Atmospheric Environment 162 (2017) 45–54

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Atmospheric emission of mercury due to combustion of steam coal and domestic coal in China

Shaobin Wang, Kunli Luo*

Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China

HIGHLIGHTS

- We quantified Hg emission rates from steam coal, domestic coal combustion and coal gangue.
- Hg emission into the atmosphere is estimated to be 292 tons in China in 2014.
- The trend of Hg emission shows an accelerating growth after 2002.
- Hg emission due to coal utilization and coal gangue should draw more attention in the future.

ARTICLE INFO

Article history:
Received 7 October 2016
Received in revised form
5 May 2017
Accepted 12 May 2017
Available online 12 May 2017

Keywords:
Mercury
Atmospheric emission
Steam coal
Domestic coal-stove
Spontaneous combustion of coal gangue

ABSTRACT

To study the mercury emission due to the combustion of steam coal and domestic coal in China, we analyzed the mercury contents of coal, fly ash, bottom ash and sluicing water in thermal power plants, steam boilers as well as domestic coal-stoves, in Shaanxi, Shanxi, Shandong and Yunnan Provinces. This study conduct an estimate of the Hg emission rates from steam coal and domestic coal combustion based on the method of mass distribution ratio of fly ash and bottom ash. The results show that the Hg emission rate of coal combustion in thermal power plants is about 50.21% (electrostatic precipitators + wet flue gas desulfurization), and that in heating boilers is about 67.23%, and 92.28% in industrial boilers without flue gas desulphurisation equipment. Furthermore, Hg emission rate is 83.61% due to domestic coal combustion in coal-stoves. The Hg emission amount into the atmosphere from power and heat generation, industrial boilers, domestic coal-stoves and spontaneous combustion of coal gangue is roughly estimated to be 133 ± 4, 100 ± 17, 11 ± 0.1 and 47 ± 26 tons in China in 2014, respectively, and the total Hg emission amount from this paper is estimated at 292 tons. The trends of Hg emission in China from 1991 to 2014 show an accelerating growth after 2002. The proportion of mercury emission due to thermal power, heating generation and industrial energy utilization continuously increased. The atmospheric emission of mercury due to combustion of steam coal, domestic coal and coal gangue accounts nearly 50% in total anthropogenic Hg emissions in China, indicating one of the largest sources of Hg emission in China which should draw more public and scientific attention in the future.

1. Introduction

Coal combustion, as an important source of mercury emission caused by anthropogenic activities, attracts global attention for its severe effects on the environment and human health (e.g. Fthenakis et al., 1995; Finkelman et al., 2002; Zhang and Smith, 2007; Zhang and Wong, 2007; Luo et al., 2011; Dai et al., 2012; Liu et al., 2013; Zhao et al., 2015; Wang et al., 2016a,b). Global emission inventories indicate that China is the largest country of anthropogenic mercury emissions (UNEP, 2013a; Pacyna et al., 2010; Pirrone et al., 2010). Additionally, coal combustion is identified as the major source of particulate Hg in China (e.g. Fang et al., 2001; Schleicher et al., 2015; Zhang et al., 2015).

Coal accounts for 65.6% of all energy consumed (coal equivalent calculation) in China in 2014, much more than petroleum, natural gas and hydropower (Editorial Committee of CCEY, 2014). Since 1989, the production and consumption of coal in China have been ranked the first in the world. In China, thermal power generation and heating supply consume the largest part of coal (Editorial

* Corresponding author. Chinese Academy of Sciences, A11 Datun Road, Anwai, Beijing 100101, China.
E-mail address: luokl@igsnrr.ac.cn (K. Luo).

http://dx.doi.org/10.1016/j.atmosenv.2017.05.015
1352-2310/

Committee of CCEY, 2014). Meanwhile, a large number of coal-fired industrial boilers, supplying heat to industrial processes, are widely equipped. What's more, household combustion of raw coal and briquette by domestic coal-stoves are still common for Chinese rural residents (Bonjour et al., 2013). For example, coal as the main fuels accounts for 21.4% in rural households (Duan et al., 2014). Besides coal combustion modes mentioned above, a large amount of coal gangue is discharged during the processing and utilization of coal, which has become one of the largest industrial residues in China (NDRCC, 2014a).

Studies have been done a lot in the assessment of mercury emission from coal combustion in China (e.g. Wang et al., 2000; Zhang et al., 2002; Belkin et al., 2005; Streets et al., 2005; Jiang et al., 2005, 2006; Wang et al., 2006; Wu et al., 2006; Tang et al., 2007; Wang et al., 2010; Chen et al., 2013; Zhou et al., 2013; Zhang et al., 2015). However, most of these studies mainly focus on coalmines or coal-fired power plants based on statistical data analysis and laboratory simulation experiments. At present, it lacks of field investigations on the mercury emission rates of different coal under varied combustion scenarios such as steam coal in power plants, steam boilers, and domestic coal in coal-stoves. Especially, in Chinese rural areas, household coal combustion processes based on raw coal or briquette are aimed at converting fuels directly into heat without dedusting and desulfurization equipment, which caused indoor air pollution of mercury, arsenic and fluorine, etc. (Finkelman et al., 1999; Zhang and Smith, 2007; Li et al., 2012; Liu et al., 2013). Besides coal combustion resources mentioned above, spontaneous combustion of coal gangue, one of the largest industrial residues in China, is considered as a potential source of air pollution of heavy metals (e.g. Zhao et al., 2008; Liang et al., 2016; Wang et al., 2016a,b). However, it still remains poorly known of the mercury emission rate from these domestic coal-stoves in Chinese rural areas based on measured data from field investigations. In addition, it is still obscure about the share of mercury emission from residues of coal processing such as coal gangue piles in the total mercury emission in China.

