# Flow Analysis of the Mercury Associated with Nonferrous Ore Concentrates: Implications on Mercury Emissions and Recovery in China

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Estimation year	Region	Zn	Pb	Cu	Nonferrous metal production	Reference
1999	China	148	40	10	198	Streets et al., 2005 <sup>1</sup>
2003 <sup>d</sup>	China	187.6	70.7	17.6	275.9	Wu et al., 2006 <sup>2</sup>
2003	China	115.0	70.7	17.6	203.3	Pirrone et al., $2010^3$
2005	China	37.59	29.75	15.84	83.19	Hylander and Herbert, $2008^4$
2005	China	c	_c	c	65.8	AMAP/UNEP, 2008 <sup>5</sup>
2006 <sup>d</sup>	China	104.2	_c	_ <sup>c</sup>	c	Li et al., 2010 <sup>6</sup>
2006	China	107.7	_c	_ <sup>c</sup>	c	Yin et al., 2012 <sup>7</sup>
2010 <sup>d</sup>	China	39.4	30.6	2.5	72.5	Wu et al., 2012 <sup>8</sup>
2010	China	43.3	3.3	24.1	70.7	AMAP/UNEP, 2013 <sup>9</sup>
2010 <sup>d</sup>	China	_ <sup>c</sup>	_c	c	222.5 <sup>a</sup>	Tian et al., 2015 <sup>10</sup>
2010 <sup>d</sup>	China	62.9	31.0	3.5	97.4	Zhang et al., 2015 <sup>11</sup>
2007	Global	19	_c	_ <sup>c</sup>	54.5 <sup>b</sup>	Muntean et al., 2014 <sup>12</sup>
All-time to 2008	Global	10300	5990	4650	20940	Streets et al., 2011 <sup>13</sup>

S1. Atmospheric Hg emission from nonferrous metal production process

Table S1. Atmospheric Hg emissions from nonferrous metal production processes (t)

a. Including emissions from zinc (Zn) smelting, lead (Pb) smelting, copper (Cu) smelting, industrial gold smelting, and mercury mining.

b. Including emissions from Zn smelting, Pb smelting, Cu smelting, industrial gold production, artisanal gold production, and mercury mining.

c. "-"represents no detailed data.

d. Only results of the latest year were provided for the historical inventories.

## S2. Detailed description of the Hg flow model

#### S2.1 Hg input sub-model

The mercury (Hg) input  $(Q_{com,ij})$  is the product of Hg concentration in the consumed concentrates  $(C_{com,i})$  and the concentrates consumption  $(M_{com,ij})$ .

$$Q_{com,ij} = C_{com,i} M_{com,ij} \tag{E1}$$

where *i* and *j* refers to province and metal production processes. The provinces involved into concentrates production or consumption are listed in Table S2 and Table S3. Various metals production processes were used in China's nonferrous metal smelters.<sup>8</sup> The zinc (Zn) production processes included oxygen pressure leaching process (OPLP), electrolytic process (EP), imperial smelting process (ISP), retort zinc smelting process (RZSP), electric zinc furnace (EZF), and artisanal zinc smelting process (AZSP). There was no atmospheric mercury emission from OPLP since it was a hydrometallurgical process. Hg in ore concentrates consumed in the OPLP was released into water or solid wastes. Lead (Pb) smelting processes were divided into four major types, namely rich-oxygen pool smelting process (RPSP), imperial sinter process (ISP), sinter machine process (SMP), and sinter pan or pot process (SPP). Copper smelting processes included flash furnace smelting process (FFSP), rich-oxygen pool smelting process (RPSP), imperial furnace smelting process (IFSP), roasting-leaching-electrolyzing process (RLEP), as well as electric furnace smelting process (EF) and the revelatory furnace smelting process (RF).  $Q_{com}$  is Hg input, t;  $M_{com}$  is the concentrates consumed, t (Table S2);  $C_{com}$  is the Hg concentration in the consumed concentrated,  $\mu g g^{-1}$ .  $C_{com.,i}$  is calculated based on the Hg content in the concentrates supplied by k province or imported country.

$$C_{com,i} = \frac{\sum_{j} \sum_{k \ge i} C_{su,k \to ij} M_{su,k \to ij}}{\sum_{j} M_{com,ij}}$$
(E2)

where  $C_{su,k\rightarrow ij}$  is the Hg concentration in the produced/imported concentrates transported to *j* technology in *i* province for metal extraction. The key characteristics of the distribution curve for the Hg concentration in the produced concentrates are shown in Table S3.  $M_{su,k\rightarrow ij}$  is the concentrates transportation matrix, which is derived from the study by Wu et al., (2012) and Wu (2015).<sup>8, 14</sup>

Drovinco	Concentra	tes consump	tion (kt)	Drovinco	Concentrates consumption (kt)			
Flovince	Zn	Pb	Cu	Province	Zn	Pb	Cu	
Anhui	30.3	85.5	473.2	Jilin	0.0	0.0	0.8	
Fujian	65.7	28.0	113.4	Liaoning	276.5	19.1	37.6	
Gansu	293.2	22.4	684.3	Ningxia	0.0	33.8	0.0	
Guangdong	119.6	35.0	11.3	Qinghai	83.5	31.7	0.0	
Guangxi	385.4	147.7	0.0	Shaanxi	709.2	19.1	1.0	
Guizhou	28.7	0.0	0.0	Shandong	0.0	0.0	757.1	
Hebei	4.7	0.0	8.0	Shanxi	0.0	4.0	99.9	
Henan	281.6	1148.0	12.0	Sichuan	172.1	0.0	61.2	
Hubei	0.0	0.0	201.5	Xinjiang	18.8	39.8	10.2	
Hunan	1169.1	974.5	23.1	Yunnan	853.7	571.3	567.5	
Inner Mongolia	370.9	107.6	304.2	Zhejiang	26.7	0.0	115.5	
Jiangxi	49.8	95.7	582.4	National	4939.3	3363.2	4064.2	

