



# Mercury emission and speciation from industrial gold production using roasting process



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## ABSTRACT

Gold production has been identified as an important source of anthropogenic mercury emissions. Few measurements have been conducted on the mercury emission in the industrial gold production. In this study, field measurements on mercury concentration and speciation profile in the roasting flue gas were conducted in a two-stage roasting gold smelter, and the corresponding mercury emission factors were obtained using mass balance analysis. The average mercury concentration in the feed gold concentrates was 730.0 µg/kg, and the daily input mercury was 94.9 g in this research. In the roasting procedure, 38.4%, 27.3% and 9.0% of input mercury was removed into the sulfuric acid, contaminated water and arsenic, respectively, while 22.8% of input mercury flowed into the cyanidation-refining procedure within the roasting residue and the dust collected from the gas cooling tower and the electrostatic precipitator. Finally, 2.3% and 5.9% of input mercury was emitted into the atmosphere with the roasting flue gas and the refining flue gas. Mercury concentration in the roasting flue gas was 4.02 µg/m<sup>3</sup>, where the proportion of Hg<sub>p</sub>:Hg<sup>2+</sup>:Hg<sup>0</sup> was 11:57:33. The balance calculation results indicated that the atmospheric mercury emission factor was  $2.27 \times 10^{-3}$  g mercury/g gold produced for the tested smelter. Both the emission factor and mercury removal efficiencies of air pollution control devices are useful for development of a more accurate emission inventory.

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## 1. Introduction

Atmospheric mercury (Hg) has aroused public concern due to its health effects on humans and other organisms, and gold production has always been considered as a large source of anthropogenic Hg emissions (Lacerda, 1995, 1997). Based on the 2010 Hg emissions inventory, the amount of atmospheric Hg emission from gold mining, including artisanal and small-scale gold mining (ASGM) and large-scale gold production (LSGP, also known as industrial gold production), reached 824.3 t, taking the largest proportion (42.1%) of the global anthropogenic Hg emissions (AMAP/UNEP, 2013). Much of the historic emission of Hg was reported from ASGM, where Hg was employed for gold recovery from milled ore via amalgamation (AMAP/UNEP, 2013). This gold recovery method, however, has been effectively ceased in many countries due to its dramatic high Hg emission, and it has also been forbidden by law in China since 1997. Therefore, more attention should be paid on the Hg emission from LSGP in China in the future (Zhang et al., 2015).

In previous studies, great uncertainties existed in Hg emission factors (EFs) of LSGP. In general, there are four conclusive factors associated with Hg EF – the Hg concentration of feed gold concentrates, the Hg distribution factor to the flue gas, the type of air pollution control devices (APCDs) combination applied and the Hg removal efficiency of a certain type of APCDs combinations, and all of them are likely to vary considerably. Take the Hg concentration of gold concentrates for example, Fang et al. (2004) indicated that the gold concentrates generally contained 1000 µg/kg or more Hg by analyzing over 4000 gold concentrates samples in Taiwan, and the highest content was over 37,500 µg/kg. However, based on 43 concentrates samples in Shihu gold mine in China, the lowest Hg concentration was only 1.76 µg/kg, reported by Li (1990). Moreover, the Hg distribution factor to the flue gas is determined by the smelting technology employed. Obviously, compared with the heap leaching, the cyanidation with roasting pretreatment has more Hg release point due to thermal pretreatment process. Finally, based on the field experiments conducted in the non-ferrous metal smelting factories, it has been proved that the Hg of flue gas could be removed to certain extent with the utilization of the APCDs with different Hg removal efficiencies ranging from 2.4% to 99.2% (Zhang et al., 2012; Wu et al., 2012). Nevertheless, the aforementioned information was barely considered in previous researches. For example, in the research conducted by Argonne National Laboratory and Tsinghua University,

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0.79 g/g (g Hg emitted/g gold produced) was chosen as the LSGP Hg EF for lack of field measurements, and it was obvious that such Hg EF was overestimated since it was originally used to estimate the Hg emission from ASGM in China (Dai et al., 2003; Jiang, 2004; Streets et al., 2005; Wu et al., 2006). Pacyna et al. (2006) presented the LSGP Hg EF of 0.5 g/g in the Atmospheric Emission Inventory Guidebook (UN ECE, 2000) with no experiment specifications. UNEP estimated that the Hg EF of LSPG were 0.025–0.027 g/g and 0.055 g/g to calculate the Hg emission inventories of 2005 and 2010, respectively (AMAP/UNEP, 2008, 2013; Pacyna et al., 2010; Streets et al., 2011). However, only the distribution factor to air (0.04) and the Hg concentration of gold concentrates (55,000 µg/kg) were provided based on US national data, indicating that it is related to the specific technologies employed in the United States (AMAP/UNEP, 2013), and it failed to provide any specific information associated with the APCDs.

