OCCURRENCE, EMISSIONS AND DEPOSITION OF MERCURY DURING COAL COMBUSTION IN THE PROVINCE GUIZHOU, CHINA

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Abstract. Forty-eight coal samples from the major coal mines in Guizhou were collected by means of subterranean channel sampling. The average mercury content in coal from this Chinese province is 0.53 mg kg^{-1}, which is substantially higher than coal produced in any other province of China. A strong link is proposed to exist between atmospheric mercury emissions and the combustion of coal. During the last two decades, the amount of mercury emitted from coal combustion in Guizhou has increased invariably to 8.3 tons in 1998. Being concentrated to the pyrite fraction in the coal, the implementation of physical coal cleaning is likely to be an effective way to reduce the mercury content. Total and fractionated mercury in air and precipitation was measured in the provincial capital. During a measurement campaign in October 1999, the reactive gaseous mercury (RGM) concentration averaged at 450 pg m^{-3}, which is much higher than that in pristine area (less than 20 pg m^{-3}). The average total mercury in rainwater of 1996 was estimated to be 33 ng L^{-1}, and the total annual wet deposition of mercury in the area was 39 g km^{-2} in 1996.

Keywords: atmospheric pollution, mercury speciation, precipitation, reactive gaseous mercury

1. Introduction

The province of Guizhou is located in Southwestern China with an area of 170 000 km^2, accounting for about 1.8% of the total area of China. Guizhou is situated on a plateau with a mean altitude of about 1000 m. Its climate represents a typical subtropical humid monsoon with an average annual temperature of 15 °C and a precipitation of 1100–1400 mm (The Government of Guizhou Province, 1987). Guizhou is one of the largest coal producing provinces and is also one of the areas in China where acid deposition occurs frequently due to coal combustion emissions. Currently about 20 million tons of coal are burnt for both industrial service and domestic use each year in Guizhou and the amount is predicted to increase due to the regional economic development.

The provincial capital of Guizhou, Guiyang is its most industrialized area and classified as one of the most seriously polluted cities in China. Acid rain caused by coal combustion emissions is regarded as 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

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1992 were 468 and 395 μg m^{-3}, respectively, exceeding the national air quality standards, which are 100 and 300 μg m^{-3} for SO_2 and TSP, respectively.

Anthropogenic mercury emission sources are usually categorized as coal combustion, solid waste incineration, oil combustion, pyrometallurgical process including Cu/Zn production and Pb production, wood combustion and a miscellaneous category which includes chlor-alkali plants, degassing of latex paint, crematoria, fluorescent lamp breakage, dental laboratory, cement manufacturing, primary and secondary mercury production, and other coal uses (Pirrone *et al.*, 1996; Nriagu and Pacyna, 1988). Coal combustion and solid waste incineration are worldwide the largest among these source categories (Pirrone *et al.*, 1996). The number of domestic municipal waste incineration facilities is still very scarce (Hunsicker *et al.*, 1996), hence coal combustion is currently by far the major anthropogenic emission source of mercury in China. A rough estimation (Wang *et al.*, 2000) showed that the annual mercury emission from coal combustion (MEFCC) in China is around 300 tons, including 210 tons into the atmosphere and 90 t into ash and cinder. In this article, the mercury concentrations in coals from the main coal production mines in Guizhou are reported and an estimation of annual MEFCC in Guizhou is given. In addition, a preliminary study of mercury in the air and precipitation in Guiyang is presented.

2. Methods and Materials

2.1. Sampling of Coal

Forty-eight crude coal samples from 4 coal basins (Shuicheng, Liuzhi, Panjiang and Guiyang coal basin), which are the major coal production sources in Guizhou, were collected by means of underground channel sampling. The main coal-bearing stratum in these coal basins is that of the Longtan Formation from the Permian system. Anthracite and bituminous coal are two major types of coal in Guizhou. Among the 48 coal samples, 44 are bituminous coals and the rest are anthracites. After homogenization, milling and riffling following ISO recommendations, coal samples were milled and ground to <250 μm prior to chemical analysis.

