Current Status and Atmospheric Mercury Emissions Associated with Large-Scale Gold Smelting Industry in China

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ABSTRACT

Mercury, being bound with metal concentrate, is released to atmosphere in the smelting process, and has adverse effects on surrounding ambient air quality and public health. In this study, based on literature and field investigations, onsite measurements of mercury emissions to the atmosphere for four large-scale gold smelters were estimated. Almost all of the atmospheric mercury released from the roasting pretreatment process occurred before acid plants. All the concentrations of mercury emitted from the four tested gold smelters were much lower than the emission standard of air pollutants for GB 9078–1996 Grade II level areas of 1000 $\mu g\ m^{-3}$. The speciation of Hg_p took up the largest proportions for the total mercury concentrations. Because of using high mercury oxidation efficiency of double-conversion double absorption (DCDA) technology, the proportions of Hg^{2+} in the exhaust flue gas was higher than Hg^0. Average emission factors of atmospheric mercury for these four tested gold smelters were calculated to be 1.7 kg of Hg per ton gold produced, and the national total amount of mercury emitted from large-scale gold smelting industry in the year of 2014 was estimated to 639.9 kg. However, mercury emission factors obtained from present study were much lower than that in previous research. Thus, with the exact emission factor obtaining from more specific field-test data and the detailed activity data information about gold smelting industry, an accurate inventory of mercury emissions to the atmosphere from large-scale gold smelters in China can be updated in the future.

Keywords: Mercury (Hg) emissions; Hg speciation; Ontario Hydro Method (OH Method); EPA Method 30B; Large-scale gold smelter.

INTRODUCTION

Mercury (Hg) has become one of the most widely-viewed global environmental pollution problems, because of its persistence, mobility and high bioaccumulation in the environment. There are three types of mercury speciation, including elemental gaseous mercury (Hg^0), oxidized gaseous mercury (Hg^{2+}), and particle-bound mercury (Hg_P). Hg^0 and Hg^{2+} are collectively known as total gaseous mercury. Three forms of mercury in the atmosphere can experience a complicated interchangeable process. In the process of photochemical reaction, Hg^0 can be oxidized to Hg^{2+} by some strong oxidizing substances, such as ozone, hydrogen peroxide, halogen, etc. Hg^0 and Hg^{2+} are easily adsorbed by

aerosol and formed Hg_P. Previous study found that Hg^0 could transport for very long distance, participate in global mercury cycle, and stay in the atmosphere for a long time. Hg^{2+} could be spread to hundreds of kilometer, freely soluble in water and fall to the ground with the precipitation. The deposition of Hg_P generally occurred in the vicinity of the emission sources (Liu and Luo, 2012).

Nonferrous metal smelting is one of the leading anthropogenic mercury emission sources and takes up a large proportion of the anthropogenic mercury emission inventory (Pacyna *et al.*, 2006; Wu *et al.*, 2006; Pirrone *et al.*, 2010; Tian *et al.*, 2015). Being bound with metal concentrate, mercury can be released to atmosphere in the smelting process, and has adverse effects on surrounding ambient air quality and public health. So far, several mercury contamination events due to gold mining have been reported, such as fatal accidents concerning mercury poisoning of miners in China, the Philippines, and Indonesia (Lin *et al.*, 1997; Lacerda and Salomons, 1998; Appleton *et al.*, 1999). In the Dexing city of Jiangxi province, where small-scale gold mining in China appears, high concentrations of mercury have been found in

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environmental samples and human hair and blood samples (Lin *et al.*, 1997; de Lacerda, 2003). In January 2013, the United Nations Environment Programme (UNEP) led the way in controlling and reducing global mercury emissions by issuing the Minamata Convention, in which made a detailed provisions on the limitation inventory of mercury emissions.

Formerly, small-scale and artisanal gold mining was the major source of atmospheric mercury due to the amalgam burning under generally uncontrolled conditions (de Lacerda, 2003). Since September 1996, most of the small-scale gold mining activities have been strictly prohibited by China's national environmental legislation. Recent investigation indicated that most of those small mines were shut off and production technologies of amalgam burning were eliminated gradually. Nowadays, the large-scale and centrally managed gold smelting industry develops rapidly and the new production technique and air pollution control technology are constantly emerging. Consequently, the current status of atmospheric mercury emissions from gold smelting industry in China will be changed and it needs to be further investigated and summarized.