Since little information of field measurements on Hg emission of the industrial gold production is available, the main purpose of this work is to update the EF to the atmosphere in industrial gold smelter based on field measurements. In this paper, the concentration and speciation of Hg released from the roasting flue gas has been tested and the Hg removal efficiencies of the APCDs have been comprehensively discussed. Moreover, the behavior of Hg in the roasting flue gas and the fate of Hg through the whole smelting process have been systematically analyzed.

## 2. Experimental methods

### 2.1. Tested smelter

In China, the three technologies used in LSGP are heap leaching, roasting (including one-stage and two-stage roasting) cyanidation and bio-oxidation cyanidation, the latter two of which are utilized to deal with refractory gold ore. Compared with the heap leaching and bio-oxidation cyanidation, the roasting cyanidation has additional thermal pretreatment process, which is considered to be the largest Hg emission sources of LSGP (Miller and Jones, 2005). Besides, the processes in the heap leaching technology are included in the roasting cyanidation, and based on our investigation, bio-oxidation cyanidation is merely employed by one factory in Xinjiang province in China. Thus, in

this research, a two-stage roasting smelter in Shandong province, which has the biggest gold production in China, was chosen for field measurements.

The process flow diagram of tested smelter is illustrated as Fig. 1. In this research, the whole smelting process is divided into two procedures – roasting and cyanidation-refining. The roasting procedure includes processes from the roasting to the flue gas emitting (processes illustrated in the green dot frame in Fig. 1); while the cyanidation-refining is the procedure during which the gold-bearing material, including the residue from the roasterII as well as the dust from U-shape gas coolerII (UGCII), gas cooling tower (GCT) and electrostatic precipitator (ESP), is cyanided to produce dore (processes illustrated in the blue dot frame in Fig. 1).

The operation temperature of the first and second roasting process is 550 °C for 1 h and 600 °C for half an hour, respectively. After the flue gas from two roasters passes through the corresponding UGCs, they merge together to go through the GCT and ESP successively. A small quantity of coarse particles collected by UGCI and the roasting residue of the roasterI go into the roasterII for further roasting. The temperature of the roasting flue gas at the inlet of the quench tower (QT) and the outlet of the fiber filter dust collector (FF) is 376 °C and 158 °C, respectively, so arsenic trioxide in the flue gas is condensed due to this dramatically cooling and then collected by FF. Recycled dilute sulfuric acid is utilized to clean the flue gas in venturi tube (VT) and packet tower (PT), and then electrostatic demister (ESD) is employed to remove the water vapor from the gas stream. The contaminated water and contaminated acid sludge is discharged after the roasting flue gas goes through VT, PT and ESD, and then the flue gas with high SO<sub>2</sub> concentration enters into the acid plant (AP). In the AP, the flue gas goes through the dehydration tower (DHT) and the double-conversion double-absorption (DCDA) process to produce H<sub>2</sub>SO<sub>4</sub>. In the cyanidation procedure, the gold-bearing material will dissolve in the NaCN solution to generate the pregnant solution with gold and other metallo-cyanide complexes (including Hg), in which the metals will be precipitated on zinc dust. Eventually, the zinc precipitate taken from the filter presses is refined at high temperature (over 1200 °C) to produce dore, during which all the available Hg will be released into the atmosphere since there is no APCD applied in tested smelter.