2.2. Determination of Mercury in Coal

Forty samples were analyzed at the State Key Laboratory of Environmental Geochemistry (SKLEG), Institute of Geochemistry, Chinese Academy of Sciences, and 8 samples were processed at the Department of Inorganic Chemistry (DIC), Göteborg University in Sweden. At SKLEG, 0.1 g dry sample was oxidized with 3 mL concentrated HNO_3 in an PTFE-lined steel pressure bomb for 24 hr at 140 °C. After cooling down to the room temperature, the digested solution then was transferred to a 25 mL volumetric flask (Feng and Hong, 1998). At DIC, about 0.2 g dry sample was put in a quartz tube, inside a sealed acid-digestion bomb, together

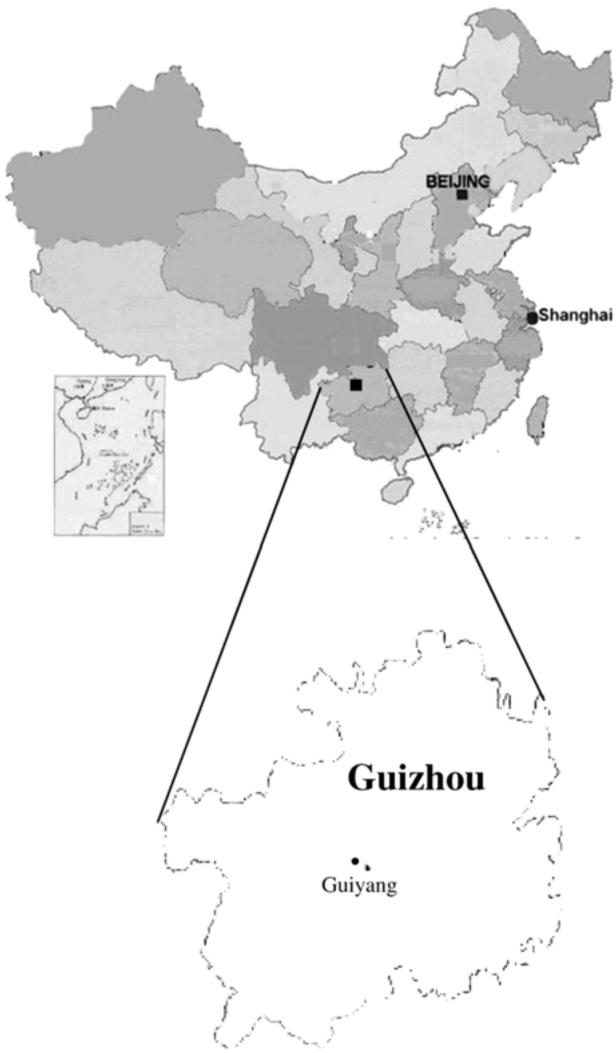

Figure 1. The location of the sampling site in Guiyang (■).

with 15 mL mixture of concentrated HNO_3, $HClO_4$, and H_2SO_4 (7:5:3) solution. These bombs were then placed in an oven at 150–160 °C for 6 hr. One mL concentrated HCl was then added to each of the digested solutions in order to stabilize the dissolved mercury. The solution was then transferred to 25 mL volumetric flasks. At both laboratories, the concentration of Hg was determined by two-stage amalgamation coupled with CVAAS after $SnCl_2$ reduction technique. A standard coal sample SRM 1630a was used to accomplish QA/QC at both laboratories. The results showed that both pretreatment procedures can quantitatively recover mercury from the standard sample.

TABLE I
Measurement results of mercury concentrations in coals of Guizhou

Coal type	Concentration (mg kg^{-1})				Sample
	Min	Max	Average	Std. dev.[a]	number (n)
Anthracite	0.19	0.69	0.42	0.20	4
Bituminous coal	0.10	2.67	0.54	0.52	44
Over all			0.53	0.55	48

[a] Std. dev. = Standard deviation.