In addition, according to previous study, approximately 46% of the total Hg emission in 2003 came from non-ferrous metal smelting in China, and the gold production source contributed 5.1% of the total Hg emission from non-ferrous metal smelting (Wu *et al.*, 2006). Pacyna *et al.* (2006) estimated the global anthropogenic Hg emission for the year of 2000. It showed that gold production source was responsible for 11.3% of the total global anthropogenic Hg emission and the Asian countries contributed about 19.0% to the global Hg emission from gold production sources in 2000. However, due to lack of actual field-tests data in China, the emission factor of Hg from gold smelting industry and its emissions to the atmosphere have still suffered from large uncertainties.

In this paper, the detailed information including gold distribution characteristics, mainstream production technology and atmospheric mercury emission control technology of large-scale gold smelting industry in China are reviewed. Based on literature and field investigations, onsite measurements in four large-scale gold smelters are carried out in Henan province of China. This paper focuses on the current atmospheric mercury emission situation and the mercury speciation distribution in flue gas associated with large-scale gold smelting industry of China.

METHODS AND DATA

Experimental Sites and Measurements

The four tested gold smelters are located in Henan Province, which is one of the major gold producing provinces of China. Air samples are collected in July 2014. As gold smelting is a relatively small industry and gold is a noble metal, the management of this kind of enterprise is very strict. Thus, there are no sampling points that cover all the inlets and outlets of the air pollution control devices (APCDs). Only one detection port occurs at the end of the flue after acid plant, which meets the environmental protection standards. The basic information about the four tested smelters is presented in Table 1. The sampling sites in the tested smelters are all located in the flue gas at the outlets of the acid plants. A sample is collected in about 50 minutes (min) and the sampling flow rate is about 0.8 L min^{-1}. Totally about 13 effective gaseous samples and 60 solid samples are collected and analyzed at four smelters.

The Ontario Hydro Method (OH Method) (ASTM, 2002) and EPA Method 30B (USEPA, 2002) are employed in the flue gas mercury measurements. Both of these two methods are standard methods that recommended by EPA and other energy agencies of United States. OH Method is used to analyze mercury speciation, including Hg^0, Hg^{2+} and Hg_P. EPA Method 30B, with relatively simple operation, is designed to measure the mass concentration of total gaseous mercury in flue gas, including Hg^0 and Hg^{2+}. It is intended for use as a reference method of the OH Method for the total gaseous mercury only under relatively low particulate conditions (i.e., sampling after all air pollution control devices). A series of preliminary investigations indicate that the flue gases at the sampling points of four tested gold smelters are cleaner and the particulate matter concentrations are relatively low. Therefore, in this study, OH Method is applied to analyze the concentrations of three mercury speciation, EPA Method 30B is used as a reference method of OH Method for the total gaseous mercury. To get a comprehensive understanding of the fate of mercury in gold smelters, both of raw materials (gold concentrate) and output samples (roasting dust) are collected simultaneously in all tested process.

All the samplings collected from flue gas and liquid samples by these two methods are analyzed in accordance with EPA Method 7470A (USEPA, 1994). The impinger solutions are recovered and analyzed with F732-V Intelligent Mercury Analyzer using Cold Vapor Atomic Absorption Spectrophotometry (CVAAS), which has a detection limit of 0.05 µg L^{-1}. In addition, most of the solid samples are analyzed in terms of EPA Method 7473 (USEPA, 1998).

QA/QC

In order to insure the reliability and accuracy of the results, the sampling and analysis are conducted strictly in accordance with the operating procedures in standard methods. Prior to each onsite measurement, the mercury sampling system is calibrated carefully with thoroughly cleaning of the sampling line, and a leak test is performed. Parallel samplings are conducted to make sure the validity of the results. Before the analysis of absorption samples, the F732-V is calibrated by drawing a standard curve with a correlation coefficient over 0.999. The analysis results are all over 10 times higher than detection limit of the instrument, and the samples with high mercury concentration were diluted before analysis. The blanks of all the reagents are low and deducted in the analysis. Two or more parallels of each sample are analyzed with the relative standard deviation less than 10%.

