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Mercury speciation and emissions from coal combustion in Guiyang, southwest China

Shunlin Tang[a,b,c,*], Xinbin Feng[a], Jianrong Qiu[b], Guoxun Yin[c], Zaichan Yang[c]

[a]State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Science, Guiyang 550002, PR China
[b]State Key Laboratory of Coal Combustion, Huazhong University of Science & Technology, Wuhan 430074, PR China
[c]Resource and Environment Academy, Henan Polytechnic University, Jiaozuo 454000, PR China

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Abstract

Although China has been regarded as one of the largest anthropogenic mercury emission source with coal combustion, so far the actual measurements of Hg species and Hg emissions from the combustion and the capture of Hg in Chinese emission control devices were very limited. Aiming at Hg mercury species measurements in Guiyang, the capital city of Guizhou province in Southwest China, we studied flue gases of medium-to-small-sized industrial steam coal-firing boiler (10–30 t/h) with no control devices, medium-to-small-sized industrial steam coal-firing boiler with WFGD and large-scale coal combustion with ESPs using Ontario Hytro method. We obtained mercury emission factors of the three representative coal combustion and estimated mercury emissions in Guiyang in 2003, as well as the whole province from 1986 to 2002. Coal combustion in Guiyang emitted 1898 kg mercury to the atmosphere, of which 36% Hg is released from power plants, 41% from industrial coal combustion, and 23% from domestic users, and 267 kg is Hg^p, 813 kg is Hg^{2+} and 817 kg is Hg^0. Mercury emission in Guizhou province increased sharply from 5.8 t in 1986 to 16.4 t in 2002. With the implementation of national economic strategy of China's Western Development, the annual mercury emission from coal combustion in the province is estimated to be about 32 t in 2015.

Keywords: Mercury species; Emission factors; Coal combustion; Guiyang; China

1. Introduction

Hg is released into the atmosphere from both natural and anthropogenic sources (Lindqvist et al., 1991; Pacyna et al., 2003; Gustin et al., 1999, Feng et al., 2004) and has gained worldwide attention due to its high toxicity, bio-accumulation, and difficulties to control. The emitted mercury could be transported globally, even to regions without significant mercury emission sources. The Arctic, for example, is reported to be affected by the transcontinental mercury transportation (Fitzgerald, 1995; Seigneur et al., 2004).

China has been regarded as one of the largest anthropogenic mercury emission sources with coal combustion in particular. Seigneur et al. (2004) estimated that anthropogenic emissions of mercury in Asia contributed 21% to the total mercury deposition in the contiguous United States in 1998. Dastoor and Larocque (2004) estimated that China and Japan contributed 28% to the total global anthropogenic emissions of mercury in 1990. Pacyna and Pacyna (2002) estimated that China's emissions from coal combustion contributed more than 25% to the total global emissions. Before 2005, there is only one estimate of mercury emissions from coal combustion in China made by Chinese researchers, which has been reported in three papers (Wang et al., 1999, 2000; Zhang et al., 2002). In recent years, more attention has been given to mercury emissions in China. Streets et al. (2005) estimated Hg emissions from China in 1999 where 537 ($\pm$236) t, and 38% of the Hg comes from coal combustion. Wu et al. (2006) estimated Hg emissions from coal combustion increased from 202 t in 1995 to 257 t in 2003 at an average

*Corresponding author. State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Science, Guiyang 550002, PR China.
E-mail address: tangshunlin@vip.gyig.ac.cn (S. Tang).

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annual rate of 3.0%. Jiang et al. (2006) reviewed mercury pollution in China including mercury emissions from coal combustion. However, all the estimates were inadequate due to the lack of direct measurements of mercury species, concentration in flue gases and capture of Hg in emission control devices in China.

Mercury released from natural sources is believed to be mainly the elemental mercury (Hg^0), whereas anthropogenically emitted mercury has a significant portion in gaseous oxidized mercury (Hg^{2+}) and particulate-phase mercury (Hg^p) depending on the specifically conditions (Schroeder and Munthe, 1998). Based on the current global atmospheric burden, it is estimated that about 50% of mercury in the atmosphere was emitted from anthropogenic sources (Lamborg et al., 2002; Mason and Sheu, 2002). Coal combustion is one of the largest anthropogenic mercury emission sources (Mason et al., 1994). It has been demonstrated that about 34% of Hg emitted annually on global scale comes from coal combustion (Prestbo and Bloom, 1995).