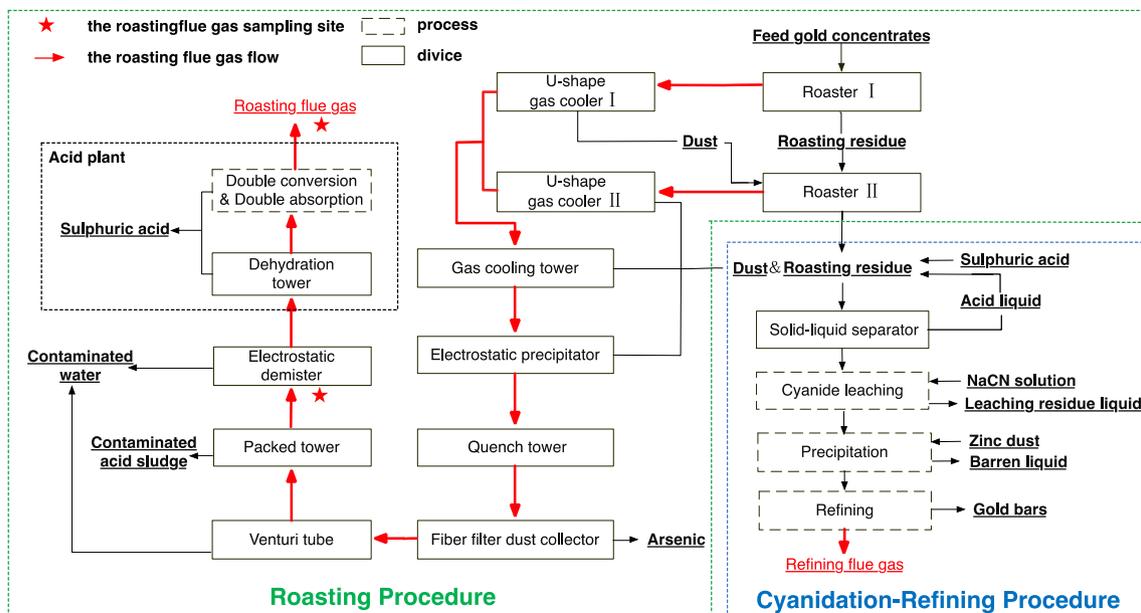


Fig. 1. Flow diagram in a two-stage roasting gold smelter.

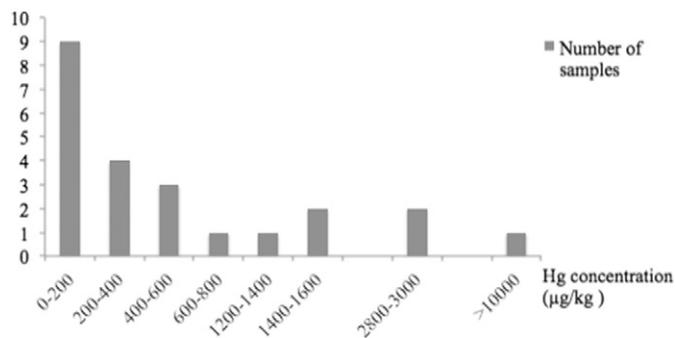


Fig. 2. The Hg concentration of 23 gold concentrates from 9 provinces in China.

## 2.2. Sampling and analytical methods

The flue gas sampling sites, subject to test feasibility, cannot completely cover all the inlets and outlets of the APCDs. Thus, based on the field investigation, one sampling site has been located at the inlet of the ESD and the other has been located at the outlet of the AP, as shown in Fig. 1. The Ontario Hydro Method (OH method) (ASTM, 2002) was employed to measure the Hg speciation profile in the flue gas of two sampling sites. Method 30B (US EPA, 2011) was applied to quantify the total Hg in the flue gas at the outlet of the AP for comparison. The method of mass balancing was also adopted to get a comprehensive understanding of the fate of Hg in the tested smelter. During the flue gas sampling, the samples of both the input (feed gold concentrates and zinc dust) and output (roasting residue, dust captured by the GCT and ESP, arsenic, contaminated water, contaminated acid sludge, sulfuric acid, cyanide leaching residue and barren liquor) were collected in the tested smelter together with the mass flow data to calculate the Hg flow.

Liquid samples (including all the impinger solutions obtained from flue gas sampling procedure) were analyzed according to the United States Environment Protection Agency (US EPA) Method 7470A (US EPA, 1994). The impinger solutions were recovered by  $\text{SnCl}_2$  and analyzed with F732-V Intelligent Mercury Analyzer based on Cold Vapor Atomic Absorption Spectrophotometry (CVAAS), whose detection limit is 0.05 µg/L. As for the solid samples, Method 7473 (US EPA, 1998) was adopted to determine the Hg concentrations. The solid samples were air dried and grounded into 100 meshes for further analysis

with Lumex RA915+ based on Cold Vapor Atomic Fluorescent Spectrophotometry (CVAFS), whose detection limit is 0.5 µg/kg. The arsenic sample with high toxicity was first digested for 30 min by aqua regia at ambient temperature in an ultrasonic water bath, and then a suitable aliquot of the digested sample solution was measured using CAVVS.