Activity Data

Activity data is obtained by conducting literature and field investigations and cross-checked for coincidence and a comprehensive analysis of annual statistics. The detailed

Table 1. Basic information about the four tested gold smelters.

	Smelter 1	Smelter 2	Smelter 3	Smelter 4
Roasting furnace scale (m^2)	120.00	24.85	30.86	16.00
Disposal capacity of gold concentrate (t d^{-1})	600	130	150	100
Flue gas volume (Nm^3 h^{-1})	40000	16000–17000	20000	8000–10000
Air pollution control devices in acid plant	flue gas scrubber (FGS) - electrostatic demister (ESD) - dehydration tower (DHT) - the conversion and absorption (C&A) - the adsorption of SO_2 with hydrogen peroxide (H_2O_2)			
Annual gold production (t)	8.0	2.3	1.2	1.3
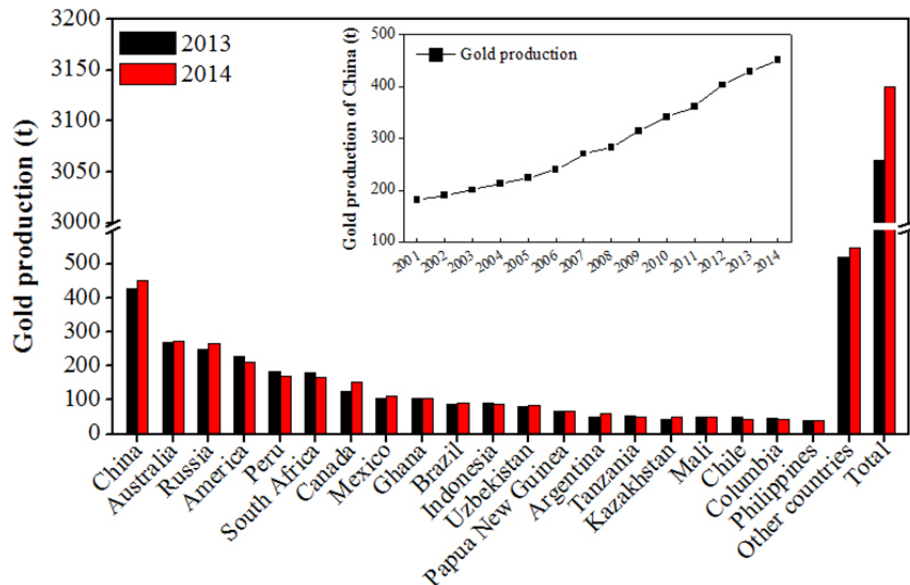
Fig. 1. Total gold production of global top 20 major producing countries, 2013–2014.

information includes gold distribution characteristics, mainstream production technology and atmospheric mercury emission control technology of large-scale gold smelters. Based on the collection and analysis of these activity data, it will be of value to a systematic and comprehensive understanding of the current status for gold smelting industry, as well as the further estimation of mercury emissions to the atmosphere from gold smelting industry in China.

RESULTS AND DISCUSSION

Gold Production and Its Distribution Characteristics

The development of China's gold industry started in the early days of political reform and globalization. During the period of 1978–2014, the annual growth rates of gold production were about 9.1%. While in the recent years, an increasing base of gold production entered into a relatively stable growth stage. By the end of 2014, the gold production of China has reached to 451.80 tons (t), reflecting an annual growth ratio of 7.3% compared with the year 2001 (see Fig. 1). It could be seen in Fig. 1, the global gold production amounted to about 3133.00 t in 2014, showing a growth ratio of 2.4% compared with that in the year of 2013. The total gold production of global top 20 major producing countries reached to 2575.20 t, accounting for 82.2% of the global production (China Gold Association,

2015). The gold production of China in 2014 took up 14.4% of the world's total gold production (see Fig. 1), ranking first in the world for eight consecutive years.

The distribution of gold production in China is countrywide but occurs in select provinces in accordance with geologic controls on deposit formation. In 2014, except for Beijing, Tianjin and Chongqing areas, gold production reports of other regions have already been presented. The provincial distribution of gold production in the year of 2014 is illustrated in Fig. 2. It is found that the gold production of Shandong province is responsible for one third of the national total. The total gold production of Shandong, Henan and Jiangxi province are more than 50% of the national total. In addition, Shandong and Henan regions are the two traditional provinces of gold production, especially for Shandong province, which is the first province that the gold production exceeded one hundred tons. While Jiangxi, Yunnan and Inner Mongolia are the three rookie provinces that have been emerged as gold producers this century.