The identification and quantification of individual Hg species, at least the three species (Hg^0, Hg^{2+}, Hg^p) in coal combustion flue gases, are imperative to understand problems concerning Hg emission control, toxicity, mobility, bioaccumulation, and atmospheric transportation because different species have distinctive physical, chemical, and biological properties. For example, it has been shown that the ability of scrubbers and particulate control devices on mercury removal from flue gas is highly dependent upon the mercury species. Elemental mercury (Hg^0) is volatile, relatively inert, and virtually insoluble. As a result, it can pass through conventional scrubbers and particulate control devices and contributes to the global mercury emissions inventory. In contrast, oxidized mercury (Hg^{2+}) is water-soluble, thus more apt to react with particles to form particulate mercury, and part of Hg^{2+} can be captured in scrubbers while the elemental mercury may be transported a few hundred kilometers away from the source. Particulate-phase mercury (Hg^p) is likely to deposit at an intermediate distance depending on the aerosol diameter. Therefore, to effectively control the emission of mercury from coal combustions, the insights of mercury species in flue gas are of great importance.

This study aims to (1) determine the Hg species in flue gases from two representative coal combustion systems, (2) to study the Hg species emission factors and resulting emissions, (3) to estimate the coal combustion mercury emissions in Guiyang, the provincial capital city of Guizhou province, Southwest China in 2003 and the whole province from 1986 to 2002.

2. Method

2.1. Site description

The provincial capital Guiyang is regarded as one of the most seriously polluted cities in China. It consists of six districts (Yunyan, Nanming, Xiaohe, Huaxi, Wudang, Baiyun), one sattelite city (Qingzhen), and three counties (Xiuwen, Xifeng, Kaiyang) (Fig. 1), and has a typical subtropical humid monsoon climate with an annual average temperature of 15 °C and an annual precipitation of 1100–1400 mm. Acid rain from coal combustion pollution is the main environmental burden. According to Statistical Bureau of Guizhou Province (1995), the daily average concentrations of SO_2 and total suspended particulate matter (TSP) in the air of Guiyang in 1992 were 468 and 395 $\mu g/m^3$, respectively, both exceeding the national air quality standards of, i.e. 100 and 300 $\mu g/m^3$ for SO_2 and TSP, respectively.

Mercury emission rates and the speciation profiles depend greatly on the applied combustion technologies. Coal in Guiyang is mainly burned in three types of combustion systems. The most common is power plants. There are two large-scale coal-fired power plants with ESPs in Guiyang: Guiyang Coal-fired Power Plant (GCPP) and Qingzhen Coal-fired Power Plant (QCPP), which consume 34% of annual coal supply. The second type is hundreds of medium-to-small-sized industrial steam (10–30 t/h) coal combustion boilers, of which, only a few of them have flue gas control devices; they consume 44% of the annually supplied coal. Coal consumption in these two types of coal combustion is relatively consistent over the year. The third type is domestic users, which consume 22% of the coal supplied annually, and the coal consumption changes in different seasons, e.g. winter is the main coal consumption season of domestic users.