Therefore, to attain a better understanding of the mercury emission of steam coal and domestic coal under different combustion conditions, we collected representative samples of steam coal, domestic briquette, bottom ash, fly ash and sluicing water from coal-fired power plants, industrial boilers and domestic coal-stoves in Shaanxi, Shanxi, Shandong and Yunnan Provinces in China. In sum, this paper aims to: *i*) study the amount of mercury emission from steam coal (mainly Permo-Carboniferous coal) combustion under dedusting equipment in coal-fired power plants and industrial boilers respectively; *ii*) investigate the mercury emission from domestic coal (mainly early Carboniferous and late Permian coal) in domestic coal-stoves in rural areas of southwestern China; *iii*) provide a rough quantitative estimate of the amount of mercury emission from different combustion conditions based on energy balance sheets instead of consumption amount of coal by different sectors, and conduct a long-term comprehensive evaluation of atmospheric emission of mercury due to coal processing and combustion in China.

2. Material and methods

2.1. Distribution of mercury in Chinese coal and sampling representativeness

Coal depositions in north China account for about 90.3% of its all coal reserves, within which, coal in the North China Plate and Northwest China accounts for 77.4%, while coal in southern China only accounts to 9.6% (Mao and Xu, 1999). The content of mercury in Chinese coal is unevenly distributed as well: mercury content in northwestern and central China is relatively low, but relatively high in the northeastern and southwestern China (Zheng et al., 2007). Ren et al. (2006) calculated mercury content in Chinese coal at the average value of mercury in coal is 0.188 mg/kg, which is close to the average mercury content of coal in U.S. at 0.17 mg/kg (Finkelman, 1993) and higher than mercury Clark value of world coal at 0.10 mg/kg (Ketris and Yudovich, 2009).

The Permo-Carboniferous coal, widely distributed in the North China Plate and constitutes the largest reserve in China, includes mainly bituminous coal and a small quantity of anthracite (Yuan, 1999). Non-coking coal of Permo-Carboniferous bituminous coal is the main steam coal in power plants and industrial boilers. Anthracite of early Carboniferous and late Permian is the main coal for household in southwestern China. Therefore, it is reasonable to select these coal as representative samples of steam coal and domestic coal in China.

2.2. Sampling sites

Shaanxi, Shanxi and Shandong Provinces are the most important coal resource bases in China. Large-sized thermal power plants such as Baqiao Power Plant and Pucheng Power Plant in Shaanxi Province, Xishan Power Plant in Shanxi Province, and Shiheng Power Plant in Shandong Province, were selected in this paper (Fig. 1). Meanwhile, steam boilers in Wangcun Coalmine in Chengcheng County in Shaanxi Province, Duerping, Dongqu and Tunlan Coalmines near Taiyuan City, Shanxi Province are also included in this study (Fig. 1).

In addition, Zhaotong Prefecture in Yunnan Province is a typical area with heating by domestic coal-stoves in southwestern China (Luo et al., 2011). The briquette blended with clay and bottom ash samples are also collected from the domestic coal-stoves in Weixin and Zhenxiong Counties in Zhaotong Prefecture (Fig. 1).

2.3. Determination of mercury contents and quality control

Samples of power generation coal, fly ash and bottom ash are dried until constant weight. Then samples are crushed by agate mills and passed through the 100-mesh-nylon-sieve. Hydride Generation - Atomic Fluorescence Spectrometry (HG-AFS) is an ideal detection technique for mercury determination (Sánchez-Rodas et al., 2010). So mercury in samples of coal, bottom ash and fly ash at dry weight and ash sluicing water is determined in by AFS-820 double channel HG-AFS (Beijing Jitian Instrument Corporation) at the Laboratory of Analytical and Testing Center of the Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences (LATC, IGSNRR, CAS).

The analysis process of coal, bottom ash and fly ash is shown as follows: weigh a sample of 0.2000–0.2500 g and put it into a test tube with cover, add aqua regia (1 mL HNO_3: 3 mL HCl), put the cover in the bottle and mix them up, then place the test tube removing away its cover in a boiler with boiling water for 2 h, shaking every 30 min, add 1 or 2 drops of 5% potassium dichromate solution ($K_2Cr_2O_7$) and dilute it to 25 mL with redistilled water. Mercury is determined in 5% HCl with potassium borohydride (KBH_4) as the reducing agent. Mercury in ash sluicing water of thermal power plants is also determined by HG-AFS based on GB/T 5750 after filtration.

For quality control, standard reference materials [GBW07401 (GSS-1), GBW07403 (GSS-3) and GBW07408 (GSS-8), Chinese Standard Sample Study Center, Chinese Academy of Measurement