2.3. Mercury in Air and Precipitation of Guiyang

A measurement site in Guiyang, where the Institute of Geochemistry, Chinese Academy of Sciences is located, was chosen for measurement of mercury in the air and precipitation (Figure 1). In 1996, total gaseous mercury (TGM) in the air was collected on gold traps at a flow rate of 1 L min^{-1} for 2 to 4 hr, and analyzed by CVAAS. In 1999, an automated mercury analyzer (Gardis 1A) (Urba *et al.*, 1995) was used to measure TGM continuously at a sampling time of 5 to 10 min. Reactive gaseous mercury (RGM) was collected in KCl coated denuders and analyzed by thermal desorption coupled with CVAAS detection (Feng *et al.*, 2000a). The sampling flow rate was 1 L min^{-1} and the sampling time was 20 hr.

From June till November 1996, rain samples were collected through a Teflon funnel with a diameter of 30 cm to a Teflon bottle. Every single event sample was collected during that period and after the collection, the samples were delivered to the laboratory and the reactive mercury and total mercury concentrations were analyzed within 24 hr (Feng *et al.*, 1998).

3. Results and Discussion

3.1. Mercury Concentrations in Coal of Guizhou

The measurement results of mercury concentrations in 48 crude coal samples from Guizhou are summarized in Table I, and it can be seen that Hg contents are 0.42±0.20 (n = 4) and 0.54±0.52 (n = 44) mg kg^{-1} in anthracite and bituminous coal, respectively. The average mercury concentration in crude coal of Guizhou is 0.53 mg kg^{-1}. Comparing the average mercury concentration in crude coal of Guizhou with that in other provinces in China, which are reported by Wang and coworkers (2000) as shown in Table II, we can easily see that mercury concentration in crude coal of Guizhou is much higher than that in other provinces in China. The low-temperature thermal fluid activities in the Mesozoic and Cenozoic Eras caused high background of mercury in bed rocks in the survey area (Chen, 1999). It can be

TABLE II

Mercury concentration in coal of main coal production provinces in China (mg kg^{-1})

Province	Range	Average	Std. dev.[c]
Heilongjiang[a]	0.02–0.63	0.12	0.11
Jilin[a]	0.08–1.59	0.33	0.28
Liaoning[a]	0.02–1.15	0.20	0.24
Inner Mongolia[a]	0.06–1.07	0.28	0.37
Beijing[a]	0.23–0.54	0.34	0.09
Anhui[a]	0.14–0.33	0.22	0.06
Jiangxi[a]	0.08–0.26	0.16	0.07
Hebei[a]	0.05–0.28	0.13	0.07
Shanxi[a]	0.02–1.95	0.22	0.32
Shaanxi[a]	0.02–0.61	0.16	0.19
Shandong[a]	0.07–0.30	0.17	0.07
Henan[a]	0.14–0.81	0.30	0.22
Sichuan[a]	0.07–0.35	0.18	0.10
Xinjiang[a]	0.02–0.05	0.03	0.01
Guizhou[b]	0.10–2.67	0.53	0.51

[a] Data from Wang *et al*., 2000.
[b] This work.
[c] Std. dev. = Standard deviation.

speculated that these thermal fluid activities might bring mercury in coal seam as well, resulting in high mercury content in coal. However, further research is needed to verify this hypothesis.

It has been shown that mercury mainly occurs in pyrite in coals from Guizhou (Feng and Hong, 1999). While pyrite has a high density, much of it is liberated during conventional physical coal cleaning process (CPCCP) (Couch, 1991) and the mercury associated with this fraction may be released and removed from coal. To the best of our knowledge, no explicit data on removal efficiency of mercury by the coal cleaning procedure employed here exists in the open literature. However, a substantial reduction is implied by comparing the mean mercury content in refined coals of Guizhou obtained in a previous study (Lindqvist *et al*., 2001) with that of crude coals, 0.15 and 0.53 mg kg^{-1}, respectively. At present, only 7.8% of coal output per year has been performed with CPCCP in Guizhou. Therefore, with the increase of the ratio of cleaned coal, the average mercury contents in coal utilized will decrease drastically. Taking into account of the fraction of fuel subject to CPCCP, the current average mercury concentration in commercial coals of Guizhou can be computed to be 0.50 mg kg^{-1}.