## 2.3. Quality assurance and quality control

To improve the accuracy of the results and reduce the accidental error for each measurement, we strictly followed the operating procedures in standard methods for sampling and analysis. Parallel samplings were conducted to ensure the validity of the results. For OH method and 30B method, four and three valid test results were obtained under the stable operating condition, respectively, with the relative standard deviation < 20%. Each of the seven impingers in the sampling train of the OH method was recovered and analyzed separately. Prior to the analysis of absorption samples and other liquid samples, the F732-V was calibrated by drawing a standard curve with a correlation coefficient over 0.995. The analysis results were all over 10 times higher than detection limit of the instrument, and the samples with high Hg concentration were diluted before analysis. The blanks of all the reagents were low and deducted in the analysis. The standard soil sample (Hg% is  $0.029 \pm 0.003$  µg/kg) was used to draw a standard curve with a correlation coefficient over 0.999 before analyzing the solid sample with Lumex RA915+. Three or more parallels of each sample were analyzed with the relative standard deviation < 10%. The standard reference materials from the National Institute of Standards and Technology (NIST), USA (1632c, coal, NIST, Gaithersburg, USA) and National Research Center for Certified Reference Materials (NRCCRM), China (GSS-5, soil, NRCCRM, Beijing, China) were used to validate the test methods in this study.

## 3. Result and discussion

### 3.1. Hg in gold concentrates

During the two-day flue gas sampling period, two feed gold concentrate samples were collected daily to analyze the Hg concentrations. The average Hg concentration in the feed gold concentrates was 730.0 µg/kg. Also, as shown in Fig. 2, 23 gold concentrate samples used in this plant came from 9 provinces in China were analyzed for the Hg concentrations. The wild range of the Hg concentration is from 5.9 to

Table 1

Mass distribution of mercury in a two-stage roasting gold smelter in Shandong Province in China, chosen as the tested smelter.

Roasting													
	Input			Output									
	Feed gold concentrate	Total (g)	Roasting residue	Dust from UGC II	Dust from GCT	Dust from ESP	Arsenic	Contaminated water	Sulfuric acid	Contaminated acid slime	Roasting flue gas	Total (g)	
Hg concentration (µg/kg)	730.0	94.9	220.0	Nd	90.2	142.9	567.3	52.5 (µg/L)	174.6	0.5	4.2 (µg/m <sup>3</sup> )	94.2	
Mass flow (t/d)	130		64	<0.01	32	32	15	680 (m <sup>3</sup> /d)	150	<0.1	525,000 (m <sup>3</sup> /d)		
Hg mass flow (g/d)	94.9		14.1	Nd	2.9	4.6	8.5	25.7	36.2	0.0	2.2		
Percentage (%)	100%		15.0%		3.1%	4.9%	9.0%	27.3%	38.4%	0%	2.3%		
Recovery rate	99%												
Cyanidation-refining													
	Input								Output				
	Roasting residue	Dust from UGC II	Dust from GCT	Dust from EP	Sulfuric acid	NaCN solution	Zinc dust	Total (g)	Cyanide leaching residue	Barren liquor	Gold	Refining flue gas	Total (g)
Hg concentration (µg/kg)	220.0	Nd	90.2	142.9	108.1	Nd	Nd	21.7	94.1	0.5	Nd	Nd	16.1
Mass flow (t/d)	64	<0.01	32	32	<0.5	Nd	Nd		150	900	1.16 (t/y)	Nd	
Hg mass flow (g/d)	14.1	Nd	2.9	4.6	0.1	Nd	Nd		15.6	0.5	Nd	Nd	
Percentage (%)	65.0%		13.4%	21.2%	0.5%				95.0%	5.0%			
Recovery rate	72%												

Nd – no data.

**Table 2**  
Mercury removal efficiencies of APCDs installed in the tested smelter located in Shandong Province in China.

APCDs	Mercury removal efficiency (%)
Gas cooling tower ( $\eta_{GCT}$ )	4%
Electrostatic precipitator ( $\eta_{ESP}$ )	6%
Fiber filter dust collector ( $\eta_{FF}$ )	12%
Venturi tube ( $\eta_{VT}$ )	7%
Electrostatic demister ( $\eta_{ESD}$ )	31%
Acid plant ( $\eta_{AP} = \eta_{DHT\&DCDA}$ )	96%

11,207.9  $\mu\text{g}/\text{kg}$ . Hg concentrations in 16 samples (70% of all the samples) were below 600  $\mu\text{g}/\text{kg}$ , but the average Hg concentration is 1103.3  $\mu\text{g}/\text{kg}$  due to the existence of gold concentrates with high Hg concentration.