Table S2. Provincial consumption of ore concentrates (metallic kt)

	Hg concentration in the Hg concentration in the			n in the	Hg concentration in the				
Province	Zn Con	centrates	$(\mu g \ g^{-1})$	Pb Con	centrates	(µg g <sup>-1</sup> )	Cu Con	centrates	$(\mu g g^{-1})$
	P10	P50	P90	P10	P50	P90	P10	P50	P90
Anhui				1.7	2.8	67.0	0.1	0.2	1.7
Chongqing				109.4	119.0	119.2			
Fujian	0.0	0.4	3.2	3.5	11.6	38.6			
Gansu	217.3	638.1	1873.3	3.8	9.7	24.7	0.3	2.9	24.8
Guangdong	42.7	209.0	580.7	12.9	34.7	93.5	0.0	0.1	0.1
Guangxi	1.2	11.1	102.4	1.5	10.4	71.6	0.4	1.5	2.1
Heilongjiang				21.4	24.1	32.9			
Henan	21.4	24.1	32.9	0.2	3.2	52.0			
Hubei				6.4	6.9	0.0	0.2	0.7	10.4
Hunan	0.3	1.9	17.8	0.5	2.5	9.0			
Inner	0.4	2 4	144	30.1	48 2	773	14	2.0	23
Mongolia	0.4	2.4	14.4	50.1	40.2	11.5	1.4	2.0	2.5
Jiangsu	12.8	29.9	34.6	7.0	22.7	73.3	0.1	0.1	0.1
Jiangxi	0.4	2.7	12.3	19.1	19.5	20.0	1.1	2.8	17.6
Jilin				41.4	55.0	77.4			
Liaoning				39.0	71.5	97.5			
Qinghai				0.0	0.4	4.6	1.7	1.8	1.8
Shaanxi	48.7	283.1	1644.6						
Shandong				4.3	5.0	5.6	1.0	1.6	2.4
Shanxi				51.8	52.2	52.5	0.1	0.1	0.3
Sichuan	11.5	27.6	124.9	5.0	26.9	145.7			
Tibet	0.1	0.2	0.9	0.0	0.0	0.0			
Xinjiang	2.4	12.5	64.1				0.1	1.3	45.9

Table S3. Key parameters of the distribution curve for Hg concentration in the produced/imported concentrates

Yunnan	0.4	3.3	166.4	18.3	20.9	24.8	1.8	16.9	157.8
Zhejiang	0.2	0.8	2.9	0.9	6.7	48.8			
China	31.8	77.5	147.3	13.6	27.1	46.1	2.0	3.7	6.5
Other countries	3.1	13.2	57.2	0.6	6.6	72.8	0.2	0.7	3.0

## **S2.2** Hg distribution sub-model for the metals production processes

Generally speaking, the metals production processes were generally divided into four stages, including dehydration, initial oxidation, extraction, and refining, as shown in Figure S1.



Figure S1. Metals production processes and wastes disposal processes

# S2.2.1 Hg distribution in the dehydration stage

In the dehydration stage, a small amount of Hg in the concentrates was released into the flue gas. Generally, the flue gas was de-dusted via dust collectors, and the dust and dehydrated concentrates were generally sent to the initial oxidation stage. Thus, only atmospheric Hg was emitted in this stage. The Hg distribution in this stage was calculated as follows. The Hg emitted to atmosphere is

$$GE_{d,ij} = Q_{d,ij}\gamma_{d,j}(1 - \eta_{d,j})$$
(E3)

Table S4. Other parameters for the Hg flow model<sup>6, 14-18</sup>

Metal	Process	Release rate (%)		<b>ó</b> )	Dis fac	Distribution factor (%)		Hg effic	Hg removal efficiency (%)		MC (%)	RR (%)	
		γd	$\gamma_{s}$	γe	$\gamma_r$	$\xi_{of}$	$\xi_{ss}$	$\xi_{se}$	$\eta_{d}$	$\eta_e$	$\eta_{r}$	α	φ
	EP	0.8	99.4	0.0	0.0	0.9	0.0	0.0	11.5	0.0	0.0	49.9	94.9
	EZF	0.5	99.4	59.1	57.2	0.6	0.0	0.6	11.5	11.5	11.5	49.9	96.9
Zn	RZSP	0.5	99.4	59.1	57.2	0.6	0.0	0.6	11.5	11.5	11.5	49.9	94.6
	ISP	0.1	99.1	65.7	57.2	1.0	0.0	0.9	40.1	40.1	40.1	49.9	96.9
	AZSP	0.0	99.4	59.1	57.2	0.6	0.0	0.6	0.0	0.0	0.0	49.9	94.6
	RPSP	0.0	98.7	60.1	57.2	0.6	0.0	39.4	40.1	0.0	0.0	59.3	95.9
DI	SMP	0.1	98.6	65.7	57.2	0.6	0.0	20.3	11.5	0.0	0.0	59.3	95.9
Pb	ISP	0.1	99.1	65.7	57.2	1.0	0.0	0.0	40.1	0.0	0.0	59.3	95.9
	SPP	0.0	98.6	65.7	57.2	0.6	0.0	20.3	11.5	0.0	0.0	59.3	95.9
	FFSP	1.3	97.7	89.1	60.0	0.6	0.0	0.8	40.1	40.1	40.1	21.4	97.7
	RPSP	0.0	98.1	60.1	54.3	0.1	1.8	0.3	40.1	40.1	40.1	21.4	97.7
Cu	RLEP	0.0	97.9	68.8	57.2	0.6	1.8	0.3	40.1	40.1	40.1	21.4	97.7
	IFSP	0.5	97.9	68.8	57.2	0.6	1.8	0.3	40.1	40.1	40.1	21.4	97.7
	EF/RF	0.0	97.9	68.8	57.2	0.6	1.8	0.3	0.0	0.0	0.0	21.4	97.7