TABLE III

Annual coal productions and consumptions in Guizhou and coal consumptions in Guiyang (in Mt) (Statistical Bureau of Guizhou, 1999)

Year	Production in Guizhou	Consumption in Guizhou	Consumption in Guiyang
1981	10.1	4.3	1.5
1982	12.1	4.6	1.7
1983	13.2	4.9	1.8
1984	14.5	5.3	1.9
1985	16.7	6.9	2.0
1986	18.2	7.6	2.2
1987	22.2	9.9	3.0
1988	22.9	11.0	2.7
1989	25.0	11.3	3.0
1990	26.5	10.9	3.1
1991	26.7	11.9	3.2
1992	29.0	13.2	3.4
1993	32.3	14.1	3.4
1994	36.6	16.1	3.7
1995	39.1	17.3	4.4
1996	43.9	18.7	4.8
1997	47.1	20.3	5.2
1998	46.9	21.7	5.5

3.2. MEFCC IN GUIZHOU AND GUIYANG

Coal is the principal energy source in Guizhou, accounting for about 80% of the total energy consumption. Both production and consumption of coal have been increasing significantly in parallel with the increase of economy and population during the last two decades. Table III lists annual coal production and consumption in Guizhou and coal consumption in Guiyang City. Approximately a quarter of the coal consumed in Guizhou was burnt in Guiyang.

A detailed coal consumption scenario in Guizhou in 1998 is listed in Table IV. Coal-fired power plants consumed nearly one half of the total coal consumption. The powdered coal boilers are widely used in most power plants and only particulate control devices that mainly include electrostatic precipitator and wet scrubber are applied. According to mercury mass balance studies conducted by Wang and co-workers (2000), from 53.4 to 93.5% and with an average of 74.3% mercury in coal will be released into the atmosphere during coal combustion in such

TABLE IV

Mercury emissions from all kinds of coal combustions in Guizhou (1998)

Consumer	Coal consumption (10^4 t)	Hg emissions (t)
Farming, forestry, animal husbandry, fishery	132.12	0.42
Industry	1474.64	5.26
Mining and quarrying	223.61	0.72
Manufacturing	195.89	0.63
Others	20.22	0.06
Electric power	1034.92	3.85
Construction	1.62	0.005
Transportation, postal and telecomunications	3.68	0.012
Wholesale, retail trade, and catering service	50.20	0.16
Domestic use	507.74	2.48
Total	2170	8.3

powder boilers in China. A mercury emission factor (MEF) of 0.37 g t^{-1} from power plant can be calculated from the data on the average mercury concentration in commercial coal in Guizhou and the mercury balance study during coal combustion.

Domestic coal consumption constituted nearly one quarter of the total consumption in Guizhou in 1998. More than 80% of the total population (36.6 million) use coal to process food in Guizhou. A consequence might be food poisoning, such as endemic arsenism that occurs in Guizhou (Zheng *et al*., 1999). Coal is burnt in a small scale without any pollution control device by almost every family in countryside and some small cities and it was estimated that $\sim$98% mercury in coal was released into air during coal combustion (Feng, 1997). An MEF of 0.49 g t^{-1} for domestic combustion can thus be estimated from the average mercury concentration in commercial coals.

In such users as farming, forestry, animal husbandry and fishery, mining and quarrying, manufacturing, construction, transportation and so forth, layer-burning boilers are used widely. Mercury mass balance studies showed that 64% of total mercury in coal was emitted into the air (Wang *et al*., 2000), and the corresponding MEF is 0.320 g t^{-1}.

In the light of the average MEFs and the coal consumption of different sections in 1998, the mercury emissions from different coal combustion processes in Guizhou were computed (Table IV). While the four coal basins (Shuicheng, Liuzhi, Panjiang and Guiyang coal basin) mentioned in Section 2.1 have since 1981 been the major coal production sources in Guizhou and the ratio of coal performed with