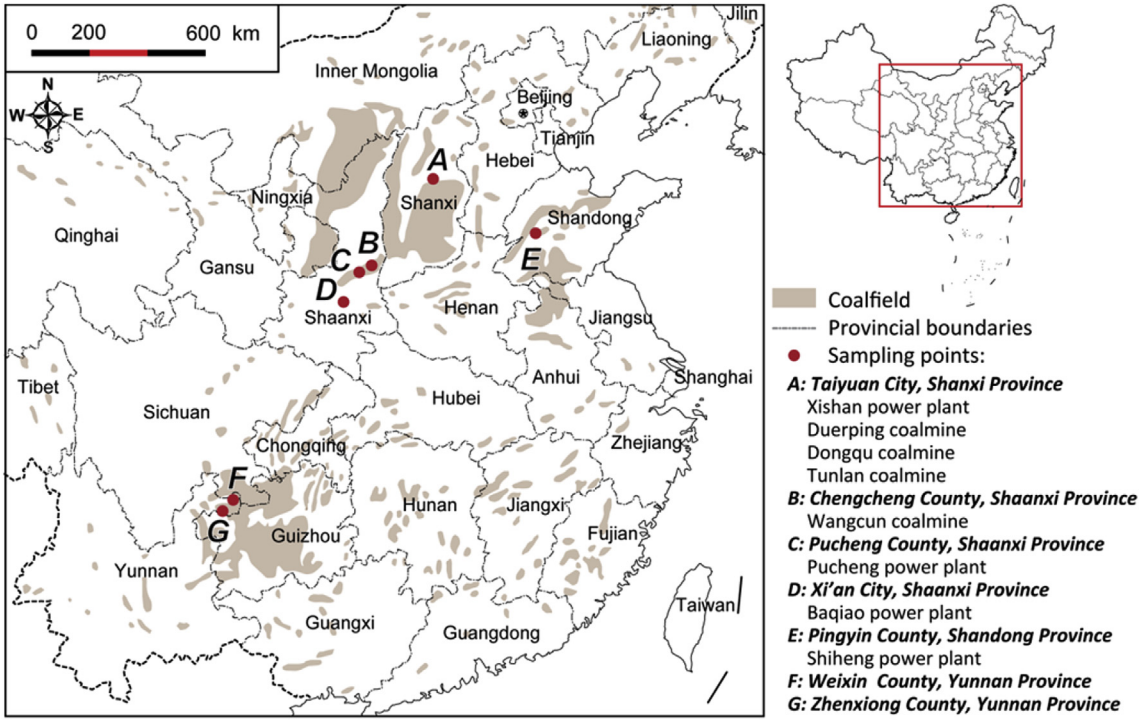

Fig. 1. Geographic setting and location of sampling points (the distribution of Chinese coal fields is after Dai et al., 2012).

Sciences] are randomly analyzed with the respective batch of samples. The correlation coefficient is more than 0.999, and the relative standard deviation was ±5% for mercury contents.

2.4. Calculation of mercury emission rate

Mercury is one of the most volatized trace elements in coal (Stein et al., 1996). Its final transformation matter can be divided into three parts: *i*) remains in coal bottom ash; *ii*) trapped in fly ash by dust catcher and dissolved in sluicing water; *iii*) goes directly into the atmosphere (gas phase and aerosols). To obtain the mercury emission rate from industrial coal-fired boilers, mass balance technique has been widely used by many studies (Wang et al., 2000; Streets et al., 2005; Tang et al., 2007, 2012; Dabrowski et al., 2008). In deed, the mass distribution ratio of fly ash and bottom ash are closely associated with coal burning temperature and other combustion conditions (Luo et al., 2004). 1) In high-temperature boilers under well controlled combustion conditions, the fly ash and bottom ash have similar compositions (mainly SiO_2 and Al_2O_3) and their carbon content is quite low under complete burning. The mass of fly ash and bottom ash accounts for 90% and 10% respectively of all ashes produced. 2) The fly ash and the bottom ash generally account for 60% and 40% of all ashes respectively when coal is burnt at low temperature (Luo et al., 2004). Therefore, we estimate mercury emission from coal combustion by multiplying a coefficient or directly subtract mercury content in each ton of coal ash from that in each ton of coal.

2.4.1. Mercury emission rate in high-temperature boilers of thermal power plants

In fact, the ash content of coal is defined as the amount of residuals after complete combustion. Mercury emission from coal combustion (E) can be expressed as follows:

$$E = C_{\text{coal}} - (C_{\text{bottom ash}} \times f_{\text{bottom ash}} + C_{\text{fly ash}} \times f_{\text{fly ash}}) \quad (1)$$

in Eq (1), C_{coal}, $C_{\text{bottom ash}}$ and $C_{\text{fly ash}}$ are the mercury concentrations of coal, bottom ash and fly ash, respectively, and f is the mass fractions of bottom ash or fly ash relative to raw coal.

The furnace hearth temperature is high in the large-sized power plant, so coal is burnt more completely and the fly ash plus bottom ash quantity is approximately equal to the ash content of coal. For the large-sized power plant with good combustion condition and high-temperature furnace hearth, the mercury emission rate (R_1) from coal combustion based on the following Equation (2):

$$R_1 = \frac{C_{coal} - \left(C_{bottom\ \ ash} \times 10\% + C_{fly\ \ ash} \times 90\%\right) \times Ash}{C_{coal}} \times 100\% \quad (2)$$

2.4.2. Mercury emission rate in steam boilers and domestic coal-stoves

In general, the hearth temperature of regional boilers for hot water supply varies from 800 to 1200 °C. As a result, coal burnt incompletely and about 5–20% of coal remains in fly ash and bottom ash (Luo et al., 2004). Therefore, for mid-low temperature power plants and hot water boilers, the total amount of fly ash and bottom ash is more than the ash content in coal, which should be equal to the ash content in coal plus the coal remaining in the fly ash and bottom ash. For regional boilers for hot water supply, we can calculate the mercury emission rate (R_2) based on the following Equation (3):

$$R_2 = \frac{C_{coal} - \left(C_{bottom\ \ ash} \times 40\% + C_{fly\ \ ash} \times 60\%\right) \times Ash - C_{coal} \times M}{C_{coal}} \times 100\% \quad (3)$$

where M is used to express the percent of coal remaining in the fly ash and bottom ash, and it is considered at 7% for regional boilers.