### 3.2. Hg release rate in roasting process

As shown in Table 1, the daily input Hg is 94.9 g within the feed gold concentrates, 14.1 g of which is left in the residue, indicating that only 85.1% Hg in the gold concentrates will evaporate into the flue gas. Wu et al. (2012) reported that the release rates of Hg into flue gas in various smelting processes of primary zinc, lead and copper smelters in China are in the range of 97.7–99.4%. The highest operation temperature of the gold ore concentrates roasting smelters is over 600 °C, and thus most of the Hg should go into the flue gas after roasting same as other nonferrous metals smelting. The low Hg release rate in the tested smelter may relate to the special chemical properties of Hg and gold. According to the research conducted by Li (Li, 1990), Hg at certain chemical speciation in the gold ores will sublime only on the condition that the temperature exceeds 780 °C.

### 3.3. Hg removal efficiencies of APCDs

The Hg concentration in all the collected samples were tested and shown in Table 1, and the Hg removal efficiencies of APCDs were calculated though the balance calculation and listed in Table 2. To be specific, the Hg removal efficiencies of GCT and ESP, where Hg is removed with the collection of the dust, are 4% and 6%, respectively. The dust removal efficiencies of GCT and ESP are 85% and 98%, respectively, and thus, particulate mercury ( $\text{Hg}_p$ ) in the roasting flue gas should be almost removed. The quench tower (QT) is designed to decrease the temperature of the roasting flue gas from 375 °C to below 130 °C by spraying circulating cooling water. The cooling water quantity is about 1  $\text{m}^3/\text{h}$  and there is no by-product from the QT. As a result of the dramatic temperature reduction, arsenic trioxide ( $\text{As}_2\text{O}_3$ ) in the roasting flue gas condensed and then is captured by the FF. Meanwhile, part of Hg vapor is condensed into arsenic due to the tremendous temperature drop. The total Hg content decreased by 12% after the roasting flue gas goes through the FF. These Hg would be elemental mercury ( $\text{Hg}^0$ ) and oxidized mercury ( $\text{Hg}^{2+}$ ), as  $\text{Hg}_p$  has been almost removed by the GCT and ESP. In the tested smelter, there are two APCDs (VB and ESD) where the contaminated water is released and the properties of the roasting flue gas at their inlets have been detailed in Table 3. Based on the results of OH method (as shown in Table 4), the Hg concentration of the roasting flue gas at the inlet of ESD is 89.4  $\mu\text{g}/\text{m}^3$  with a gas

**Table 3**  
The properties of the roasting flue gas at inlets of APCDs.

Parameter	VT	PT	ESD	DT
Flow rate <sup>a</sup> ( $\text{km}^3/\text{d}$ )	914	929	642	656
Temperature (°C)	100	68.64	35	35
Pressure (kPa)	95.83	93.63	92.33	90.33
Hg concentration ( $\mu\text{g}/\text{m}^3$ )	70.17	Nd	89.36	Nd

Nd - No data.

<sup>a</sup> Flow rate - Under working condition.

**Table 4**  
Hg concentration and speciation in the roasting flue gas ( $\mu\text{g}/\text{m}^3$ ).

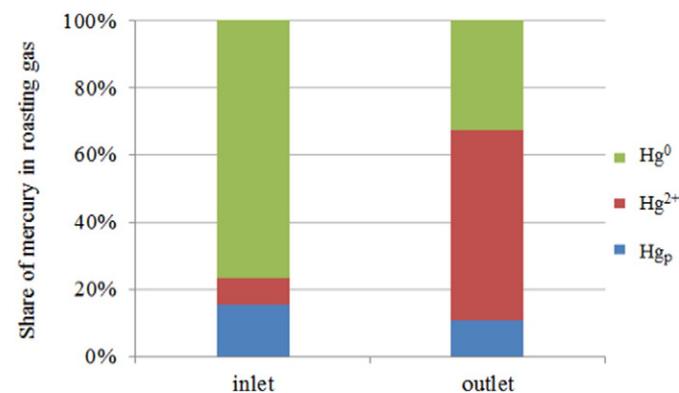
	OH method			30B method	
	$\text{Hg}_p$	$\text{Hg}^{2+}$	$\text{Hg}^0$	$\text{Hg}_t$	$\text{Hg}_t$
Before ESD	13.8 (15%)	7.2 (8%)	68.4 (77%)	89.4	-
After acid plant	0.5 (11%)	2.4 (57%)	1.4 (32%)	4.2	4.0