MC: Metal content; RR: Recovery rate; EP: Electrolytic process; ISP: Imperial smelting process; RZSP: Retort zinc smelting process; EZF: Electric zinc furnace; AZSP: Artisanal zinc smelting process; RPSP: Rich-Oxygen pool smelting process; SMP: Sinter machine process; SPP: Sinter pan or pot process; FFSP: Flash furnace smelting process; RPSP: Rich-oxygen pool smelting process; IFSP: Imperial furnace smelting process; RLEP: Roasting-leaching-electrolyzing process; EF: Electric furnace smelting process; RF: Revelatory furnace smelting process. where  $Q_d$  is Hg input into the dehydration stage, which equals to the Hg amount in the ore concentrates consumed, t;  $GE_d$  is the Hg emitted to the atmosphere in the dehydration stage, t; *i*, *j* and *d* refer to the province, metals production processes and dehydration stage, respectively;  $\gamma_d$  is the Hg release rate in the dehydration stage, % (see **Table S4.**);  $\eta_d$  is the Hg removal efficiency of the dust collectors in the dehydration stage, % (see **Table S4.**).

# S2.2.2 Hg distribution in the initial oxidation stage

The Hg input into the initial oxidation stage  $(Q_s)$  is the Hg amount in the dehydrated concentrates and dust.

$$Q_{s,ij} = Q_{d,ij} (1 - \gamma_{d,j} + \gamma_{d,j} \eta_{d,j})$$
(E4)

Most Hg in the dehydrated concentrates and the dust was released to the flue gas in the smelting or roasting furnace, leaving a small amount of Hg in the furnace slag or intermediate products.<sup>16, 18</sup> The furnace slag and intermediate products from the Zn and Pb production processes were further used in their corresponding production processes.<sup>19, 20</sup> Only the smelting furnace slags from the Cu production process was released out of the metals production processes, and certain amount was reused in the wastes disposal processes.<sup>21-23</sup> The Hg amount in the Cu smelting furnace slags ( $I_{ss,ij}$ ) is calculated as follows.

$$I_{ss,ij} = Q_{s,ij} \xi_{ss,j} \tag{E5}$$

where  $\xi_{ss}$  is the percentage of Hg in the slags in total Hg input of initial oxidation stage, % (see **Table S4.**).

The initial oxidation stage was generally considered as the largest release point for air pollutants. Thus, flue gas typically must undergo a thorough de-dusting and required additional Hg removal being desulfurization in certain cases.<sup>8, 24</sup> The air pollution control devices were combined into 8 types in 2012.<sup>14</sup> The Hg in the emitted gas ( $GE_s$ ) from the initial oxidation stage is:

$$GE_{s,ij} = Q_{s,ij}\gamma_{s,j}\sum_{m}\theta_{m,ij}(1-\eta_m)$$
(E6)

The Hg in the flue gas was removed into wastes and byproducts, including dust, waste acid, calomel, sulfuric acid and FGD slag. The dust was reused in the initial oxidation stage or in the extraction stage or was sent to another metal production system to recover valuable metals.<sup>19, 25, 26</sup> Only a subset of smelters used the dust to recover valuable metals in 2012.<sup>27</sup> Thus, we assumed that dust was reused in the metals production processes. Waste acid was generally sent out of the metals production systems and disposed at the waste water stations. Thus, the Hg amount in the waste acid output of the metal production system ( $I_{wa}$ ) is as follows.

$$I_{wa,ij} = Q_{s,ij} \gamma_{s,j} \sum_{m} \theta_{m,ij} (1 - \eta_{dc}) \eta_{fgs+esd}$$
(E7)

where  $\gamma_s$  is the percentage of Hg released from the concentrates and dust into the flue gas, % (**Table S4.**);  $\theta$  is the application percentage of a certain type of air pollution control device, % (**Table S4**); *m* refers to the type of air pollution control device;  $\eta_{dc}$  and  $\eta_{fgs+esd}$  are the Hg removal efficiency of the dust collectors, and the flue gas scrubber+electrostatic demister, % (**Table 1**).