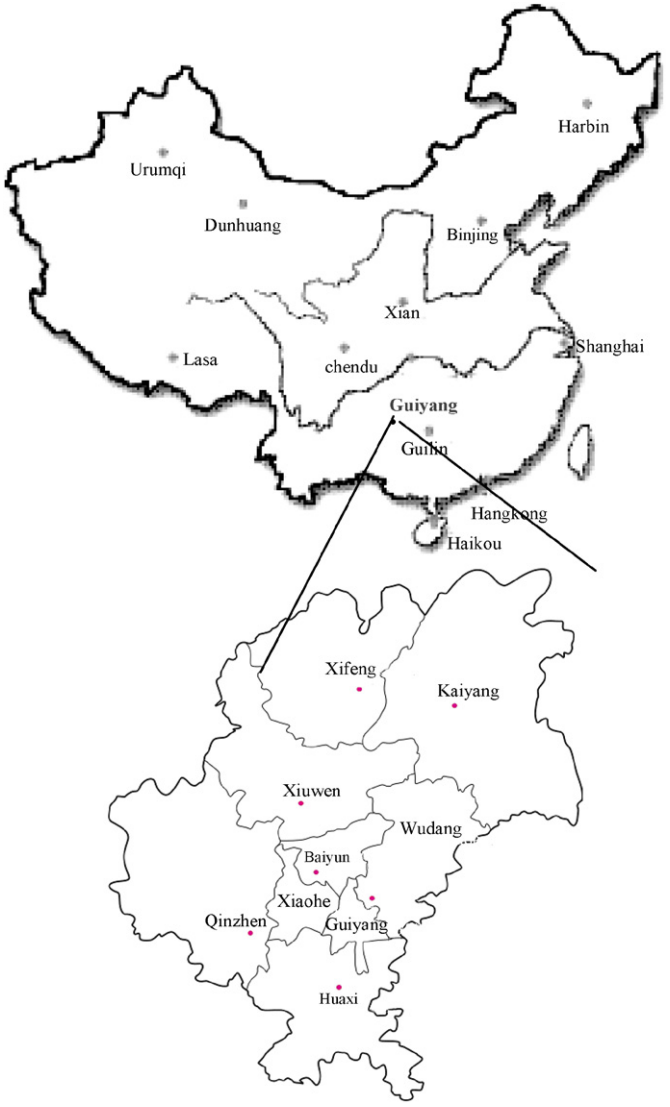

Fig. 1. The location of the sampling site in Guiyang (1—Guizhou coal-fired Boiler of Beer Plant; 2—Guiyang Power Plant).

In this study we chose GCPP to represent the large-scale coal combustion in Guiyang. The GCPP is situated 10 km away to the north of the city center. We also chose the 10 t/h coal-fired steam boiler of Guizhou Beer Plant (GBP) as a representative of industrial medium-to-small coal combustion to study mercury species in flue gas. The GBP is located 5 km away to south of the city center and is equipped with a wet flue gas precipitator and desulfurization (WFGD) device. Meanwhile, the samples taken at the flue gas inlet of WFGD represents hundreds of medium-to-small-sized industrial coal-fired boilers without flue gas cleaning devices.

2.2. Sampling and analysis

One campaign was conducted through June 25–28 in 2002 to sample the flue gas inlet and outlet of WFGD of GBP. Although the coal fired in GBP comes partially from raw coal of Guiyang Basin and partially from conventional physical coal cleaning process (CPCCP), coals from different sources was not sampled in this work because Hg content of raw coal from Guiyang Basin was previously studied in detail (Feng and Hong, 1999; Feng et al., 2002). During the full load operation, samples of the fired-CPCCP at stokehole, clinker, process water used in flue gas precipitator and desulfurization, wastewater with WFGD-reactants and flue gas were collected. The CPCCP, coal ash, process water and wastewater were sampled every 3 h and 3 times per day with duration of 4 days. The CPCCP and clinker samples were dried to constant weight at 40 °C and then analyzed in laboratory. The inlet and outlet of WFGD flue gas were sampled about 1 m^3 with Ontario Hydro Method (OHM) to determine the mercury species in flue gases. The OHM has now been validated as a mercury speciation measurement method in a full-scale test program at a Midwestern power plant (ERRI, 1999; Laudal et al., 2000). Nearly one Particulate Hg (Hg^p) was collected on a 47 mm diameter glass fiber filter, and gas phase Hg (Hg^{2+}, Hg^0) were collected in seven impingers solutions that connected in a series. Hg^{2+} was collected in the first three impingers containing KCl solution and Hg^0 was collected in the fourth impinger containing H_2O_2–HNO_3 solution and the last three impingers containing $KMnO_4$–H_2SO_4 solution. The sampling lines, i.e. Teflon tubings, were heated to 120 °C to prevent flue gas condensation. The eighth impinger containing silica gel was used to remove the moisture from the sampling line before entering the gas meter. All impingers were placed in an ice bath to prevent evaporation of the sampling solutions. Particulate mercury collected on the filters were digested with microwave oven in a Teflon container and determined using dual gold amalgamation coupled with CVAFS technique.

Another campaign was made for the sampling at the electrostatic precipitators (ESPs) outlet of the 200 MW large-scale boiler of GCPP with OHM method through July 14–16 in 2003. The fired-CPCCP was also sampled every 3 h and 3 times per day with duration of 3 days, and dried before analysis for laboratory measurement.

In all, 0.2 g dry sample of CPCCP, clinker, 10 ml process water and wastewater with fly ash were put into the PTFE vessel of micro-wave digestion system (MDS-2000) with 15 ml mixture of concentrated HNO_3, $HClO_4$, H_2SO_4 (7:5:3) solution, and then digested at 150–160 °C for 6 h. After cooling to room temperature, 1 ml concentrated HCl was added to each of the digested solutions in order to stabilize the dissolved mercury. The solution was then transferred to 25 ml volumetric flasks as preparative samples. A standard coal sample of US NIST 1630a was used to accomplish QA/QC.