The temperature in the majority of domestic coal-stoves is quite low. Coal combustion residue is mainly bottom ash, and there is more un-burnt coal remaining in coal bottom ash. The carbon content in bottom ash (M) is about 15% in domestic coal-stoves, where the hearth temperature is about 700–850 °C (Luo et al., 2004).

2.5. Long-term data collections and uncertainty analysis

We collect the data of total coal consumption in China, the growth of coal consumption of thermal power, heat generation, industrial (for energy use) and residential utilization from 1991 to 2014 (China Energy Statistical Yearbook of 1997–1999, 2005, 2013, 2014, 2015). In addition, energy balance sheets, based on the physical energy content method, can reflect the entire energy throughput of one country's energy structure, and it can also represent different energy flow including the entire process from production to consumption (IEA, 2005). So we conduct an estimation of long-term emission trends of Hg due to coal combustion based on energy input with standard quantity from Chinese energy balance sheets (China Energy Statistical Yearbook of 1997–1999, 2005, 2013, 2014, 2015). Furthermore, coal gangue production in China from 1992 to 2014 is also collected (Li and Sun, 2007; CNCA, 2016; NBSC, 2013a,b, 2014; NDRCC, 2012, 2014a,b).

Additionally, we estimate the uncertainty for each emitting sector such as steam coal, domestic coal and coal gangue by combining the coefficients of variation (relative standard deviation, RSD) of the contributing factors (Wu et al., 2006). We then combine these uncertainties to estimate the total uncertainty of Hg emission estimates.

3. Results and discussions

3.1. Mercury emission rates of steam coal combustion in coal-fired power plants and heating boilers

The average rate of mercury emission from coal combustion is calculated at 92.28% (before dedusting and desulfurization) in high-temperature power plants (Table 1). Xishan Power Plant is both equipped with wet FGD (flue gas desulfurization) and ESP (electrostatic precipitators). The mercury emission rate by the former method is 83.65%, while the rate of the latter is 60.02%, which indicates that ESP is a more effective mercury control method than water film scrubber (Table 2). The emission rate by wet FGD is based on the ratio of sluicing water: fly ash at 20:1, which means that 20 ton of sluicing water can flush 1 ton of fly ash. In addition, the average rate of mercury emission from steam boilers is 67.23% (Table 3), which is higher than mercury emission rate of power plants.

The same kind of coal (clean coal from Chenghe and the Pubai Mining Bureau, Shaanxi Province), which have mercury content of 0.198 mg/kg, was used as steam coal in both Pucheng Power Plant and Wangcun Coalmine (Tables 3 and 4). But in Pucheng Power Plant, the carbon content and mercury content in fly ash (about 0.066 mg/kg) were much lower than those of Wangcun Coalmine (about 0.48 mg/kg).

3.2. Mercury emission rates of domestic coal combustion in coal-stoves

The domestic coal consumed by local rural residents are mainly #5 and #6 coal in Longtan Formation of late Permian in Zhenxiong and Weixin counties in Zhaotong Prefecture, Yunnan Province (Luo et al., 2008). The average content of these domestic coal is 0.099 ± 0.048 mg/kg. Although the variation range of mercury contents and ash contents in briquette is quite large, the rates of mercury emission have not changed distinctly. The rates of mercury emission are 80.77–84.50% (the average rate at 83.61%) from domestic coal combustion in coal-stoves (Table 4).

The relation between mercury emission rates and mercury/ash contents in briquette is discussed in this paper (Fig. 2). Regression analysis shows that: i) logarithm regression between mercury contents in briquette (μg/kg) and mercury emission rate (%), which indicates quite low mercury emission rates are associated with mercury contents about 50 μg/kg, while increased mercury emission rates had little relation with higher contents of mercury; ii) binomial regression between ash contents (%) and mercury emission rate (%), which indicates higher mercury emission rates at lower ash contents around 40% while lower mercury emission rates are associated with the increased ash contents (Fig. 2). It indicated that inorganic salts and oxides in coal are more conducive to catching mercury during the coal combustion process. It may furtherly indicate a possible way to reduce mercury emission from

Table 1
Mercury emission per ton of coal combustion in thermal power plants (before dedusting and desulfurization).

Sampling points	Coal no.	Average mercury in coal ($g \cdot t^{-1}$)	Mercury in bottom ash ($g \cdot t^{-1}$)	Mercury in fly ash ($g \cdot t^{-1}$)	Mercury to atmosphere ($g \cdot t^{-1}$)	Ash content (%)	Mercury emission rate (%)
Pucheng Power Plant	5#	0.198	0.054	0.066	0.184	22.23	92.72
Baqiao Power Plant	5#	0.183	0.060	0.073	0.169	20.01	92.16
Shiheng Power Plant	5#	0.163	0.050	0.068	0.148	23.12	90.61
Xishan Power Plant	8#	0.311	0.092	0.099	0.157	20.16	93.63
Average							92.28

Table 2
Mercury emission per ton of coal combustion in Xishan thermal power plants (after dedusting and desulfurization).

	Mercury in bottom ash (g ·t^{-1})	Mercury in fly ash (g·t^{-1})	Mercury in ash sluicing water (mg/L)	Mercury to atmosphere (g·t^{-1})	Mercury emission rate (%)
wet FGD	0.092	0.223	0.002	0.260	83.65
ESP	0.092	0.675	–	0.187	60.02

Table 3
Mercury emission per ton of coal combustion in steam boilers in Shanxi Province.