The calomel released out of the metal production process was sent to the qualified Hg recovery company. The Hg in the calomel  $(I_{smr})$  is as follows.

$$RM_{smr,ij} = Q_{s,ij}\gamma_{s,j}\sum_{m}\theta_{m,ij}(1-\eta_{dc})(1-\eta_{fgs+esd})\eta_{smr}$$
(E8)

where  $\eta_{smr}$  is the Hg removal efficiency of the specific Hg removal tower, % (Table 1).

The sulfuric acid was released out the metals production systems as byproduct. Hg

in the sulfuric acid  $(I_{sa})$  is as follows.

$$I_{sa,ij} = Q_{s,ij} \gamma_{s,j} \sum_{m} \theta_{m,ij} (1 - \eta_{dc}) (1 - \eta_{fgs+esd}) (1 - \eta_{smr}) \eta_{sa}$$
(E9)

where  $\eta_{sa}$  is the Hg removal efficiency of the sulfuric acid plants, % (**Table 1**).

The FGD slags from the initial oxidation stage were mainly produced in Cu smelters in 2012. The FGD slags were divided into sodium sulfate, activated coke, ammonia sulfate and FGD gypsum according to the desulfurization technology applied. The Hg in the first three types was primarily input into chemical plants, and the Hg in FGD gypsum was mostly dumped. We assumed that 50% of the Hg in desulfurization wastes ( $I_{fgd}$ ) from the initial oxidation stage was passed to the chemical plants and the remainder was dumped as discarded slags.

$$I_{fgd,ij} = Q_{s,ij} \gamma_{s,j} \sum_{m} \theta_{m,ij} (1 - \eta_{dc}) (1 - \eta_{fgs+esd}) (1 - \eta_{smr}) (1 - \eta_{sa}) \eta_{fgd}$$
(E10)

where  $\eta_{fgd}$  is the Hg removal efficiency of the FGD tower, % (**Table 1**).

APCD combinations	Туре	Zn	Pb	Cu
None	Type 1	4.5	5.7	0.3
DC	Type 2	3.0	6.2	0.4
FGS	Type 3	0.0	0.0	0.0
DC+FGS	Type 4	0.7	12.6	0.3
DC+FGS+ESD+SCSA	Type 5	1.3	16.1	1.6
DC+FGS+ESD+DCDA	Type 6	79.6	59.4	48.3
DC+FGS+ESD+SMR+DCDA	Type 7	10.9	0.0	0.0
DC+FGS+ESD+DCDA+DFGD	Type 8	0.0	0.0	33.2

Table S5. Application percentage of different types of APCD combinations (%)

APCDs: Air pollution control devices; None: no APCDs is applied; DC: Dust collectors; FGS: Flue gas scrubber; ESD: Electrostatic demister; SMR: Specific Hg removal tower; DCDA: Double contact and double absorption tower; SCSA: Single contact and single absorption tower; WFGD: Wet flue gas desulfurization; DFGD: Dry flue gas desulfurization.

# S2.2.3 Hg distribution in the extraction and refining stage

The Hg distribution in the pyrometallurgical extraction and refining stage was quite similar as that in the dehydration stage.

In the extraction stage, exhaust overflow gas was present in addition to the extraction gas. Dust collectors were used for gas cleaning. Dust was primarily returned to the extraction process except for a small portion with high Pb concentration.<sup>22</sup> In this case, we assumed that all dust was returned to the extraction furnace.

The Hg input into the extraction stage  $(Q_e)$  is

$$Q_{e,ij} = Q_{d,ij} (1 - \gamma_{d,j} + \gamma_{d,j} \eta_{d,j}) (1 - \gamma_{s,j} - \xi_{ss,j})$$
(E11)

The Hg output of the metals production processes with the extraction furnace slags  $(I_{es})$  is

$$I_{es,ij} = Q_{e,ij} \xi_{es,j} \tag{E12}$$

where  $\xi_{es}$  is the percentage of Hg in the furnace slag in total Hg input into extraction stage, % (Table S4.).

The Hg emitted to the atmosphere from the extraction stage is

$$GE_{e,ij} = Q_{e,ij} \gamma_{e,j} (1 - \eta_{e,j} + \xi_{of} \eta_{e,j})$$
(E13)

where  $\gamma_e$  is the percentage of Hg released into the flue gas from the material for the extraction stage, % (**Table S4.**);  $\eta_e$  is the Hg removal efficiency of the dust collectors for the extraction stage, % (**Table S4.**);  $\xi_{of}$  is the percentage of Hg into the overflow gas in the total Hg of the flue gas, % (**Table S4.**).

In the refining stage, the Hg input  $(Q_r)$  is

$$Q_{r,ij} = Q_{d,ij} (1 - \gamma_{d,j} + \gamma_{d,j} \eta_{d,j}) (1 - \gamma_{s,j} - \xi_{ss,j}) (1 - \gamma_{e,j} - \xi_{es,j})$$
(E14)

The Hg concentration of the materials input to the refining stage (such as blister Cu and Pb bullion) was generally less than 30 ng g<sup>-1.5</sup> Thus, the mobile Hg in this stage was notably lower than that in other stages. Solid wastes  $(OD_r)$  including furnace slag and dust produced in this stage were assumed to be discarded.