The regional distribution feature of gold production in China is presented in Table 2. In 2014, the final gold production of Zhaoyuan City for Shandong Province ranks the first, follows by Lingbao and Laizhou Cities. The total gold production of national top 10 major producing counties/cities reaches to 127.90 t, accounting for 28.3% of the national total production.
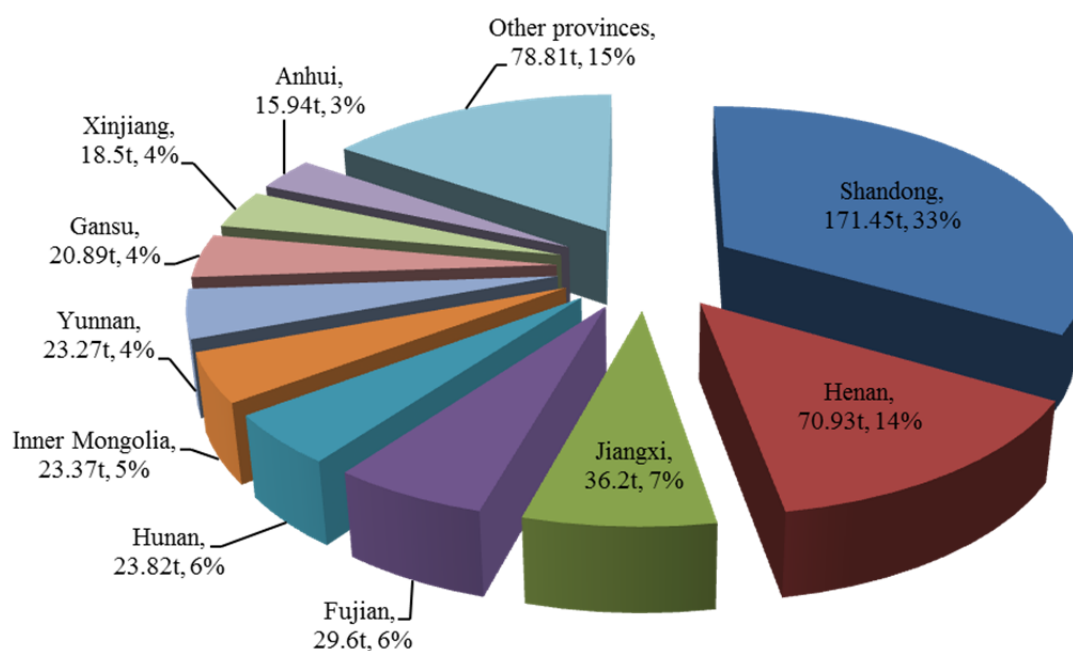
Fig. 2. Provincial distribution characteristic of gold production in China, 2014.

Table 2. Total gold production of top 10 major producing counties/cities of China, 2014.

Rankings	City/County	Final gold production (ton)
1	Zhaoyuan City of Shandong Province	32.97
2	Lingbao City of Henan Province	27.11
3	Laizhou City of Shandong Province	20.15
4	Shanghang County of Fujian Province	12.25
5	Tongguan County of Shaanxi Province	7.87
6	Zhenfeng County of Guizhou Province	7.14
7	Song County of Henan Province	6.25
8	Wulate Midum Banner in Inner Mongolia	5.10
9	Heqing County of Yunnan Province	5.04
10	Aohanqi Banner of Inner Mongolia	4.02

Mainstream Production Technology of Large-Scale Gold Smelting Industry in China

Since September 1996, most of the small-scale gold mining activities have been strictly prohibited by national environmental legislation of China. So a large amount of small-scale gold mines were shut off and production technologies of amalgam burning were eliminated gradually. The gold smelting industry continues to develop rapidly with advances in new production techniques and technologies. Consequently, there will be equal amendments to the atmospheric mercury emission limits in China.