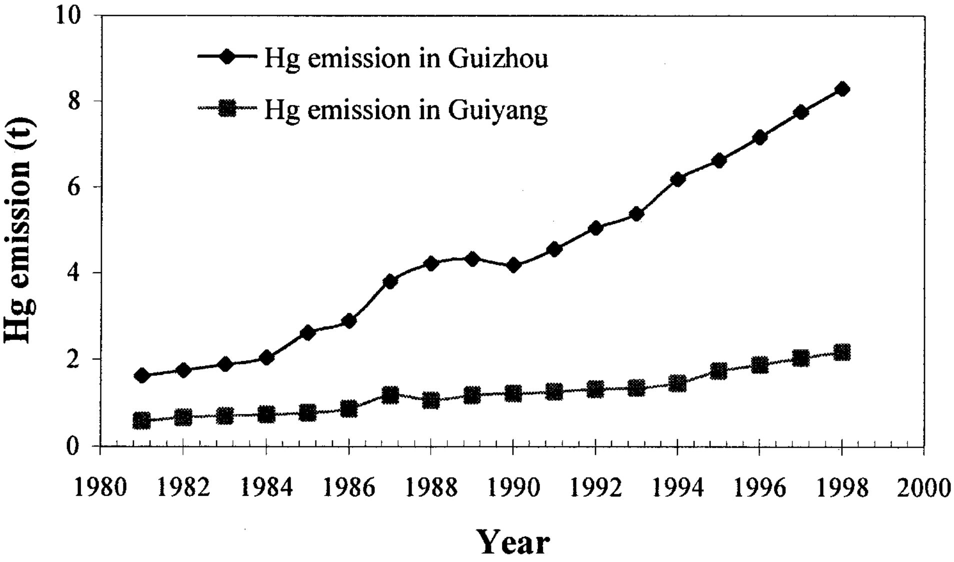

Figure 2. Mercury emission from coal combustion in the province Guizhou and its capital Guiyang.

CPCCP in Guizhou has been stationary, it is therefore reasonable to assume that the average mercury concentration in the commercial coal of Guizhou and the MEF is temporally invariable. Correspondingly, the annual MEFCC in Guizhou and Guiyang from 1981 to 1998 were calculated based on the coal consumption data (Figure 2). According to our estimation, the total mercury emission into the air from coal combustion in Guizhou was 6.6 t in 1995, which is significantly different from that (22.58 t) estimated by Wang and co-workers (2000). In their calculation, they assumed that all coal produced in Guizhou were consumed in the province, it is actually not true, however, from the statistic data listed in Table III (Statistic Bureau of Guizhou, 1999). In 1995, only 44% coal produced in the province was combusted in Guizhou. The average mercury concentration in coal of Guizhou (0.6 mg kg^{-1}) applied in their calculation corresponds to eastern Guizhou (Ni *et al.*, 1998), which is slightly higher than that used in this article. Moreover, in their estimation the reduction of mercury emission resulted from implementation of CPCCP was not properly considered. Taking into account the factors mentioned above, it is obvious that Wang and co-workers overestimated the amount of MEFCC in Guizhou 1995.

A clearly increasing trend of MEFCC in Guizhou and Guiyang in the past 20 yr can be seen, and of 8.3 t Hg emitted into the air from coal combustion in Guizhou 1998, 2.2 t was emitted in Guiyang. From 1996 to 1998, MEFCC in Guiyang increased at an average annual rate of 7.1%. Since the CPCCP will significantly remove mercury from coal, the implementation of the process, which is much