Sampling points	Coal no.	Average mercury in coal (g·t^{-1})	Mercury in bottom ash (B) (g·t^{-1})	Mercury in fly ash (C) (g·t^{-1})	Mercury to atmosphere (g·t^{-1})	Ash content (%)	Mercury emission rat (%)
Wangcun	5#	0.198	0.130	0.480	0.109	10.85	74.41
Duerping	8#	0.177	0.021	0.339	0.247	22.51	66.06
Dongqu	8#	0.177	0.087	0.295	0.247	20.76	68.16
Tunlan	8#	0.177	0.137	0.528	0.214	15.59	60.27
Average							67.23

Table 4
Mercury emission from coal combustion in coal-stoves in Zhaotong Prefecture, Yunnan Province.

Sampling points		Ash content (%)	Mercury content in briquette (g·t^{-1})	Mercury in bottom ash (g·t^{-1})	Mercury to atmosphere (g·t^{-1})	Mercury emission rate (%)
Weixin county	Sample 1	37.83	0.0749	0.0073	0.0626	83.53
	Sample 2	36.37	0.0575	0.0064	0.0480	83.39
	Sample 3	31.52	0.0285	0.0049	0.0236	82.83
	Sample 4	43.59	0.0766	0.0047	0.0643	83.93
	Sample 5	36.02	0.0730	0.0059	0.0612	83.84
	Sample 6	56.65	0.0641	0.0058	0.0531	82.94
	Sample 7	42.04	0.0740	0.0045	0.0622	83.98
	Sample 8	42.18	0.0961	0.0051	0.0809	84.11
	Sample 9	41.12	0.1729	0.0053	0.1461	84.50
	Sample 10	56.12	0.0734	0.0054	0.0612	83.35
	Sample 11	32.41	0.1508	0.0058	0.1274	84.50
	Sample 12	59.39	0.0577	0.0082	0.0471	81.62
	Sample 13	45.53	0.0940	0.0070	0.0786	83.64
	Sample 14	48.33	0.1216	0.0117	0.1011	83.14
	Sample 15	37.23	0.1047	0.0066	0.0881	84.06
	Sample 16	36.08	0.1215	0.0111	0.1017	83.69
	Sample 17	39.56	0.0880	0.0069	0.0738	83.76
Zhenxiong county	Sample 1	41.19	0.0799	0.0071	0.0668	83.54
	Sample 2	56.79	0.0602	0.0112	0.0487	80.77
	Sample 3	49.72	0.0527	0.0060	0.0436	82.75
	Sample 4	43.64	0.0865	0.0032	0.0729	84.35
	Sample 5	45.23	0.2307	0.0067	0.1949	84.48
	Sample 6	37.66	0.0767	0.0027	0.0648	84.46
	Sample 7	36.60	0.1518	0.0067	0.1280	84.35
	Sample 8	36.22	0.2029	0.0057	0.1717	84.60
Average						83.61

domestic coal combustion in rural area.

3.3. Atmospheric mercury emission due to combustion of steam coal, domestic coal and spontaneous combustion of coal gangue in China

Based on Chinese energy balance sheet with standard quantity, about 1219 and 143 million tons of standard coal are burnt in China in 2014 as thermal power and heating fuel, respectively (China Electric Power Yearbook, 2015). The total mercury emission rate in thermal power plants is calculated at 50.21% (ESP + wet FGD), which is quite closed to the average Hg removal efficiency of about 49% in coal-fired power plants (Zhou et al., 2013). Therefore, the annual mercury emission from power to the atmosphere is about 115.08 tons and 18.13 tons for heating, respectively. Also, about 582 million tons of coal are used as industrial energy in China in 2014 (China Electric Power Yearbook, 2015). It still lacks of efficient flue gas desulphurisation equipment in small industrial boilers (<10 t/h) (NDRCC, 2014b). Accordingly, it is about 100.93 tons from industrial boilers as the mercury emission rate is 92.28%. Additionally, about 69 million tons of standard coal are burnt in China in 2014 for residential consumption (China Energy Statistical

