficiency.

4.3 Perspectives on future mercury emission control in China

The UNEP technical background report [14] established three future scenarios for the global mercury emissions by 2020, including Status Quo (SQ) scenario, Extended Emissions Control (EXEC) scenario and Maximum Feasible Technological Reduction (MFTR) scenario. The SQ scenario assumed that current patterns, practices and uses that result in mercury emissions to air would continue, and economic activity was assumed to increase with emission control practices unchanged; the EXEC scenario assumed economic progress at a rate dependent on the future development of industrial technologies and emission control technologies, and that emission control measures currently implemented in Europe would be implemented around the world; the MFTR scenario assumed implementation of all available solutions, leading to the maximum degree of mercury emission reduction. Under the SQ scenario, the total mercury emissions in China would increase from 632 t to 843 t during 2005–2020. With the emission control measures implemented under the EXEC and MFTR scenarios, the total emissions would reduce to 378 t and 290 t, respectively.

Wang et al. [150] estimated the future mercury emissions from coal-fired power plants in China. With hree coal consumption scenarios (high, medium, and low) and three control strategy scenarios (base, reference, and strict), totally nine scenarios were projected for 2015 and 2020. From the comparison of different scenarios, control

strategies determine the trend of mercury emissions whereas coal consumption mainly has an impact on the speed of the increase or decrease of mercury emissions. Under the most probable scenario, the total mercury emissions from Chinese coal-fired power plants would develop a reversed U-shaped curve and eventually be reduced to 130 t by 2020. Based on the most recent studies, Wang et al. [151] updated the projection for the coal power sector by 2020 and 2030 using a probabilistic emission factor model. Two energy scenarios were projected, namely, reference energy scenario and alternative energy scenario. Three control scenarios, namely, baseline (BAU) scenario, and extended emission control (EEC) scenario, and accelerated control technology (ACT) scenario, were developed in this study. The BAU scenario assumed that the air pollution control would follow the laws and regulations by 2008. The EEC scenario assumed more advanced air pollution control technologies gradually spread out based on the policies implemented after 2008 and those with the potential to be implemented in future. The ACT scenario would speed up the implementation of all of the air pollution control technologies. With the alternative energy scenario, the mercury emissions in BAU, EEC and ACT scenarios would 27%, 56% and 71% lower than that in 2008, respectively. The high growth rate of the installation of FGD and SCR will play an important role during 2008–2020. The increase of specific mercury control technology and FF applications, as well as the widespread application of SCR, has a significant impact from 2020 to 2030. The mercury emissions in the ACT scenario would be 34% lower than that in the EEC scenario, mainly due to further enhancement of the applications of specific mercury control technologies.

5 Conclusions

This paper reviews the current status of atmospheric Hg emissions, pollution and control in China. Atmospheric Hg originates from anthropogenic and natural sources. With about 500–800 t of anthropogenic Hg emissions, China contributes 25%–40% to the global Hg emission inventory. Coal combustion, non-ferrous metal smelting, gold production, cement production and iron and steel production are considered as the dominant Hg emission sources in China. The uncertainties of Hg emissions from coal combustion and non-ferrous metal smelting are about ±40% and ±85%, respectively, while those of Hg emissions from other sources are even larger. The natural Hg emissions in China, with larger uncertainty, are equivalent to the anthropogenic Hg emissions.

The atmospheric mercury concentration in China is about 2–10 times the background level of north hemisphere. The atmospheric Hg concentrations at urban sites are higher than those at rural sites which reveal the regional backgrounds in China. Remote sites have the lowest Hg concentrations which are a little higher than the north hemispheric background level. Dry Hg deposition is higher than wet deposition in urban area in China. Hg deposition fluxes in rural and remote areas in China are usually in the range of 50–200 $\mu g \cdot m^{-2} \cdot yr^{-1}$ and 10–50 $\mu g \cdot m^{-2} \cdot yr^{-1}$, respectively. Wet deposition in the most remote area in Tibet is as low as 1–2 $\mu g \cdot m^{-2} \cdot yr^{-1}$.

Mercury emission control legislations in China currently involve the sectors of power plants, non-ferrous metal smelters and waste incinerators. The Hg emission control technologies in China are so far mostly based on co-benefit from existing air pollution control devices. The particle, SO_2 and NO_x control strategies implemented or to be implemented in coal combustion, non-ferrous metal smelting and cement production in China have considerable Hg removal efficiency. Dedicated Hg control technologies, e.g. activated carbon injection, are required in the future to further improve the Hg abatement in China.

Although many studies have been conducted, current knowledge is not sufficient to fully understand the atmospheric Hg emissions, transport, deposition, pollution and effects in China owing to the lack of accurate mercury emission inventory, synchronous observations of atmospheric Hg, and validated models with state-of-the-art chemistry mechanisms. Therefore, there is a need of greater coordinated study on mercury incorporating emission, modeling, and observation of atmospheric Hg. The large size of various regions, different types of sources, and regional atmospheric compound pollution in China provide a unique opportunity to improve the understanding of the Hg cycle and understanding the role of China as source or sink for Hg on regional and global scales.

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