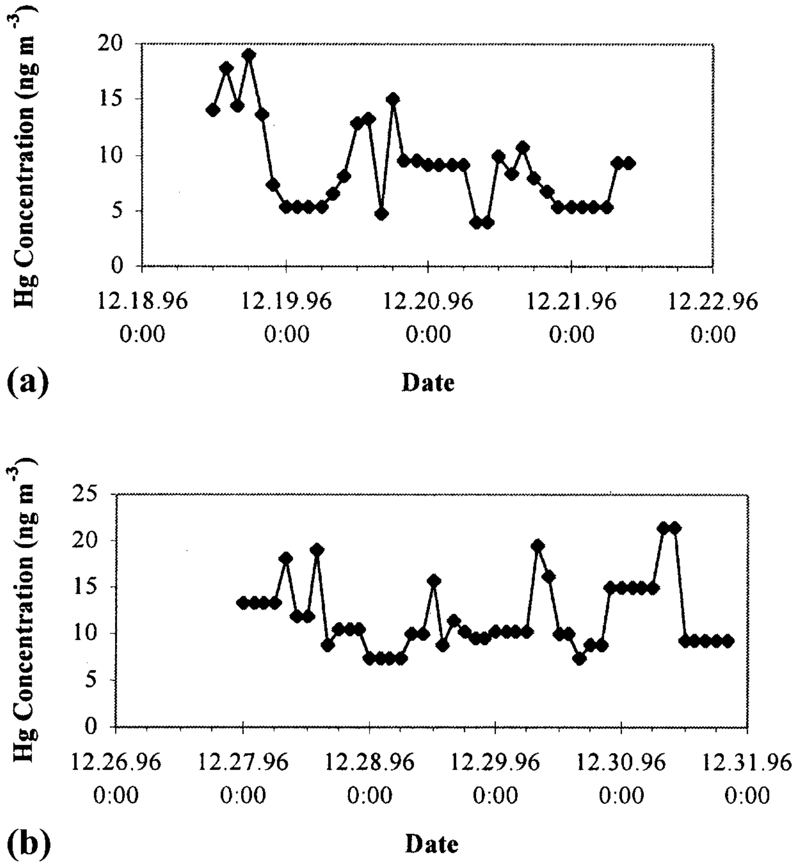

Figure 3. Total gaseous mercury in the air of Guiyang in December 1996.

easier to set up than installation of flue gas cleaning, is a crucial way to reduce MEFCC in Guizhou for the moment.

3.3. Mercury in Air and Rain in Guiyang

3.3.1. *Mercury in Air*

Two measurement campaigns were carried out in December 1996 and in October 1999, respectively. In the first campaign, only TGM was measured using manual method with a time resolution of 2 to 4 hr, and the measurement results are shown in Figure 3. During the second campaign, an automated mercury analyzer was used and a high time resolution data set of TGM in air was achieved and presented in Figure 4. Generally, TGM concentrations in air during nighttime were relatively low and stationary since the dominant nocturnal wind direction in Guiyang is northeastern, and therefore not from the source areas which are located at 5 km southeastern from the measurement point. In daytime, anthropogenic activities in-

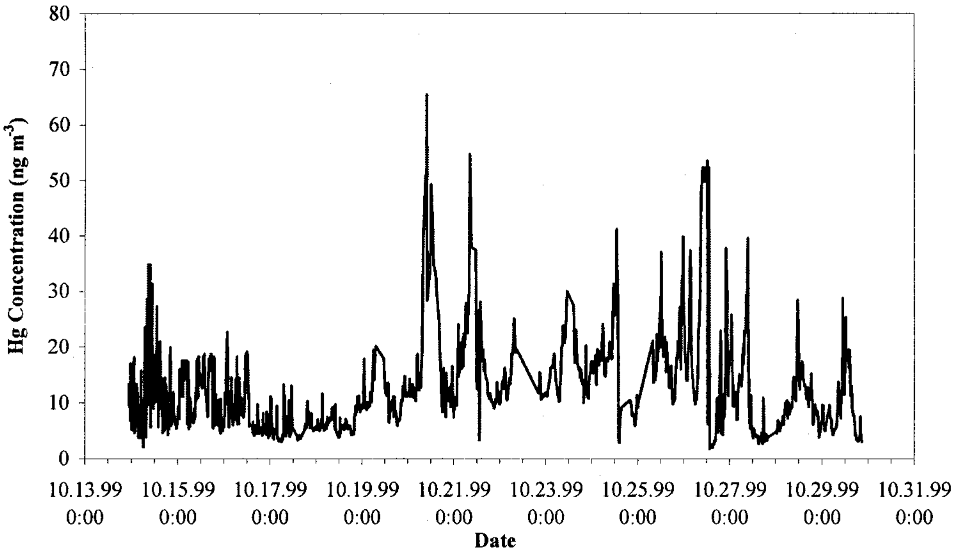

Figure 4. Total gaseous mercury in the air of Guiyang in November 1999.