The mineralogical characteristics of ore, containing its composition and structural properties, are important factors that influenced the selection of gold production technology. According to the processing characteristics of gold ore, it can be divided into manageable gold ore and refractory gold ore. Refractory gold ore is a kind of ore that the gold recovery rate of less than 80% only by leaching technology without pretreatment process (Nieves and Francisco, 1994). Thus, pretreatment is a necessary production process for refractory gold ore. Nowadays, the reserves of refractory

gold ore account for 60% of the world's total gold reserves, and also in China. One third of the world's gold productions come from refractory gold ore. Therefore, the mainstream production technology of large-scale gold smelting industry in the world includes ore dressing, pretreatment, leaching, extraction and recovery, refining process (see Fig. 3). The common methods of pretreatment for refractory gold ores before conventional cyanide leaching contain roasting, chemical oxidation and biological oxidation. Roasting technology is the most widely used in the world. Many countries and regions, such as Fairview in South Africa, Getchell in America, Campbell in Canada, have all established the roasting factory of gold concentrate (China Gold Association, 2015). And, crucially, almost all of the atmospheric mercury during gold smelting process will be released from roasting pretreatment process.

Status of Atmospheric Mercury Emission Control Technology in Large-Scale Gold Smelting Industry

Mercury associated with gold ore will be released to the flue gas during the smelting process, and has adverse
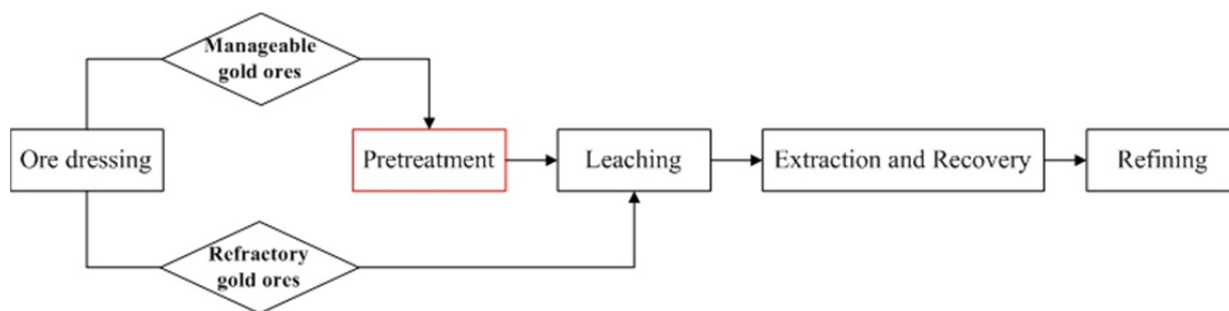
Fig. 3. Mainstream production technology of large-scale gold smelting industry in China.

influence on the local and regional environment, as well as human health. The main process of large-scale gold smelting industry for mercury emissions is the roasting pretreatment unit. The flue gas from this process first goes through the exhaust-heat boiler and then the particle control devices. A cyclone dust collector and electrostatic precipitator (ESP) are used in all large-scale gold smelters. After dust collector device, the flue gas with high SO_2 concentration enters into the acid plant. During the acid plant, the flue gas generally passes through the flue gas scrubber (FGS), electrostatic demister (ESD), dehydration tower (DHT), the conversion and absorption (C&A) process, and process for the adsorption of SO_2 with hydrogen peroxide (H_2O_2) successively. A previous study in a Chinese zinc smelter showed that FGS and ESD device in the tested acid plant could remove 17% and 30% of the total mercury, and acid plant with C&A processes displayed average mercury removal efficiency up to 97.4% (Wang *et al.*, 2010a). Zhang *et al.* (2012) investigated that the double-conversion double absorption (DCDA) process in acid plant had mercury removal efficiency of over 99.0%. The catalytic mercury oxidation process took place simultaneously when SO_2 converted to SO_3 with the existence of a catalyst in the conversion tower (Kamata *et al.*, 2008; Lee and Bae, 2009). The oxidized mercury could easily be absorbed in the sulfuric acid inside the absorption tower. Therefore, acid plants in gold smelters have remarkable co-benefit on mercury removal. Industrial sulfuric acid obtained from acid plant is mainly used in fertilizer, metallurgy and metal processing industry, petroleum industry, chemical industry, and so on.

In general, the removal of mercury inside the acid plant can be normally divided into two stages. In the first stage, is the FGS and ESD, mercury in the flue gas enters into the waste water. In the second stage, is the DHT and C&A process, mercury in the flue gas goes into the sulfuric acid product. The removal efficiency of each stage is determined by the speciation of mercury in the flue gas going into the acid plant.