flow rate of 642  $\text{km}^3/\text{d}$ , and therefore the total amount of Hg enters into ESD is 57.5 g/d. Moreover, since the amount of Hg flowing into the sulfuric acid and the air is 36.2 g/d and 2.2 g/d, respectively, the Hg removed by ESD into the contaminated acid is calculated to be 19.1 g/d. Rest of Hg in the contaminated acid is removed by VT, which is 6.6 g/d. Thus, the Venturi process removes 7% of the Hg, which is absorbed into the contaminated water, and the similar method for Hg removal is also applied in ESD with the Hg removal efficiency of 31%. The DCDA process, which is the last procedure for Hg removal, is capable of eliminating 96% of the Hg in the roasting gas into the sulfuric acid.

As discussed above, the  $\text{Hg}_p$  has almost been removed when the roasting flue gas goes through the GCT and ESP. According to the results of the OH method (as shown in Table 4), at the inlet of the DHT, the proportion of the  $\text{Hg}_p$  of the total Hg ( $\text{Hg}_t$ ) is 14%. The element composition of the particulate matter on the filters has been analyzed by ICP-MS, and the result demonstrates that the average As content is 21%. So the particulate matter captured at the inlet of the ESD probably is  $\text{As}_2\text{O}_3$  in the roasting flue gas, and the Hg in the particulate matter is  $\text{Hg}^0$  and  $\text{Hg}^{2+}$  condensed with the arsenic.

### 3.4. Hg concentrations and speciation in roasting flue gas

The Hg concentrations in the roasting flue gas emitted to the air from OH method and 30B method were given in Table 4, which are 4.2 and 4.0  $\mu\text{g}/\text{m}^3$ , respectively. The Hg speciation profile in the flue gas varied a lot between the two sampling sites, as showed in Fig. 3. The proportions of  $\text{Hg}_p$ ,  $\text{Hg}^{2+}$  and  $\text{Hg}^0$  in the roasting flue gas before the ESD are 15%, 8% and 77%, respectively. Through the ESD and DCDA,  $\text{Hg}_p$  and  $\text{Hg}^{2+}$ , which can be captured by water, as well as a small proportion of  $\text{Hg}^0$  is removed into the contaminated water and sulfuric acid. The content of  $\text{Hg}_p$  in the flue gas drops from 13.8  $\mu\text{g}/\text{m}^3$  to 0.5  $\mu\text{g}/\text{m}^3$ . The DCDA process has strong Hg oxidation efficiency to oxidize  $\text{Hg}^0$  to soluble  $\text{Hg}^{2+}$ , and  $\text{Hg}^{2+}$  is then removed by  $\text{H}_2\text{SO}_4$  (Song, 2010). Therefore, the concentration of  $\text{Hg}^0$  in the flue gas after the acid plant drops from 68.4  $\mu\text{g}/\text{m}^3$  to 1.4  $\mu\text{g}/\text{m}^3$ . The ESD and DCDA process removes 96%, 67% and 98% of  $\text{Hg}_p$ ,  $\text{Hg}^{2+}$  and  $\text{Hg}^0$ , respectively. The removal efficiency of  $\text{Hg}^{2+}$  is lower compared with the removal efficiency of  $\text{Hg}_p$  and  $\text{Hg}^0$  due to the growing concentration of  $\text{Hg}^{2+}$  in DCDA process.