TABLE V

Reactive gaseous mercury concentrations in the air of Guiyang in 1999

Date	RGM (pg m^{-3})	Average (pg m^{-3})	Weather conditions
October 15	785.8	785.8	Sunny
October 16	167.7, 191.5	179.6	Cloudy/sunny
October 17	171.8, 205.2	188.5	Cloudy/sunny
October 18	593.7, 513.2	553.4	Sunny
October 19	739.0, 700.7	719.9	Sunny
October 20	592.5, 589.3	590.9	Sunny
October 21	697.3, 678.6	687.9	Sunny
October 23	625.3, 299.3	462.3	Cloudy/sunny
October 24	89.7, 73.9	81.8	Cloudy with short period drizzle
Average		453.8±248.0	

creased dramatically and the wind direction was relatively unstable, which caused increased and unstable TGM concentrations. The average TGM concentration in air was 11±4 ng m^{-3} in the 1996 campaign, and was 13±9 ng m^{-3} in the 1999 campaign, which were significantly higher than the reported levels for background continental areas in the range of 1.0 to 4.0 ng m^{-3} (i.e. Lindqvist and Rodhe, 1985; Fitzgerald *et al.*, 1984).

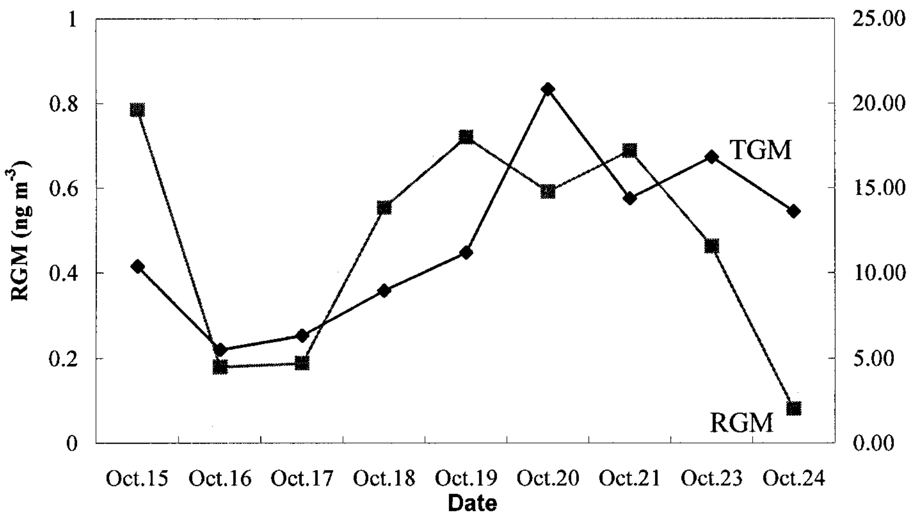

Figure 5. Daily average total gaseous mercury and reactive gaseous mercury concentrations in the air of Guiyang, 1999.

During the second campaign in 1999, daily average RGM concentrations in air were measured. Results from two parallel sampling lines are generally concordant, as shown in Table V. The average RGM concentration during the sampling period was 450±250 pg m^{-3}, which are much higher than that measured at background areas in Europe (Sommar *et al.*, 1999; Feng *et al.*, 2000a) and in U.S.A. (Lindberg and Stratton, 1998). RGM constituted 3.9% of TGM in the air of Guiyang. Figure 5 shows daily average TGM and RGM in the air during the measurement period. It can be seen that RGM generally followed the temporal trend of TGM quite well, implying that RGM and TGM came from the same sources. With development of new reliable methods for speciation of mercury in coal combustion flue gases (i.e. Prestbo and Bloom, 1995; Feng *et al.*, 2000b), it is demonstrated that flue gases from coal combustion do contain RGM species such as $HgCl_2$, which are shortly lived in the air contrary to Hg^0. Again, the evident highlights that the major RGM sources in air of Guiyang came from coal combustion.