Concentrations and Speciation of Mercury in Tested Gold Smelters

The mercury contents in four tested gold smelters are summarized in Table 3. As can be seen, mercury contents in gold concentrate for four smelters ranges from 2.84–5.06 μg g^{-1}. The highest one is found in Smelter 4, leading to very high total mercury concentrations in the flue gas

after acid plants. Thus, mercury concentration in input materials (gold concentrate) being processed is a key factor affecting the mercury concentrations in exhaust flue gases. Additionally, one main byproduct from gold smelter is roasting dust. The mercury contents in roasting dust are in the range of 0.26–3.06 μg g^{-1}. Special concerns should be paid to these byproducts, which would be potential mercury emission sources to soil if not properly disposed.

The configuration of APCD in smelting process is another key factor affecting the mercury concentrations in exhaust flue gases. Because of adopting a variety of efficient APCDs, such as ESP, FGS, ESD, DHT and C&A process, the emissions of total mercury in the exhaust flue gas are relatively low. The concentrations of mercury in the flue gas at the outlets of acid plants range from 1.52–32.81 μg m^{-3}, much lower than the currently implemented mercury emission standard for GB 9078-1996 Grade II level areas of 1000 μg m^{-3} (MEP, 1996). The lowest mercury concentrations are occurred in Smelter 1, which is likely due to the cumulative removal by adopting the devices of two-stage cyclone dust collector, quadrupole ESP, three-stage FGS and two-stage ESD. When combining, it created a more efficient cooperative removal efficiency of mercury in flue gas. By comparison of these two testing methods, the total gaseous mercury concentrations detected by OH Method are 0.12–2.43 μg m^{-3}, which are about 42.9%–95.3% lower than those detected by EPA Method 30B. Due to the differences of sampling and analysis principle, comparative study of these two methods is needed in the future research.

Nowadays, with the development of production technology and pollution control technology, the current implementation of the mercury emission standard has been conservative. Therefore, it is urgent to amend the air pollution emission limits of gold smelting industry in order to meet the requirements of the national environmental protection regulations.

Moreover, as can be seen in Table 3, the speciation split of mercury in the exhaust flue gas after acid plants are similar. The proportions of Hg_p in the exhaust flue gas for four tested gold smelters are highest within the range of 90.6%–99.0% for the total mercury concentrations. While the proportions of Hg^{2+} and Hg^0 for the total concentrations of mercury in the exhaust flue gas are in the range of 0.7%–9.0% and 0.3%–1.8%, respectively. In addition, due to apply the double-conversion double absorption (DCDA) process in four tested smelters, it displays high mercury

Table 3. Concentration and speciation of mercury in the four tested gold smelters.

	Smelter 1	Smelter 2	Smelter 3	Smelter 4
Hg contents in input and output samplings, $\mu g \ g^{-1}$				
Gold concentrate (input)	2.84 ± 1.54	4.95 ± 3.17	2.79 ± 1.67	5.06 ± 2.97
Roasting dust (output)	0.26 ± 0.09	2.93 ± 1.27	0.71 ± 0.19	3.06 ± 1.58
Hg concentrations in flue gas after acid plant, $\mu g \ m^{-3}$				
OH Method	1.52 ± 0.21	8.60 ± 1.32	26.02 ± 1.58	32.81 ± 1.67
EPA Method 30B	0.75 ± 0.34	2.39 ± 0.33	5.43 ± 0.61	3.41 ± 0.74
Hg speciation in flue gas after acid plant, %				
Hg_p	92.1	93.7	90.6	99.0
Hg^{2+}	6.1	5.3	9.0	0.7
Hg^0	1.8	1.0	0.4	0.3

oxidation efficiency (Wang *et al.*, 2010b; Zhang *et al.*, 2012). Thus, the proportions of Hg^{2+} in the exhaust flue gas after acid plant is higher than Hg^0.

A previous study (Kim *et al.*, 2011) found that a flue gas scrubber after the acid plant had high removal efficiency for Hg^{2+}, because Hg^{2+} was water soluble (Wang *et al.*, 2010b). So the low concentration of mercury in non-ferrous metal smelters might be resulted from the influence of the flue gas scrubber. Therefore, the configuration of APCD plays an essential role in affecting the mercury concentrations in exhaust flue gases. Further installation of the flue gas scrubbing after the acid plant may be a possible method for heightened mercury removal in the flue gas of gold smelters.