**Fig. 3.** Mercury speciation in the roasting flue gas at the inlet of the ESD and outlet of the AP.

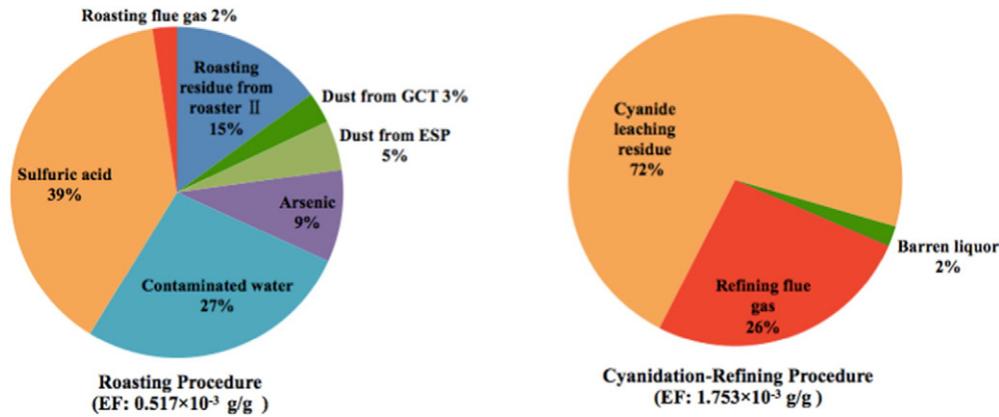


Fig. 4. Mass distribution of mercury in outputs for a two-stage roasting gold smelter in China.

### 3.5. Fate of Hg in roasting gold smelting

The Hg mass balance of tested smelter is shown in Table 1. The calculated mass balance recovery of the roasting procedure is 99%, indicating that the test results are in acceptable accuracy; while for the cyanidation-refining procedure, the calculated mass balance recovery is 72%. To further analyze the possible error sources, there are three sources where the Hg measurements are missed – dore, NaCN solution and the refining gas. Since there should be no Hg left in dore due to the extreme high refining temperature (over 1200 °C), and there is little Hg in chemically pure NaCN solution, the inaccuracy of mass balance recovery is primarily caused by the deficiency of the refining flue gas test.

The mass distributions of Hg in both roasting and cyanidation procedures are shown in Fig. 4. In the roasting procedure, except for the part of Hg which enters into the gold-bearing material for further cyaniding processing, most Hg is captured into the sulfuric acid, contaminated water, and arsenic with the proportion of 38.4%, 27.3% and 9.0%, respectively, and only 2.3% of Hg is emitted into the air with the roasting flue gas. The contaminated water from VT and ESD is mildly acidic and will be treated in sewage treatment plant before discharge or re-use. Sulfuric acid and arsenic are two by-products from the two-stage roasting gold smelters for sale. Both of them could be potential Hg emission sources to the environment if they are not properly disposed.

During the cyanidation-refining procedure, part of Hg is converted to highly water soluble di- or tetra-cyano Hg complex in the presence of cyanide. Based on the test data, 28% of the Hg reacts with  $CN^-$  and goes into pregnant gold/silver bearing solution, leaving 72% of Hg in the cyanide-leaching residue. Afterwards, the pregnant gold/silver bearing solution is processed with the zinc dust to have the precious metal precipitated in the high gold/silver concentration sludge, which is squeezed from the “barren liquor”. On the basis of our analysis, almost all the Hg (99%) in the pregnant solution is precipitated with the precious metal into the high gold/silver concentration sludge. And it will be totally released into the air with the refining flue gas if no APCDs are utilized. Both the cyanide leaching residue and the barren liquor are the tailing of the cyanidation-refining procedure, which are deposited in impoundment for storage and then delivered to the qualified tailing disposal facilities for treatment.

### 3.6. Hg emission factor for tested smelter

The Hg emission factor (EF) is defined as the amount of Hg emitted into the atmosphere per gram of gold produced during the smelting process. For the tested smelter, a mass balance method is applied to get a comprehensive understanding of Hg emission from the whole gold smelting process.

Since the Hg emission results from both the emission of the roasting flue gas in the roasting procedure and the refining flue gas in the cyanidation-refining procedure, the whole EF of Hg is the sum of the Hg EFs of the two procedures mentioned above, as shown in Eq. (R1). As for the roasting procedure, the Hg EF can be determined by subtracting the amount of Hg removed by APCDs from the whole amount of Hg in the flue gas which comes from the roasting of the gold concentrate to produce 1 g of gold, as shown in Eq. (R2). While in the cyanidation-refining procedure, Hg in  $H_2SO_4$  solution will not be used for calculation since the daily usage of  $H_2SO_4$  solution is no  $>0.01$  t, and the Hg emitted into the atmosphere is merely from the refining flue gas with no APCDs applied. Therefore, the Hg EF of the cyanidation-refining procedure can be calculated with Eq. (R3).