$$OD_{rs,ij} = Q_{r,ij}(\xi_{rs,j} + \gamma_{r,j}\eta_{r,j})$$
(E15)

where  $\gamma_r$  is the percentage of Hg released into the flue gas from the material for the refining stage, % (**Table S4.**);  $\xi_{rs}$  is the percentage of Hg in the furnace slag in total Hg input into refining stage, % (**Table S4.**).  $\eta_r$  is the Hg removal efficiency of the dust collectors for the refining stage, % (**Table S4.**). The limited Hg in the refined metal is negligible. The atmospheric Hg emissions from the refining stage (*GE<sub>r</sub>*) are

$$GE_{r,ij} = Q_{r,ij} \gamma_{r,j} (1 - \eta_{r,j})$$
(E16)

For the electrolytic Zn process, the wet methods were used in both the extraction and refining stages. Thus, the Hg was distributed into several slags and waste acid.

$$I_{ex,i} = \theta_x Q_{e,iep} \tag{E17}$$

$$\sum_{x} \theta_{x} = 1 \tag{E18}$$

where  $Q_{e,ep}$  is the Hg input into the extraction stage of the electrolytic Zn process, t;  $I_{ex}$  is the Hg amount in x slag, t;  $\theta_x$  is the Hg distribution factor during the Zn extraction and refining stages (**Table S6**), %; x refers to the leaching slags, Ag slag, Cu slag, Cd slag, Co slag, In slag, Pb slag and waste acid.

Zn smelting slag and waste acid	Leaching slag	Ag concentrates	In slag	Co slag
$\theta_x$ (%)	28.4	4.2	10.5	10.0
Zn smelting slag and waste acids	Cu slag	Cd slag	Pb slag	Waste acid
$\theta_x$ (%)	3.5	7.1	6.1	30.2

Table S6. Hg distribution in the slags and waste acid from Zn extraction/refining stages<sup>14, 18</sup>

#### S2.3 Hg distribution sub-model for the wastes disposal processes

The wastes disposal processes considered in this study primarily included sulfuric acid utilization processes, waste acid disposal processes, and smelting slags disposal processes. The recoveries of Hg from acid slags and calomel were calculated in the sector of Hg distribution as a component of the waste acid disposal processes. The Hg distribution in the flue gas desulfurization (FGD) slags disposal processes was based on the assumption that Hg was either dumped or placed in interim storage at the chemical plants. The detailed disposal methods and assumptions applied for the main intermediated wastes and byproducts are shown in **Table S7**.

	Wastes/	Disposal method							
Stage	Byproduc t	Zn smelters	Pb smelters	Cu smelters					
Dehydr ation	Dust	Used as raw materials in the oxide	Used as raw materials in the oxidation stage						
	Dust	<ol> <li>Used as raw materials in the extraction stage; 2) Returned to the oxidation process;</li> <li>Note: the dust produced in the retort zinc smelting process and electric zinc process was not suitable to re-roasted in the oxidation process, but it was the case in China by lacking better disposal methods.</li> <li>Mainly returned to the oprocess; 2) Several smelters recy valuable metals in the dust through the dust the dust through the dust the dust the dust the dust the dust the dust through the dust the dust t</li></ol>							
	Waste acid	Disposed in the waste acid station	ι.						
Initial oxidati	Sulfuric acid	Used in the fertilizer plants, concentrating mill.	chemical plan	ts, dealers, smelters and					
on	Calomel	Sold to Hg recovery company No calomel produced.							
	FGD slags	Including FGD gypsum, ammonia sulfate, sodium sulfate, zinc sulfate. 1) the FGD gypsum was mainly sent to cement plant; 2) the ammonia sulfate and sodium sulfate were sent to chemical plants; 3) the zinc sulfate was used in the extraction process. The Zn smelters rarely installed the desulfurization towers in 2012. Thus, we assumed no desulfurization slags were	Mainly as FGD gypsum used in the cement plants. The Pb smelters rarely used the desulfuriza tion towers	Including desulfurization gypsum, ammonia sulfate, sodium sulfate, activated coke. 1) The desulfurization gypsum was mainly dumped in the smelters; 2) the ammonia sulfate and sodium sulfate were sent to chemical plants; 3) the used activated coke was recycled in high					

Table S7. Disposal methods and assumptions made for the wastes/byproduct produced from the metals production processes

		produced in 2012.	in 2012. Thus, we assumed no desulfuriza tion slags were produced in 2012.	temperature to recover sulfur dioxide. Hg in the activated coke was released into the flue gas. We assumed that the Hg in the desulfurization slags was either dumped or temporarily stored in the chemical plants.
	Furnace slags	Only a small amount produced in several smelters. We assumed no furnace slag was produced in the Zn smelters.	Used in the extraction stage.	Several large-scale smelters recover the Cu in the slags, most smelters discarded the slags.
	Dust	Assumed no dust produced since more than 80% of Zn was extracted with hydrometallurgical processes.	Returned to the extraction stage.	Returned to the extraction stage. Dust in some smelters with high Pb concentration was sold to recover Pb. We assumed that all dusts were returned for extraction.
Extract	Furnace slags/Lea ching slag/Purif ication slags	Most leaching slags were roasted in the volatilization kiln at the temperature larger than 800 °C. The purification slags generally included Ag slag, In slag, Co slag, Cu slag, Cd slag, and Pb slag. The Ag, Co and In slag were used to recover metals with hydrometallurgical process. The smelters generally recovered Cd and Pb slags with pyrometallurgical process. Both the pyrometallurgical process and hydrometallurgical process were used to recover Cu.	Used to recover ZnO in the fuming furnace at the temperatur e larger than $800 \ C$ .	Mainly discarded in the smelters.
Refinin g	Furnace slags	Assumed discarded.	Assumed discarded.	Assumed discarded.