Estimating Mercury Emissions from Large-Scale Gold Smelters in China

Atmospheric mercury emissions from large-scale gold smelters are based on gold productions multiplied by mercury emission factors. The emission factors of mercury are calculated in the forms of mercury emissions per ton gold produced. As a result of limited information and lack of field tests on mercury emissions for large-scale gold smelting industry in China, some emission factors of atmospheric mercury in previous studies for this source category have to be cited from some published literatures and only with nationally averaged levels. Pacyna *et al.* (2006) reported that the global average emission factor of mercury for large-scale gold production is 0.5 g g^{-1} gold mined, and this data is also used in a recent study to estimate the global mercury emissions to the atmosphere from large-scale gold production industry (Pirrone *et al.*, 2010). Streets *et al.* (2005) calculated the anthropogenic mercury emissions from large-scale gold production industry in China by using the average mercury emission factor of 0.79 t t^{-1} Au. Based on the activity data and the mercury emission factor of 0.79 t t^{-1} Au, Wu *et al.* (2006) estimated that the mercury emissions from large-scale gold production industry in the year of 2003 were 16.2 t, contributing 2.3% of the total anthropogenic mercury emissions in China. However, in our present study, it can be seen that mercury emission factors for these four tested gold smelters are calculated to be 0.5–3.4 kg of Hg emitted to the environment for every ton of gold produced, with an average value of 1.7 kg t^{-1}.

Notably, the emission factors of atmospheric mercury for large-scale gold smelting industry used in previous studies are much higher than that in our present research. On account of shutting off many small-scale gold mines in China since 1996 and emerging lots of new production techniques and technologies, the actual emissions of atmospheric mercury from large-scale gold smelting industry might be decreased. Using the field-tested average mercury emission factor of 1.7 kg t^{-1} and the available gold production figures from countries in the year of 2014, the total emissions of mercury from four tested gold smelters are calculated to be 14.7 kg, and the annual mercury emissions of large-scale gold smelting industry in China are estimated to 639.9 kg according to an operational time of 320 day $year^{-1}$, and 24 hour day^{-1} for each gold smelter. In future, with the emission factor of mercury obtaining from lots of specific field-test data and the detailed activity data information about gold smelting industry, an accurate inventory of mercury emissions to the atmosphere from large-scale gold smelters in China can be updated.

CONCLUSIONS

In this study, current status and atmospheric mercury emissions associated with large-scale gold smelting industry in China were investigated and measured. Our results showed that the mineralogical characteristics of ore were important factors that influenced the selection of gold production technology. The mainstream production technology of large-scale gold smelting industry included ore dressing, pretreatment, leaching, extraction and recovery, refining process. Almost all of the atmospheric mercury would release from the process of roasting pretreatment before acid plant. Acid plants in gold smelters had significant co-benefit on mercury removal. Mercury concentration in input materials (gold concentrate) and the configuration of APCD in smelting process were two key factors affecting the mercury concentrations in exhaust flue gases. All the concentrations of mercury in the flue gas at the outlets of acid plants for the four tested gold smelters were in the range of 1.52–32.81 $\mu g \ m^{-3}$, which could meet the emission standard of GB 9078-1996 Grade II levels. The proportions of Hg_p in the exhaust flue gas were highest within the range of 90.6%–99.0% for the total mercury concentrations. The proportions of Hg^{2+} in the exhaust flue gas after the acid plant were higher than Hg^0, due to the high mercury oxidation efficiency in double-conversion double absorption (DCDA) process.

Atmospheric mercury emission factors for these four tested gold smelters were ranged from 0.5–3.4 kg of Hg emitted to the environment for every ton of gold produced, with an average of 1.7 kg t^{-1}, which was much lower than that in previous research. Through a rough calculation, national mercury emissions from large-scale gold smelting industry in the year of 2014 might be 639.9 kg. Henceforth, after a series of specific field-test about large-scale gold smelting industry in the future, an accurate inventory of mercury emissions to the atmosphere from large-scale gold smelters in China can be updated.

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