Mercury content in the impingers and preparative samples was analyzed using dual gold amalgamation coupled with CVAFS after $SnCl_2$ reduction technique. Tekran 2500 AFS Hg detector was used and it was calibrated with the injection of a known amount of saturated mercury vapor (Tekran 2505) and analyzed regarding the CVAFS analysis of the Hg in solution of OHM. The absolute detection limit of the instrument is 0.1 pg.

3. Results and discussion

3.1. Mercury concentrations in CPCCP coal, coal ash, process water, and waste water

Our previous studies showed that the raw coal consumed in Guiyang mainly come from Guiyang coal basin with an average mercury concentration of 0.38 μg/g and the average mercury concentration in coal of Guizhou province was 0.50 μg/g (Feng and Hong, 1999; Feng et al., 2002), the results of which are higher than other provinces in China. The CPCCP can remove 50% of the mercury from raw coal (Feng et al., 2002), but only 14% of the raw coal has been processed with CPCCP in 2003 in Guiyang (EPA of Guizhou Province, 2004). Control technologies used to reduce criteria air pollutant emissions (e.g., particulate matter) from various combustion boilers also remove certain amount of Hg from flue gas, however, the removal efficiencies vary widely. Only 7% of the medium-to-small-sized coal combustions have been installed with flue gas control device while the domestic coal was burned without any flue gas cleaning device (EPA of Guizhou Province, 2004).

The analysis results were listed in Table 1. The overall mean value of mercury concentration in the CPCCP coal of the GBP and GCPP were 0.17 ± 0.03 μg/g ($n = 12$), 0.20 ± 0.03 μg/g($n = 9$), respectively. These lower Hg levels also indicated that CPCCP can remove nearly 50% of the mercury from raw coal. Mercury concentration in clinker of the 10 t/h steam coal-fired boiler of GBP was higher than that in coal, possibly because of the lower combustion temperature, the incomplete burning and the sampled clinkers watered by the wastewater. The 10 t/h steam coal-fired boiler of GBP was renewed 3000 t/yr tap water for WFGD and then discharged 3000 t/yr waster water. Mercury concentration in wastewater was about 630 times higher than that of the process water, indicating that mercury in flue gas was removed effectively by process water.

Table 1
Hg concentrations in samples of fired-coal, coal ash, process water and waster water

Raw coal	CPCCP coal (μg/g)	Clinker (μg/g)	Process water (μg/l)	Wastewater (μg/l)
0.38 ± 0.18 ($n = 3$)	0.17 ± 0.03[a] ($n = 12$) 0.20 ± 0.03[b] ($n = 9$)	0.24 ± 0.04 ($n = 12$)	0.18 ± 0.02 ($n = 12$)	113.54 ± 0.23 ($n = 12$)

[a]Coal-fired boiler of Guizhou Beer Plant.
[b]Guiyang coal-fired Power Plant.

3.2. Mercury speciation in flue gases

Mercury speciation in the flue gases at the WFGD inlet and outlet of 10 t/h coal-fired steam boiler of GBP was summarized in Table 2. Mercury concentrations analyzed in flue gas in June 27 were higher than that on other days. It reflected that Hg in coal was inconsistent. The average concentrations of Hg^p, Hg^{2+}, Hg^0, and Hg^t in WFGD inlet flue gas were 0.31, 0.64, 0.78, and 1.73 μg/m^3, respectively, compared to the average Hg^p, Hg^{2+}, Hg^0, and Hg^t concentrations of 0.07, 0.04, 0.56, and 0.67 μg/m^3 in WFGD outlet flue gas, respectively. Therefore, Hg^0 was the main species in flue gas. The comparisons indicated that most of the Hg^p and Hg^{2+} were removed from flue gas by WFGD system.

Mercury removal efficiencies varied in different flue gas control devices. The mean mercury removal efficiency of coal-fired plants with dry particulate control was about 30% (Chu and Porcella, 1995), and that of ESP was 30–40% (Chow et al., 1995). Pavlish et al. (2003) reviewed US ICR data on mercury capture in boilers and existing devices indicating that both wet and dry flue gas desulfurization (FGD) systems removed 80–90% of the Hg^{2+}, but Hg^0 was not affected. The mercury removal efficiency of WFGD system in this work was consistent with Pavlish's. It is obvious that Hg^{2+} removal efficiency was much higher than that of Hg^p and Hg^0. The mean removal efficiency of Hg^p and Hg^{2+} was 93.8% and 75.9% based on the outlet and inlet average concentrations of Hg^p and Hg^{2+} in Table 2, while the WFGD Hg^0 removal efficiency was only 26.6% based on the outlet and inlet average concentrations of Hg^0 in Table 2. As a whole, the average Hg^t removal efficiency was 57.2% based on the outlet and inlet average concentrations of Hg^t in Table 2. These results were also generally consistent with the results of other researchers, e.g. 50% of the total mercury was removed by more effective dust cleaning device and 25–75% of the Hg emitted was water soluble Hg (Meji, 1991; Galbreath and Zygarlicke, 1996).