# S2.3.1 Hg distribution in the sulfuric acid utilization process

Hg distribution in the sulfuric acid utilization process is.

$$Q_{sa,k} = \sum_{i} \sum_{j} I_{sa,ij} \theta_{k,i}$$
(E19)

$$OP_{t,k} = \sum_{k} Q_{sa,k} \delta_{t,k}$$
(E20)

where  $Q_{sa,k}$  is the Hg input with sulfuric acid into k department, t;  $\theta_x$  is the percentage of Hg deliver to k department in the total Hg input with the sulfuric acid, % (**Table S8**); The k department refers to the fertilizer plant, chemical plant, concentrating mill, smelters and dealers;  $\delta_{t,k}$  is the percentage of Hg in the k department that passed to t fate, % (**Table S9**). The parameter t refers to the fate of Hg during utilization of sulfuric acid. The fates include interim storage, dumped, and emissions to air, water or soil.

Sulfuric acid users	Sulfuric acid produced in Zn smelters	Sulfuric acid produced in Pb smelters	Sulfuric acid produced in Cu smelters
Dealers	20.1	19.3	20.3
Zn smelters	16.1	1.7	2.9
Other smelters	24.2	7.8	0.5
Fertilizer	26.1	21.0	52.7
Chemical plants	6.2	34.8	23.4
Concentrating mills	7.3	15.6	0.2

Table S8. Hg distribution in different sulfuric acid users  $(\%)^{14, 28}$ 

Table S9	Hg distribution	factors in	different fate	s in the	sulfuric	acid users	14, 18, 28, 29
	ing distribution	Idetors III	uniforent fate	s m the	Sumune	acta asers	

Sulfuric acid users	Air	Water	Soil	Dumped	Interim storage
Dealers	0.0	0.0	0.0	0.0	100.0
Zn smelters	23.6	0.8	2.0	71.3	2.2

Other smelters	0.0	2.0	2.0	96.0	0.0
Fertilizer	1.4	0.1	40.6	57.9	0.0
Chemical plants	2.5	0.1	5.0	58.4	34.0
Concentrating mill	0.0	24.5	2.0	73.5	0.0

#### S2.3.2 Hg distribution in the waste acid disposal process



Figure S2. Waste acid disposal processes

The main methods used for waste acid disposal included the lime neutralization method, lime-ferric salt neutralization method, sulfide-neutralization method and biological method.<sup>23, 30</sup> The first two methods generally consisted of three stages, including settling (set), neutralization (neu) and arsenic removal (asr), which refer to as type 1 in this work. The latter two methods have one additional stage, sulfurization/coordination (sul), and the latter two methods are classified as type 2 (**Figure S2**). The Hg emitted into water ( $LE_{wa}$ ) is

$$LE_{wa} = \sum_{i} \sum_{j} (I_{wa,ij} + 30.2\% Q_{e,iep})(1 - \eta_{set}) [\theta_1 + \theta_2(1 - \eta_{sul})](1 - \eta_{neu})(1 - \eta_{asr})$$
(E21)

where  $\eta_{set}$ ,  $\eta_{sul}$ ,  $\eta_{neu}$  and  $\eta_{asr}$  is the Hg removal efficiency of the setting, sulfurization/coordination, neutralization and arsenic removal stage, respectively, %.  $\theta_1$ and  $\theta_2$  is the application percentage of type 1 and type 2, % (**Table S10**); the 30.2% here is the percentage of Hg into the waste acid in the leaching and refining stages of the Zn electrolytic process, as shown in **Table S6**.

In the waste acid disposal process, the Hg was distributed into settling slag, sulfurization/coordination slag, neutralization slag and arsenic slag. The settling slag

was sent to qualified Hg recovery companies, returned to the smelting furnace, or temporarily stored in the smelters. The percentages of Hg in the settling slag sent to each disposal method were  $\theta_{set1}$ ,  $\theta_{set2}$ , and  $\theta_{set3}$  (**Table** S11), respectively. The sulfurization/coordination slag was either temporarily stored in the smelters or sent to a qualified Hg recovery company. The percentages of Hg routed to these two disposal methods were  $\theta_{sul1}$  and  $\theta_{sul2}$  (**Table** S11). Several large smelters recovered arsenic from the arsenic slag, but most smelters contained no technology or economic drivers necessary to recover the arsenic.<sup>31</sup> Thus, we assumed that arsenic slag was temporarily stored in the smelters. The neutralization slag was primarily discarded as dumped slag.

Thus, the Hg amount into the qualified Hg recovery company  $(RM_{wa})$  is

$$RM_{wa} = \sum_{i} \sum_{j} (I_{wa,ij} + 30.2\% Q_{e,iep}) \eta_{set} \theta_{set1} + I_{wa,ij} \theta_2 (1 - \eta_{set}) \eta_{sul} \theta_{sul1}$$
(E22)

The Hg amount in the temporarily stored slags  $(IS_{wa})$  is

$$IS_{wa} = \sum_{i} \sum_{j} (I_{wa,ij} + 30.2\% Q_{e,iep}) [\eta_{set} \theta_{set2} + \eta_{set} \theta_{set3} \eta_{s,\min} + (1 - \eta_{set}) \theta_2 \eta_{sul} \theta_{sul2})]$$
(E23)