3.3.2. *Mercury in Precipitation*

The annual rainfall in Guiyang 1996 was 1186.7 mm, and most precipitations occurred in the summer (Statistic Bureau of Guizhou Province, 1997). Thirty-one rain samples were collected from June to November 1996. The pH of these rain samples varied from 4.4 to 3.1, and SO_4^{2-} was the dominant anion. It is generally accepted that the acidification of rain in Guiyang is caused by the regional coal combustion emissions (Shen *et al.*, 1993). The total mercury (Hg_{tot}) in rain ranged from 10.3 to 90.6 ng L^{-1} with an average of 33 ng L^{-1}, and the reactive (Hg_R) form varied from 0.8 to 13.2 ng L^{-1} and averaged at 4 ng L^{-1}. Table VI lists monthly-based total

TABLE VI

Total and reactive mercury in rain samples of different month in Guiyang 1996 (ng L^{-1})

Month	Hg_{tot}	Hg_R	Hg_R/Hg_{tot} (%)	N
June	33.0	2.0	6.1	1
July	28.1±3.0	1.1±0.1	3.9	8
August	22.1±6.5	1.7±0.6	7.7	10
September	34.1	2.9	8.5	1
October	28.9±15.8	5.9±4.3	20.4	5
November	50.9±21.6	7.4±1.6	14.5	6
Average[a]	32.9±17.6	4.2±3.4	12.8	31

[a] Average of 31 rain samples.

and reactive mercury concentrations in rainwater samples. On average, the reactive mercury constituted 12.8% of total mercury in rainwater.

It is reasonable to assume that the average Hg_{tot} concentration obtained from these samples represented the average Hg_{tot} concentrations in the precipitation in 1996, since these samples covered 90% of the annual rainfall in Guiyang. In the light of the average Hg_{tot} concentration in rainwater and annual rainfall, we roughly estimated that the wet deposition rate of this area in 1996 was 39 g km^{-2} a^{-1}, which is much higher than that of Nordic countries (Iverfeldt, 1991). On the other hand, as mentioned previously, RGM in air of Guiyang were quite high, which will lead a high dry deposition rate (Schroeder and Munthe, 1998). In a global scale, dry deposition accounts for around 54% of total mercury deposition (Shia *et al*., 1999). An earlier study conducted by Xiao and coworkers (1998), however, showed that the dry deposition consist of 73% of the total Hg deposition in Guiyang. High ratios of particulate mercury, which was not measured in this study and of RGM, with respect of the total mercury in the air of Guiyang, could explain the high proportion of dry deposition in this area. Considering dry deposition, the total deposition of mercury in Guiyang reached 143 g km^{-2} in 1996 and now the annual total mercury deposition in Guiyang must be increasing due to the increase of MEFCC.

4. Summary

The average mercury concentration in crude coal of Guizhou, China is 0.53 mg kg^{-1}, which is significantly higher than that of other coal production areas in China. Since mercury in the coals mainly exists in pyrite, a large part of mercury may be removed by using the CPCCP technique. However, only 7.8% of the coal output per year has been treated with the CPCCP right now. A clearly increasing trend of MEFCC in Guizhou in the past 20 yr can be seen, and totally 8.3 t Hg was emitted into the air from coal combustion in Guizhou in 1998. As the CPCCP will significantly remove mercury from coal, the implementation of the process, which is easier to set up than installation of flue gas cleaning, is a crucial way to reduce MEFCC in Guizhou for the moment. In the long run, the installation of cleaning devices in flue gas cleaning for SO_2, NO_X, dust and mercury is necessary to ultimately eliminate the emissions of pollutants from coal combustion.

Around 2.2 t of mercury were emitted from coal combustion in Guiyang in 1998. From 1996 to 1998, MEFCC in Guiyang increased at an average annual rate of 7.1%. Elevated TGM and RGM concentration in air of Guiyang were observed. Coal combustion emissions are probably the primary mercury source in Guiyang.

The average total and reactive mercury concentrations in rainwater of Guiyang in 1996 were 33 and 4 ng L^{-1}, respectively. Since mercury emission rate has been increasing, the total mercury concentration in rainwater must be increasing. In 1996, the total wet deposition of mercury was estimated to be 39 g km^{-2}. Obviously, much research is needed to scrutinize the ecological effects and human health impacts causing by such a large amount of mercury deposition in this area.

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