The inlet flue gas samples in ESP of GCPP were not obtained because our sampling tubing and glass fiber filter was obstructed by lots of particulates in the inlet flue gas of ESP. The average Hg^p, Hg^{2+}, Hg^0, and Hg^t concentration in ESPs outlet flue gas of GCPP were 0.89, 4.98, 3.32, and 9.19 μg/m^3, respectively, as shown in Table 3.

Table 2
The concentrations and relative distributions of different mercury species in flue gas from Guizhou Beer Plant (June 25–28, 2002)

Sampling sites	Sampling time	Hg^p		Hg^{2+}		Hg^0		Hg^t
		μg/m^3	%	μg/m^3	%	μg/m^3	%	μg/m^3
Inlet WFGPD	June-27	0.27	8.5	1.38	43.6	1.51	47.9	3.2
	June-28	0.28	30.4	0.26	28.0	0.36	41.6	0.93
	June-28	0.31	29.4	0.28	26.8	0.46	43.8	1.05
	Average	0.29	22.8	0.64	32.8	0.79	44.4	1.73
Outlet WFGPD	June-25	0.10	21.1	0.02	4.3	0.34	74.6	0.45
	June-25	0.06	10.3	0.09	14.0	0.46	75.7	0.61
	June-26	0.08	16.0	0.02	5.1	0.38	78.9	0.48
	June-27	0.07	4.9	0.01	0.7	1.41	94.3	1.50
	June-28	0.06	19.0	0.06	20.0	0.29	61.1	0.67
	Average	0.07	14.3	0.04	8.8	0.58	76.9	0.74

Table 3
The concentrations and relative distributions of different mercury species in flue gas from Guiyang Coal-fired Power Plant (July 14–16, 2003)

Sampling sites	Date	Hg^p		Hg^{2+}		Hg^0		Hg^t
		μg/m^3	%	μg/m^3	%	μg/m^3	%	μg/m^3
Outlet ESPs	7–14	0.81	8.8	4.92	53.4	3.49	37.9	9.22
	7–14	0.89	9.5	4.87	52.0	3.60	38.5	9.36
	7–14	0.93	9.8	4.79	50.6	3.75	39.6	9.47
	7–15	0.87	8.7	5.19	51.6	3.99	39.7	10.05
	7–15	0.84	9.9	4.86	57.2	2.79	32.9	8.49
	7–15	0.91	10.2	5.22	58.5	2.79	31.3	8.92
	7–16	0.83	9.3	4.85	54.1	3.29	36.7	8.97
	7–16	0.97	11.0	4.87	55.5	2.94	33.5	8.78
	7–16	0.93	9.9	5.25	55.7	3.21	34.0	9.43
	Average	0.89	9.7	4.98	54.2	3.32	36.1	9.19

3.3. Mercury emission factors

In order to check the reliability of OHM sampling system and the accuracy of flue gas mercury speciation data, mercury mass balance was carried out with the 10 t/yr steam coal-fired boiler of GBP in Fig. 2. Mercury input of the coal combustion resulted in 7200 t/yr CPCCP and 3000 t/yr fresh water which input 3.4615 g/d Hg to the boiler. Our studied boiler was installed with WFGD, the mercury output included 1180 t/yr clinker, 55,000 m^3/h flue gas, 3000 t/yr wastewater which output 1.44, 2.27, and 0.89 g/d Hg from the boiler, respectively. After passing through WFGD, the ratio of total mercury input/output of this boiler system was about 95% with a mercury emission factor of 26%, representing medium-to-small-sized coal-fired boilers with flue gas control device. In Guiyang only 7% of medium-to-small industrial coal combustion were installed with flue gas cleaning device (EPA of Guizhou Province, 2004), however, we did not sample medium-to-small industrial coal combustion without any flue gas control device. So flue gas output of boiler in GBP was sampled to representing medium-to-small industrial coal combustion without any flue gas control device. Mercury output of the boiler included 1180 t/yr clinker and 55,000 m^3/h flue gas, its output 1.44 and 2.27 g/d Hg from the boiler, respectively. The ratio of total mercury input/output of this boiler system were about 108% with a mercury emission factor of 66%. The mass balance calculation indicated that there was basis ($< \pm 10\%$) for the Hg emission factors because of error of OHM. This work showed that total mercury remained in clinker was relatively higher than that of other's results, possible reason was the same as the mercury concentration in sampled clinkers.