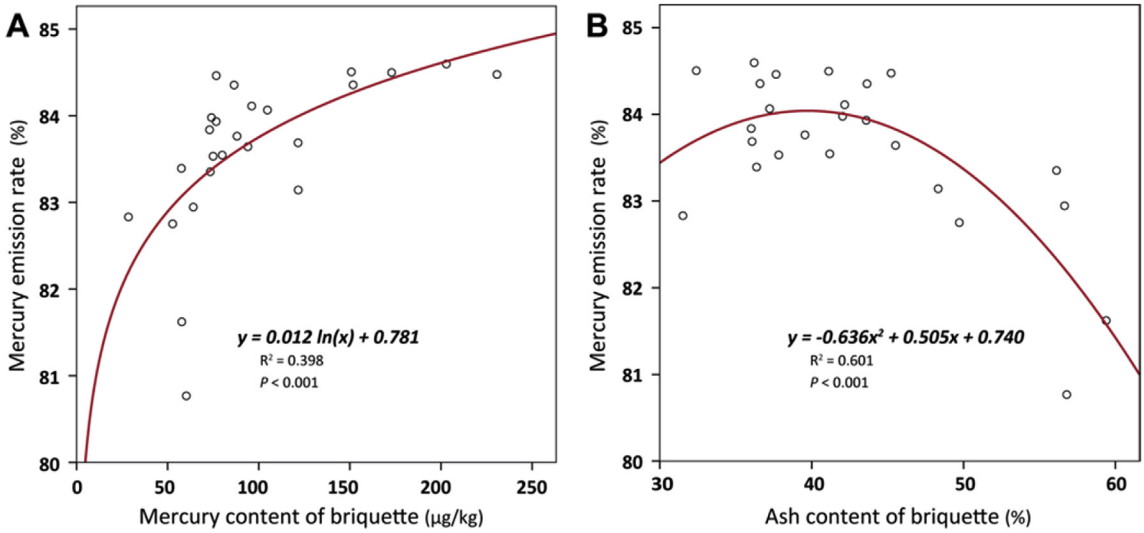

Fig. 2. Regression analysis between mercury emission rate and properties of briquette in domestic coal-stoves in Zhaotong Prefecture, Yunnan Province. A) logarithm regression between mercury content in briquette (μg/kg) and mercury emission rate (%); B) binomial regression between ash contents (%) and mercury emission rate (%).

Yearbook, 2015). Hence the annual mercury emission from domestic coals to the atmosphere is about 10.82 tons as the mercury emission rate is 83.61%. To sum up, the total annual mercury emission to the atmosphere from steam coal combustion is about 245 tons in China, 2014 (Table 5). The atmospheric emission of mercury from coal gangue spontaneous combustion in 2014 is calculated at 47.44 tons, based on the method from Wang et al. (2016a,b). So, the total Hg emission amount due to steam coal, domestic coal and coal gangue is estimated at 292 tons from this paper (Table 5).

Atmospheric emissions of mercury due to coal combustion in China are established by previous researches (Table 5). Based on these results, the calculated Hg emission due to coal combustion and power and heat generation, industrial boilers exhibit an increasing trend from 1995 to 2014 despite quite large difference (Table 5). However, our estimate results are lower than the previous calculation such as Zhao et al. (2015). It may be attributed to the different statistical definition of coal utilization. Previous researches mainly used total consumption amount of coal by different sectors. It should be noted that not all coal is used as energy by way of burning, and some of raw coal are used as a feed coal in production of a wide range of other chemical products.

Based on the calculation based on energy balance sheet in China in 2014, the largest anthropogenic Hg emission source due to coal combustion is considered as thermal power generation, followed by industrial boilers, heating boilers and domestic stoves (Table 5). Additionally, emission of mercury due to spontaneous combustion of coal gangue in China in 2014 reached to 47 tons, which is nearly one fifth of total mercury emission due to coal combustion, based on the calculation method from Wang et al. (2016a,b).

3.4. Long-term emission trends of Hg due to coal processing and combustion in China (1991–2014)

Coal consumption in China by thermal electric and industrial boilers increased obviously from 1991 to 2014 (Fig. 3A). Moreover, production of coal gangue increased with continuous growth of coal mining and coal cleaning. Coal consumption of heating shows increasing trend despite small total amount. While, coal for residential consumption shows fluctuation but persistent decline trend from 1991 to 2014 (Fig. 3A).

Emission trend of mercury due to combustion of steam coal, domestic coal and spontaneous combustion of coal gangue over the twenty-four-year study period (1991–2014) in China is examined in this paper based on the calculated emission rates (Tables 2–4) and Chinese energy balance sheet. The trend shows an accelerated growth after 2002, which is consistent with the fast growth of coal production in China (Fig. 3B). The average annual growth rate of mercury emission from 1991 to 2001 is about 2.56%, while the rate from 2002 to 2014 is about 8.14%. However, the emission exhibited a slow down trend during recent years, for that the emission in 2014 was even slightly lower than that in 2013. This trend may be

Table 5
Estimation of total emission of mercury into atmosphere from coal combustion and processing in China (tn).

Year for calculation	Total amount	Power generation	Heating boilers	Industrial boilers	Domestic coals	Spontaneous combustion of coal gangue	Reference
1995	213.8	72.86	–	–	19.05	–	Wang et al., 2000
1999	202.36	67.97	–	103.2	19.70	–	Streets et al., 2005
2000	161.6/219.5	56.6/76.8	74.3/101	–	22.6/30.7	–	Jiang et al., 2005
2003	256.7	100.1	124.3	–	21.7	–	Wu et al., 2006
2005	–	108.6	–	–	–	–	UNEP, 2011
2007	–	132	–	–	–	–	Tian et al., 2011
2009	–	172	–	–	–	–	Chen et al., 2013
2010	253.8	100.0	119.7		20.7		Zhang et al., 2015
2012	363.8	178.1	22.2	–	38.5	–	Zhao et al., 2015
2013	–	–	–	–	–	45	Wang et al., 2016a,b
2014	*292.4*	*115.08*	*18.13*	*100.93*	*10.82*	*47.44*	*This work*