$$EF_{\text{overall}} = EF_{\text{Roasting}} + EF_{\text{Cyanidation-Refining}} \quad (R1)$$

$$EF_{\text{Roasting}} = \frac{M}{G \times \delta} r_R [\prod_k (1 - \eta_k)] \\ = \frac{M}{G \times \delta} r_R (1 - \eta_{\text{GCT}}) (1 - \eta_{\text{ESP}}) (1 - \eta_{\text{FF}}) (1 - \eta_{\text{VT}}) (1 - \eta_{\text{ESD}}) (1 - \eta_{\text{AP}}) \quad (R2)$$

$$EF_{\text{Cyanidation-Refining}} = \left( \frac{M \Delta_{\text{CR}}}{G \times \delta} \right) r_{\text{CR}} \quad (R3)$$

where, EF is the Hg emission factor; M and G are mercury/gold concentration in the feed gold concentrate;  $\delta$  is the recovery rate of gold;  $r$  is Hg release rate;  $k$  is the combination type of APCDs;  $\eta$  is the Hg removal efficiency of a certain combination of APCDs;  $\Delta$  is the Hg distribution coefficient; R stands for the roasting procedure; CR stands for the cyanidation-refining procedure; GCT stands for gas cooling tower; ESP stands for electrostatic precipitator; FF stands for fiber filter; VT stands for venture tube; ESD stands for electrostatic demister; AP stands for acid plant.

The Hg removal efficiencies listed in Table 2 and the Hg concentrations in the roasting flue gas listed in Table 4 have been used for calculation. The other parameters in Eqs. (R1)–(R3) are listed in Table 5. The value of G and  $\delta$  are provided by the tested smelter. Parameter  $r_R$  with the value of 85.1% has been discussed in Section 3.2. The calculation of Hg distribution coefficient for the cyanidation-refining procedure ( $\Delta_{\text{CR}}$ ) is to divide the whole Hg in the gold concentrates by the amount of the Hg in the raw material for the cyanidation-refining procedure,

Table 5  
Parameters in EF calculation.

Parameters	G ( $\mu\text{g}/\text{kg}$ )	$\delta$ (%)	$r_R$ (%)	$\Delta_{\text{CR}}$ (%)	$r_{\text{CR}}$ (%)
Value	35,000	70	85.1	22.8	25.8

and the determined result means that 22.8% of the whole Hg in the gold concentrate gets into the cyanidation-refining procedure. In the cyanidation-refining procedure, all the Hg will be emitted into the air through the refining glue gas except for part of Hg in the cyanide leaching residue and the barren liquor, and then  $r_{CR}$  is calculated to be 25.8%. Therefore, for the tested smelter in this research, EFs are  $0.517 \times 10^{-3}$  g/g for the roasting procedure, and  $1.753 \times 10^{-3}$  g/g for the cyanidation-refining procedure, so the overall EF is  $2.27 \times 10^{-3}$  g/g.

The EF obtained in this research was considerably smaller than others from previous studies (Pacyna et al., 2006, 2010; Streets et al., 2005, 2011; UNEP, 2013). The Hg EF is recovered through the method of field measurements and mass balance calculation, which is much more precise. But the Hg EF of gold smelting can vary dramatically depending on the Hg concentration of the gold concentrates, the specific smelting technology utilized and the effectiveness of the Hg control measures.

#### 4. Conclusions

In this study, on site measurements were conducted in a two-stage roasting gold smelter, and the Hg removal efficiency of APCDs was determined based on the mass balance calculation. The result indicated that the Hg removal efficiency of GCT, ESP, FF, VT, ESD and AP was approximately 4%, 6%, 12%, 7%, 31% and 96% respectively. The Hg release rate of two-stage roasting was 85.1%. In the roasting procedure, with application of all these air pollution control devices, only 2.3% of Hg in the gold concentrates was emitted to the atmosphere and the ratio of  $Hg_p:Hg^{2+}:Hg^0$  was 11:57:33. In the cyanidation-refining procedure, 5.9% of Hg in the gold concentrate was emitted to the atmosphere. Based on the Hg content in the gold concentrates, Hg release rate to the flue gas, distribution coefficient for the cyanidation-refining procedure and Hg removal efficiencies by APCDs, the atmospheric Hg emission factor was  $2.27 \times 10^{-3}$  g/g, with  $0.517 \times 10^{-3}$  and  $1.753 \times 10^{-3}$  g/g for roasting and cyanidation-refining procedure, respectively. The mercury emission in roasting and refining procedures take up to 23% and 77% of the total emission of the whole smelting process, respectively. These results can be used to develop more accurate Hg emission inventory for industrial gold production as well as help with the mercury emission control.

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