Fig. 3 showed the Mercury distribution and emission factors of GCPP that represents large-scale coal combustion. The boiler consumed 761,462 t coal in 2003, of which 60% coal washed by CPCCP was from Guiyang Basin and 40% coal without washing from some small coal mines near Guiyang. The average mercury concentration was 0.20 mg/kg in the washed coal and 0.38 mg/kg in the raw coal. The temperature of the flue gas in sampling site was 155.5 °C. The fired coal input 206 kg mercury to the boiler. Mercury output included fly ashes, clinkers and 2,172,523.5 m^3/h flue gases. The emission factors of GCPP representing large-scale combustion boilers were obtained based on the mercury species concentration in flue gases and the mercury in the fired coal in 2003. This research indicated that about 31% of Hg^t in coal emitted to atmosphere as Hg^0, contributing to the global cycling. However, 54% of the Hg^t including 8% Hg^p and 46% Hg^{2+} in coal was emitted to atmosphere, which deposited in the vicinity area of GCPP and QCPP, resulting in serious mercury pollution (Horvat et al., 2003). In all, 85% of Hg^t in coal was emitted into atmosphere from power plants.

3.4. Mercury emission from coal combustion in Guiyang

In light of upper emission factors, coal consumption, CPCCP coal percent and the flue gas cleaning device installation percentage, mercury emissions from coal combustion in Guiyang were estimated (Tables 4 and 5).

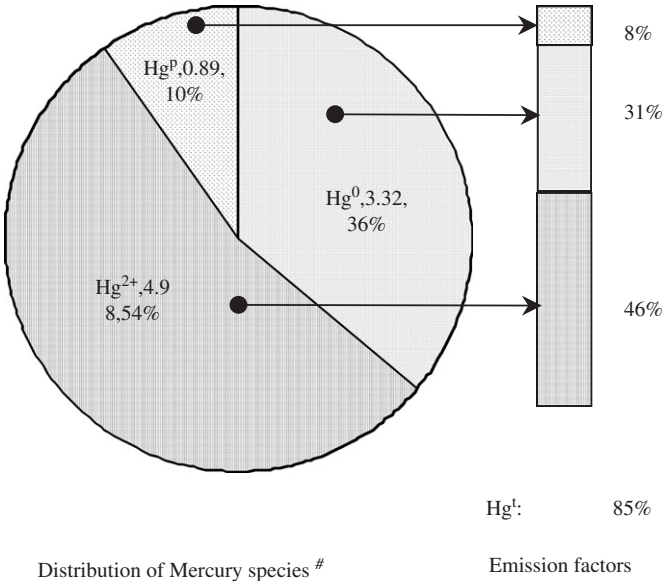

Fig. 3. Mercury distribution and emission factors of GCPP.

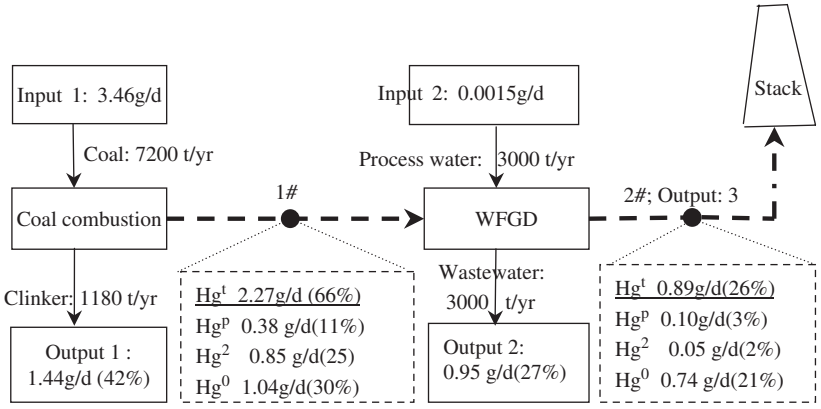

Fig. 2. Mass balance of total mercury and emission factors of mercury speciation from medium-small coal combustion (Guizhou Beer Plant).