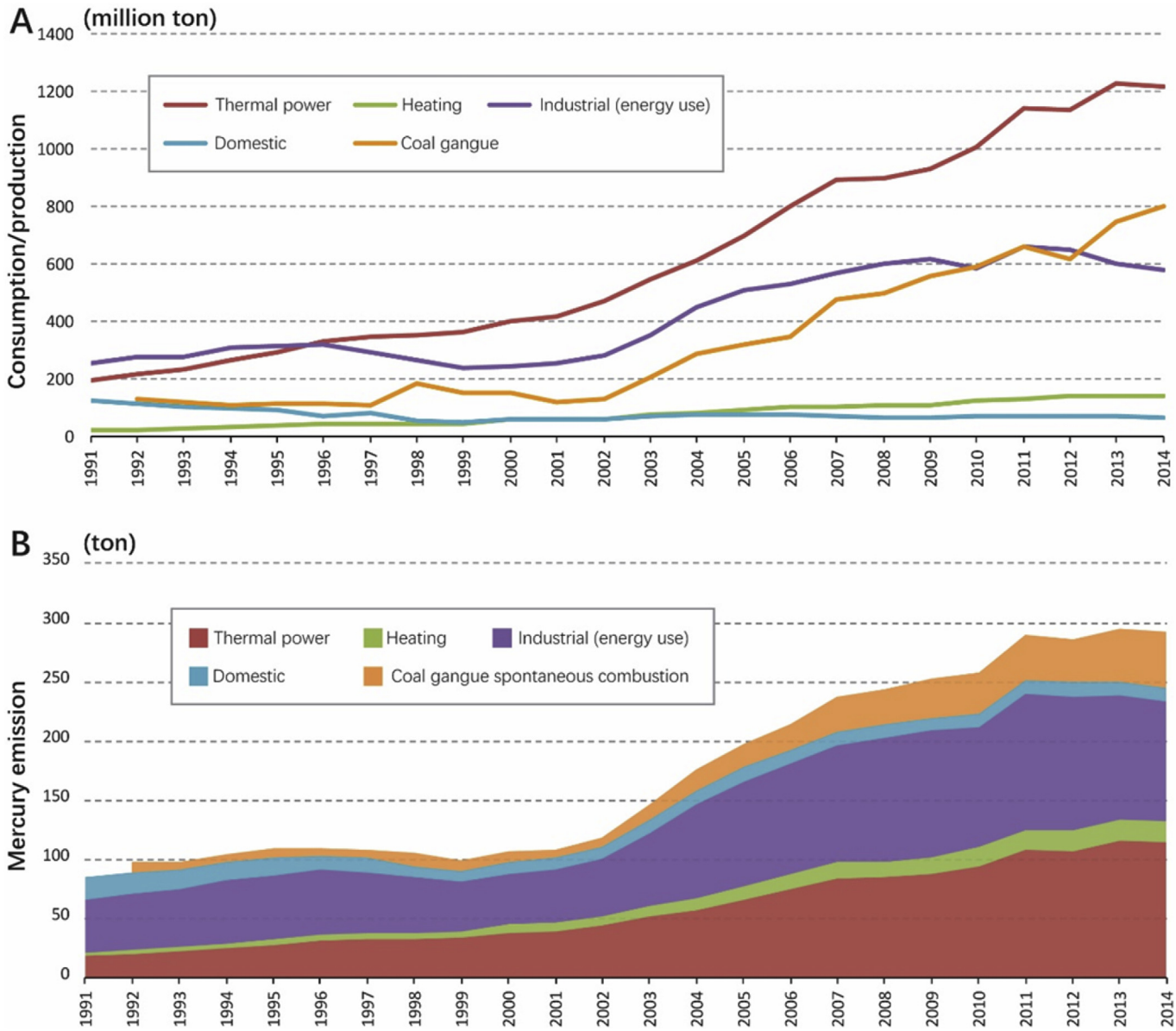

Fig. 3. Growth of coal consumption and mercury emission due to coal combustion (consumed by power generation, heating, industrial and residential utilization) and spontaneous combustion of coal gangue in China from 1991 to 2014. Original data of coal consumption is from China Energy Statistical Yearbook of 1997–1999, 2005, 2013, 2014, 2015.

attributed to the decline of coal consumption in industry and thermal power in recent years.

The amount of Hg emission due to thermal power, heat generation and industrial consumption continuously increase from 1980 to 2012 (Fig. 3B), and the proportion of the industrial utilization and thermal power became the largest Hg emission sources. In comparison, the proportion due to residential utilization keeps decreasing trend. The ratios of Hg emission from residential consumption in total Hg emission from coal combustion declined from more than 20% in 1991 to 4.42% in 2014 (Fig. 3B). This trend may be attributed to the change of decentralized heating into central heating by Chinese urban residents and development of clean coal technology in main Chinese cities (Lu et al., 2008). However, Hg emission due to spontaneous combustion of coal gangue has considerably increased after 2002, and it reached to 800 million tons in 2014 (Fig. 3B) with a continuous growth of coal mining and raw coal washing rate (CNCA, 2016).

3.5. Estimation validity and uncertainties

Several studies have attempted to discuss China's mercury emission strength by field analysis (e.g. Tang et al., 2007; Wang et al., 2010). We select typical calculation results of mercury emission factors based on field measurements in China to identify the validity of the estimation in this paper. The total mercury concentrations in the flue gas from boilers in coal-fired power plants range from 1.92 to 27.15 μg/m^3, which were significantly related to the mercury contents in steam coal, and 19–72% of Hg in flue gas is emitted to the atmosphere (Wang et al., 2010). This result is consistent with the calculation of mercury emission rate in thermal power plants in this paper.

The uncertainties in Hg emissions due to coal combustion in China in 2014 are listed in Table 6, indicated by a 95% confidence interval of the mean value. In 2014, we calculate uncertainty levels in the estimate of emissions due to combustion of steam coal and

Table 6
Uncertainties in Hg emissions due to coal combustion in China in 2014.

Emission sources	Power and heat generation	Industrial boilers	Domestic coals in coal-stoves	Spontaneous combustion of coal gangue
Uncertainties	±3.06%	±16.89%	±0.88%	±55.56%

domestic coal are ±19.95% and ±0.88%, or ±46 tons and ±0.1 tons respectively. Meanwhile, the uncertainty level of emissions due to spontaneous combustion of coal gangue is calculated at ±55.56% in the estimate of, or ±26 tons.