The Hg amount in the discarded slags  $(OD_{wa})$  is

$$OD_{wa} = \sum_{i} \sum_{j} (I_{wa,ij} + 30.2\% Q_{e,iep})(1 - \eta_{set}) [\theta_1 + \theta_2(1 - \eta_{sul})] [\eta_{neu} + (1 - \eta_{neu})\eta_{oxi}]$$
(E24)

The Hg in the settling slags returned to the metals production process  $(IR_{wa})$  is

$$IR_{wa} = \sum_{i} \sum_{j} (I_{wa,ij} + 30.2\% Q_{e,iep}) \eta_{set} \theta_{set3}$$
(E25)

The reutilization of settling slag led to the atmospheric Hg emission ( $GE_{wa}$ ) is

$$GE_{wa} = (I_{wa,ij} + 30.2\% Q_{e,iep})\gamma_s (1 - \eta_{s,\min})$$
(E26)

where  $\eta_{s,\min}$  is the Hg removal efficiency of the APCDs for the initial oxidation

stage, %. We used the lowest Hg removal efficiency of the APCD combinations of DC+FGS+ESD+DCDA shown in Table 2 for two reasons. First, acid slags were smelters installed APCD generally reused in with combinations of DC+FGS+ESD+DCDA. Second, acid slags were generally input into the roaster with a large amount of Hg in a short time, which reduced the synergic Hg removal efficiency of APCDs. For example, the larger the Hg concentration in the flue gas, the lower Hg removal efficiency of electrostatic precipitator will be, as indicated from field and lab experiments.<sup>6, 32</sup> The removed Hg was assumed to be temporarily stored in the smelters. The Hg recovered from the calomel and waste acid slag is

$$RE_{wa} = (RM_{wa} + RM_{smr,ij}) \times \lambda_{RE}$$
(E27)

where  $\lambda_{RE}$  is the recovery rate of the Hg from the calomel and waste acid slag, %. In this work, we used the 95% as the recovery rate.<sup>33</sup> We assumed that about 1% of the Hg emitted to atmosphere and the rest was discarded with the dumped slags.

Table S10. Application percentage of different methods for waste acid disposal (%)

Waste acid disposal method	Zn smelter	Lead smelter	Cu smelter
SET+NEU+ASR, $\theta_1$	63.2	68.2	24.8
SET+SUL+NEU+ASR, $\theta_2$	36.8	31.8	75.2

SET: Settling; NEU: Neutralization; ASR: Arsenic; SUL: Sulfurization/Coordination

Table S11.	Percentage	of Hg	distributed	among	different	disposal	wavs	(%)
		- 0						( )

The type of acid slags	A aid along disposal ways	Zn	Pb	Cu
	Actu stags disposat ways	smelter	smelter	smelter
Settling slags	Reclaimed in the Hg recovery company, $\theta_{set1}$	13.8	0.4	0.0
	Interim storage, $\theta_{set2}$	51.9	37.7	70.8
	Roasted in the smelter, $\theta_{set3}$	34.2	61.9	29.2

Sulfurization/	Reclaimed in the Hg recovery company, $\theta_{\text{sull}}$	29.6	0.0	0.0
coordination slags	Interim storage, $\theta_{sul2}$	70.4	100.0	100.0





Figure S3. Leaching slags disposal processes

The detailed disposal procedure for leaching slags was described in our previous studies<sup>18, 34</sup>. The method used to calculate the Hg distribution in the Zn leaching slags disposal processes was similar to that for the metals production processes. The Hg in the leaching slags was calculated in (E17). Both dust collectors and flue gas desulfurization towers were assumed to be used for air pollution control in the large smelters, whereas only dust collectors were used in other smelters. The dust (mainly zinc oxide product) and FGD slags were temporarily stored in the smelters. The furnace slag consisted of ferrous slag, which was used in building materials. The Hg in this material was assumed to be stabilized.

$$GE_{ls} = \sum_{i} \sum_{j} I_{ls,ij} \gamma_{ls} (1 - \eta_{dc}) (1 - \theta_{ds} \eta_{ds})$$
(E28)

$$IS_{ls} = \sum_{i} \sum_{j} I_{ls,ij} \gamma_{ls} (\eta_{dc} + \theta_{ds} \eta_{ds} - \eta_{dc} \theta_{ds} \eta_{ds})$$
(E29)

$$ST_{ls} = \sum_{i} \sum_{j} I_{ls,ij} (1 - \gamma_{ls})$$
(E30)

where  $GE_{ls}$ ,  $IS_{ls}$  and  $ST_{ls}$  respectively refers to the amount of Hg emitted to air, temporarily stored and stabilized during leaching slags disposal process.  $\gamma_{ls}$  is the Hg released rate in the furnace and is approximately 93.2%.  $\eta_{dc}$  and  $\eta_{ds}$  are the Hg removal efficiencies of dust collectors and FGD towers and are  $4.3\pm3.1\%$  and  $38.5\pm27.6\%$ . The values for the Hg release rate and Hg removal efficiency were assigned based on field experiments conducted at three Zn smelters.<sup>18</sup> We assumed that the Hg removal efficiencies fit the normal distributions. The standard deviation of the Hg removal efficiency was used to generate the uncertainty, and  $\theta_{ds}$  was the application percentage of FGD towers in the Zn smelters with a value 36.8%.