Table 4
Mercury emissions within district of Guiyang in 2003

District	Power plant		Industrial steam boiler		Domestic use		Total	
	M_c (10^4 t)	Hg_t (kg)	M_c (10^4 t)	Hg_t (kg)	M_c (10^4 t)	Hg_t (kg)	Hg_t (kg)	Hg_t (%)
Nanming and Yunyan	76.14	176	22.93	40	47.25	110	326	17
Huaxi			14.79	26	19.15	45	71	4
Wudang			9.78	17	12.36	29	46	2
Baiyun			92.55	163	8.98	21	184	10
Xiaohe			19.23	34	9.68	23	57	3
Qingzhen	219.98	509	233.89	412	30.76	72	993	52
Kaiyang			19.4	34	26.24	61	95	5
Xiuwen			10.32	18	18.26	43	61	3
Xifeng			16.5	29	15.45	36	65	3
Total	296.12	685	439.39	773	188.13	440	1898	100

Table 5
Mercury emissions from all kinds of coal combustion in Guiyang (2003)

Coal combustion[a]	Coal consumption (10^4 t)	Hg emission (kg)			
		Hg^t	Hg^p	Hg^{2+}	Hg^0
Power plant	296.12	685	68	370	247
Industry	439.39	773	126	276	370
Raw chemical materials and chemical products	114.62	202	33	72	97
Metals mining and processing	97.19	171	28	61	82
Industry of which coal as raw material	62.92	111	18	40	53
Construction	62.60	110	18	39	53
Ordinary machinery manufacturing	4.66	8	1	3	4
Others	97.40	171	28	61	82
Domestic use	188.13	440	73	167	200
Total	923.64	1898	267	813	817

[a]Excluding $\leqslant$4 t/h smaller-sized industrial steam coal combustion.

The basic meanings about Hg emission calculation of large-scale coal-fired power plants were described as below:

$$Hg^t = M_c E_m [Hg^p C_p + Hg^c(1 - C_p)] * 10, \quad (1)$$

where Hg^t is mercury emissions to the atmosphere(kg), M_c is the amount of coal consumption (10^4 t), E_m refers to mercury emission factors(%), Hg^p is mercury content in CPCCP coal (0.20 μg/g), C_p is the proportion of CPCCP coal (%) that is 60% for power plant and 14% for industrial coal combustion (EPA of Guizhou Province, 2004), and Hg^c is mercury concentration in raw coal (0.38 μg/g).

Only 7% of industrial coal combustion facilities were installed with flue gas cleaning device in Guiyang (EPA of Guizhou Province, 2004), therefore, Hg emissions from the medium-to-small-sized coal-fired power plants were calculated as follows:

$$Hg^t = M_c[Hg^p C_p + Hg^c(1 - C_p)][(1 - Hg^v)Hg^b + Hg^v Hg^a] * 10, \quad (2)$$

where Hg^t is the mercury emitted into the atmosphere (kg), Hg^v is the proportion of applied flue gas cleaning device (7%), Hg^b is the emission factor before the flue gas cleaning device (66%), and Hg^a is the emission factor at the flue gas cleaning device outlet (26%).

Considering domestic coal combustion without any flue gas cleaning device, mercury emissions was calculated using medium-to-small-sized mercury emission factor before WFGD, shown below:

$$Hg^t = M_c Hg^b [Hg^p C_p + Hg^c(1 - C_p)] * 10. \quad (3)$$

Using the above calculation methods (Eqs. (1)–(3)), mercury emissions were calculated from different coal combustion sources. Table 4 showed mercury emissions within district of Guiyang in 2003. Coal combustion of Guiyang in 2003 emitted about 1898 kg mercury into atmosphere, which accounts for the estimation of 2.2 t in our previous study (Feng et al., 2002). The largest mercury emission from coal combustion was Qingzhen district with the emission rate of 52% in Guiyang, the second was

central city (Nanming and Yunyan) with 17%, and the third was Baiyun with 10%.

Table 5 showed mercury and mercury species emissions from power plant, industrial coal combustion and domestic use in Guiyang in 2003. Mercury species emissions of power plant were estimated using the Hg distribution in Table 3, which emitted about 685 kg mercury into the atmosphere including 68 kg Hg^p, 370 kg Hg^{2+} and 247 kg Hg^0. Mercury species emissions from industrial coal combustion were estimated with Hg distribution in inlet flue gas of WFGD of 7% coal consumption and Hg distribution in outlet flue gas of WFGD of 93% coal consumption. The estimation showed industrial coal combustion emitted 773 kg mercury into the atmosphere which included 126 kg Hg^p, 276 kg Hg^{2+}, and 370 kg Hg^0. Mercury emissions from domestic coal combustion emitted 440 kg mercury into the atmosphere which include 73 kg Hg^p, 167 kg Hg^{2+}, and 200 kg Hg^0 by using the Hg distribution in flue gas of inlet WFGDP in Table 2, because domestic coal combustion was installed with no cleaning devices. The estimation indicated that 36% of the total Hg emission was from the two large-scale coal-fired power plants, in which 64% was released as Hg^p and Hg^{2+}. Because of relatively centralized emission sources, mercury pollution around the two power plants was serious (Horvat et al., 2003). In addition, there were some bias of mercury emissions from industrial coal combustions and domestic users because only one industrial coal combustion facility was studied. The actual Hg emission from industrial coal combustion and domestic users was probably higher than that reported in this work and needs farther study.