Many uncertainties remain in our knowledge of anthropogenic releases of mercury to the atmosphere in China. For example, the mercury emission uncertainty due to spontaneous combustion of coal gangue is quite large, for the mercury content of coal gangue and emission factor lack of in-depth study. Many of the activities that release large amounts of mercury are in remote areas in China without detailed statistical data as well. Furthermore, the coal self-ignite or coal mine fire is also a serious problem in China (Lu and Qin, 2015), but this combustion process is not included in this paper.

3.6. Policy implications

UNEP (2013a,b) indicates that east-southeastern Asia is responsible for about 40% of global anthropogenic mercury emissions, and about 75% come from China, which is about one-third of the total global amount. Global anthropogenic emission of mercury was 1960 tons in 2010, in which coal burning reached to 474 tons (UNEP, 2013a). The Hg emissions to the atmosphere from Chinese anthropogenic sources has been estimated to be 500–700 tons per year (Fu et al., 2012), which accounts for a quite large proportion of the global anthropogenic Hg emissions. The atmospheric emission of mercury due to combustion of steam coal and domestic coal accounts nearly 50% in total anthropogenic Hg emissions in China, and it contributes more than half of global anthropogenic emission due to coal burning. The estimate result in this paper is equivalent to the total Hg emission from metal and building material production (Zhang et al., 2015), which indicates that emission of mercury due to combustion of steam coal and domestic coal is one of the largest sources of Hg emission in China.

Although mercury content in coal is quite low, due to huge consumption of the power plants, industrial boilers and local residents, its contribution to global mercury emission cannot be ignored. *The Minamata Convention on Mercury*, launched in 2013 by UNEP, requires control of various large-scale coal-fired utility boilers and industrial boilers mercury emissions (UNEP, 2013b). Dedusting and pollutant control equipment in thermal power plants are widely used, while no particular technology has been applied to Hg emission control in China (Zhou et al., 2013). To further decrease the Hg emissions from combustion of steam coal, strong measures and innovative technologies such as modified fly ash sorbent (Wang et al., 2016a,b) and developed hybrid filter (Sung et al., 2017), etc. have important application prospects, under the background of coal as the dominant energy in China in the near future (Lu et al., 2008).

Additionally, compared to controlled combustion process, there is not any effective dedusting and desulfurization equipment in domestic coal-stoves in Chinese rural areas, and most of mercury are released directly into the atmosphere except a few remaining in the bottom ash based on our analysis above (Table 4). With the development of central heating system and clean coal technology in Chinese urban areas, the ratio and amount of mercury emission from residential utilization show a declining trend, but it still exhibits higher Hg emission rate. Fu et al. (2009, 2012) identified that coal and biomass consumption are important sources of the high level of total gaseous mercury during winter in southwestern China. Despite the serious indoor air pollution due to domestic coal in rural China was assessed by some studies (e.g. Finkelman et al., 1999; Zhang and Smith, 2007; Li et al., 2011, 2012; Chowdhury et al., 2013; Liu et al., 2013; Li et al., 2017), mercury emission and health effects due to rural coal utilization is urgently needed as well, which should draw more public and scientific attention in the future. In deed, Hg emission control is more difficult in Chinese rural areas than urban areas in actual policy scenarios, for the decentralized combustion of domestic coal. So, more field investigations pollution evaluation and in-depth control technology research will be needed for better understanding of Hg emission and control.

Last, mercury emission due to spontaneous combustion of coal gangue is considered as an important part of emission inventories in China (Wang et al., 2016a,b). This emission amount which is calculated as (47 ± 26) tons accounts for nearly one fifth of total emission from coal combustion. It indicates the importance of mercury emission control research and policy from coal mining waste such as coal gangue in China.

4. Conclusion

In this paper, we presented an estimate of the mercury emission rates from steam coal/domestic coal combustion and spontaneous combustion of coal gangue in China. It shows that the mercury emission rate of coal combustion in thermal power plants is about 50.21% with ESP + wet FGD, and that in heating boilers is about 67.23%, and 92.28% in industrial boilers without flue gas desulphurisation equipment. Furthermore, the rate of mercury emission is 83.61% from domestic coal combustion in coal-stoves.

The Hg emission amount into the atmosphere from power and heat generation, industrial boilers, domestic coal-stoves and spontaneous combustion of coal gangue is roughly estimated to be 133 ± 4, 100 ± 17, 11 ± 0.1 and 47 ± 26 tons in China in 2014, respectively, and the total Hg emission amount from this paper is estimated at 292 tons.

The trends of mercury emission due to combustion of steam coal, domestic coal and spontaneous combustion of coal gangue in China from 1991 to 2014 shows an accelerating growth after 2002. The proportion of mercury emission due to thermal power, heating generation and industrial energy utilization continuously increase, while the proportion due to residential utilization keeps decreasing trend. Meanwhile, Hg emission due to spontaneous combustion of coal gangue has considerably increased after 2002 as well with continuous growth of coal mining and development of clean coal technology.

In sum, the atmospheric emission of mercury due to combustion of steam coal, domestic coal and coal gangue accounts nearly 50% in total anthropogenic Hg emissions in China, indicating one of the largest sources of Hg emission in China which should draw more public and scientific attention in the future.

Acknowledgements

Financial supports for this research were provided by the National Basic Research Program of China (973 Program) (Grant no.

2014CB238906) and the National Natural Sciences Foundation of China (Grant no. 41472322 and 41172310). We thank Prof. Y. C. Zhao for reviews that improved the manuscript considerably, as well as Dr. C. Zhao for providing helps with data collecting and language editing.

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