The Hg distribution in the Zn purification slags is calculated as follows.

$$OP_{t,x} = \sum_{t} \sum_{i} I_{ex,i} \delta_t$$
(E31)

where x refers to the Zn smelting slags, including leaching slag, Ag slag, Cu slag, Cd slag, Co slag, In slag and Pb slag (The fate of Hg in the waste acid and leaching slags were calculated in S4.2 and S4.3); t refers to the Hg fates, including interim storage, discarded with slags, or emitted to air or water.  $\delta_t$  is the percentage of Hg into each fate, % (Table S12).

Table S12. The percentage of Hg into different fates in the utilization process of Zn purification slags (%).<sup>14</sup>

Slags	Pyrometallurgical process			Hydrometallurgical process			
	Air	Dumped	Interim storage	Dumped	Water	Interim storage	
Ag slag	_	_	_	5.0	90.0	5.0	
Cu slag	55.0	31.3	13.7	5.0	90.0	5.0	
Cd slag	48.5	37.8	13.7	_	_	_	
Co slag	_	_	_	5.0	90.0	5.0	
Pb slag	77.1	9.2	13.7	_	_	_	
In slag	_	_	_	5.0	90.0	5.0	

The main smelting slags from the Pb production processes were the extraction furnace slags. The extraction furnace slags were roasted in the fuming furnace. The flue gas was dedusted with dust collectors. The dust was the ZnO byproduct and the fuming furnace slag was discarded.

The atmospheric Hg emitted  $(GE_{es})$  is

$$GE_{es} = \sum_{i} \sum_{j} I_{es,ij} \gamma_{es} (1 - \eta_{dc})$$
(E32)

The Hg temporarily stored in the ZnO byproduct (IS) is

$$IS_{es} = \sum_{i} \sum_{j} I_{es,ij} \gamma_{es} \eta_{dc}$$
(E33)

The Hg discarded with the fuming furnace slag  $(OD_{es})$  is

$$OD_{es} = \sum_{i} \sum_{j} I_{es,ij} (1 - \gamma_{es})$$
(E34)

where  $\gamma_{es}$  is the percentage of Hg released from the extraction furnace slag into the flue gas, % ( $\gamma_{es}$  =93.2%, the value for the leaching slag disposal process is used here);  $\eta_{dc}$  is the Hg removal efficiency of the dust collectors, % (**Table 1**).

The Cu slags included smelting furnace slag, extraction furnace slag and refining furnace slag. The utilization rate of Cu slags was less than 12% in China.<sup>22</sup> Most slags were discarded in the slags disposal pits. Here we assumed that the smelting furnace slag in the large smelters was roasted to recover Cu, which led to atmospheric Hg emission. Other slags were assumed to be discarded.

The atmospheric Hg emission is

$$GE_{cs} = \sum_{i} \sum_{j} I_{ss,ij} \theta_{lar} \gamma_{cs}$$
(E35)

The Hg amount in the discarded slags is

$$OD_{cs} = \sum_{i} \sum_{j} I_{ss,ij} (1 - \theta_{lar} \gamma_{cs}) + I_{es,ij} + I_{rs,ij}$$
(E36)

where  $\theta_{lar}$  is the utilization percentage of smelting furnace slag, % ( $\theta_{lar}$ =79.1%);  $\gamma_{cs}$  is the percentage of Hg released from the smelting furnace slag into atmosphere, % ( $\gamma_{cs}$ =93.2%, the value for the leaching slag disposal process was used here).



# S3. The flow of Hg associated with Zn, Pb and Cu concentrates

Figure S4. The flow of Hg associated with Zn concentrates



Figure S5. The flow of Hg associated with Pb concentrates



Figure S6. The flow of Hg associated with Cu concentrates

# S4. The key parameters of the emission and recovery curve

	The type of ore concentrates used	P10	P50	P90
Hg emissions to air from metals	Nonferrous ore concentrates	33.4	100.4	181.7
	Zn concentrates	17.2	55.5	99.9
production	Pb concentrates	14.5	41.5	76.3
processes	Cu concentrates	1.7	3.4	5.5
	Nonferrous ore concentrates	3.4	47.8	102.7
Hg emissions to	Zn concentrates	2.9	41.9	90.0
disposal processes	Pb concentrates	0.2	3.8	8.5
	Cu concentrates	0.2	2.1	4.2
	Nonferrous ore concentrates	36.7	148.2	284.4
Hg emissions to air	Zn concentrates	20.1	97.4	189.9
from the whole flow	Pb concentrates	14.7	45.3	84.8
	Cu concentrates	1.9	5.5	9.6
Hg emissions to water from the whole flow	Nonferrous ore concentrates	1.1	32.6	68.7
	Zn concentrates	0.9	28.4	60.1
	Pb concentrates	0.0	3.2	6.6
	Cu concentrates	0.3	1.0	2.0
	Nonferrous ore concentrates	-4.3	21.3	53.1
Hg emissions to soil	Zn concentrates	-3.2	11.4	28.7
from the whole flow	Pb concentrates	-0.2	5.1	11.9
	Cu concentrates	-0.9	4.8	12.5
	Nonferrous ore concentrates	12.6	62.8	122.5
Ha recovery	Zn concentrates	12.5	62.7	122.3
ing recovery	Pb concentrates	0.0	0.1	0.2
	Cu concentrates	0.0	0.0	0.0

Table S13. The key parameters (P10, P50, P90) of the Hg emission and recovery curve

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