3.5. *Mercury emission from coal combustion in Guizhou*

According to the investigation of coal consumption in Guizhou (2004), Coal-fired power plants consumed about 30% of the total coal in Guizhou province, while industry was the biggest coal-consuming sector, burning 48% of the total coal and 22% of the coal was consumed by domestic users. Based on the above calculations and discussions, we estimated the yearly mercury emission from coal combustion in Guizhou from 1986 to 2002 (Fig. 4). Mercury emission increased sharply from 5.8 t in 1986 to 16.4 t in 2002. With the implementation of national economic strategy of China's Western Development, about 30 large-scale coal-fired power plants will be built outside the city of Guiyang in the Guizhou province in the next 10 years and the provincial coal consumption will increase to 78 million tones in 2015 from 38 million tones in 2002 according to statistics of environmental protection administration of Guizhou (2004). The coal consumption in power plants will increase 58 million tons while 15.8 t of mercury will be emitted into the atmosphere, and the annual mercury emission from coal combustion in the whole province would be about 32 t in 2015, which is comparable to the annual coal combustion mercury emissions in the whole USA (Pavlish et al., 2003).

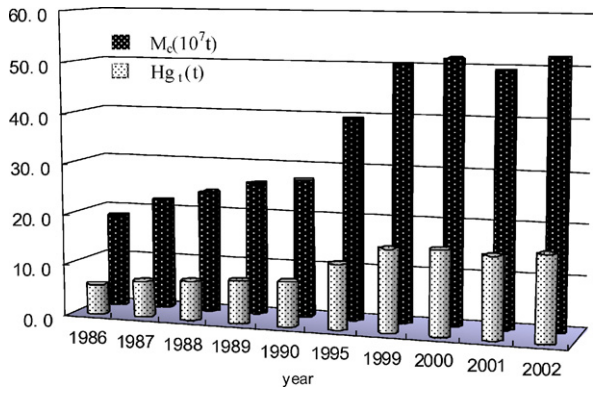

Fig. 4. Mercury emission of Guizhou from 1986 to 2002.

4. Conclusions

- We present, for the first time, a detailed mercury speciation investigation of three types of flue gases from two types of coal combustion in Guiyang.
- Mercury emission factors of medium-to-small-sized coal combustion systems without WFGD systems were 66% of Hg^t including 11% of Hg^p, 25% of Hg^{2+} and 30% of Hg^0, respectively. Mercury emission factors at WFGD system outlet were 26% of Hg^t with 3% of Hg^p, 2% of Hg^{2+}, and 22% of Hg^0. The Hg^p, Hg^{2+}, Hg^0, and Hg^t emission factors in large-scale coal combustion power plant with ESPs were 8%, 46%, 32%, and 85%, respectively.
- Coal combustion of Guiyang in 2003 emitted 1898 kg mercury into the atmosphere, including 267 kg Hg^p, 813 kg Hg^{2+}, and 817 kg Hg^0. Mercury emitted from power plant, industrial coal combustion and domestic use were 36%, 43%, and 23%, respectively, regarding to the total mercury emission of Guiyang city. Mercury pollution around the two large-scale coal-fired power plants was serious and further follow-up study was needed.
- Mercury emission increased sharply from 5.8 t in 1986 to 16.4 t in 2002 in Guizhou. With the national economic strategy of China's Western Development implementation, the newly constructed power plants will emit 15.8 t mercury into atmosphere. The annual mercury emission from coal combustion in the whole province will reach about 32 t in 2015, which was comparable to the annual coal combustion mercury emissions in the whole USA, posing high Hg pollution risk. Further study is needed for Hg speciation in flue gas in other provinces of China, and regulations of mercury emission from coal combustion must be put in place.
- Much more industrial and domestic coal combustion need in-site measurements of Hg species and fix on reliable emission factors to estimate Hg emission in Guizhou and other large coal consumption province in China, such as Shanxi and Henan, to estimate Chinese mercury emissions from coal combustion.

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