Supporting information for "Update of mercury emissions from China's primary zinc, lead and copper smelters, 2000-2010"

Submitted to ACP for Special Issue titled "Atmospheric Mercury Processes: Papers from the 10th ICMGP"

Q. R. Wu[1], S. X. Wang[1], L. Zhang[1], J. X. Song[1], H. Yang[1] and Y. Meng[1]

[1]School of Environment, and State Key Joint Laboratory of Environment Simulation and Pollution Control, Tsinghua University, Beijing 100084, China

13 pages (including cover page)

6 Tables (S1, S2, S3, S4, S5,S6)

1 Figure (S1)

1. Number of sampling mines by province or from other countries

Chinese concentrates samples were selected mainly based on provincial production of concentrates. The concentrates production from provinces with samples accounted for 94.37%, 97.50% and 93.17% of the national zinc, lead and copper production, respectively. The imported concentrates were collected from smelters with large consumption of imported concentrates. The imported zinc concentrate samples were mainly from the United States, Peru, Mexico, Australia, India and Sweden. Imported lead concentrate samples were mainly from Australia and Kazakhstan, while copper concentrates samples were from Chile, Australia, Mexico, Mongolia, Kazakhstan, Tanzania, Botswana and Canada. The Chinese ore content database in this study covered 351 zinc concentrate samples from 118 zinc mines, 190 lead concentrate samples from 83 lead mines and 174 copper concentrate samples from 55 copper mines. In addition, 39 zinc concentrate samples, 8 lead concentrate samples and 33 copper concentrate samples were collected from imported concentrates. The zinc, lead and copper supply in 2010 and the number of sampling mines by province or from other countries are shown in **Table S1**.

Table S1. Number of sampling mines by province or from other countries

Province	Zinc		Lead		Copper	
	Supply (kt)	Number of sampling mines	Supply (kt)	Number of sampling mines	Supply (kt)	Number of sampling mines
Anhui	13	1	13	2	128	4
Chongqing	24			1[a]		
Fujian	147	11	75	4	10	
Gansu	203	9	48	3	74	4
Guangdong	194	3	127	3	9	1
Guangxi	337	9	238	12	7	3
Guizhou	26		23			
Hebei	38		14		2	
Heilongjiang				1[a]	2	
Henan	60	4	70	7	7	

Hubei				1[a]	55	6
Hunan	583	26	283	11	6	
Inner Mongolia	750	6	443	4	170	2
Jiangsu	21	2	15	3	1	1
Jiangxi	55	10	47	1	208	7
Jilin	22		42	2	16	
Liaoning	47		19	6	12	
Qinghai	84		68	3	40	1
Shaanxi	211	12	52	3	9	
Shandong				1[a]	10	1
Shanxi			13	1	27	3
Sichuan	367	10	208	5	72	3
Xinjiang	26	3	12		74	7
Xizang	26	1	28	1	5	
Yunnan	560	6	107	3	201	12
Zhejiang	46	5	37	5	9	
National	3842	118	1981	83	1156	55
Other countries	1458	10[b]	881	8[b]	1733	9[b]

a. Mines in these provinces were out of production in 2010.

b. Samples from the same country were regarded as from one mine in this table.

2. Sampling, preparation and analysis methods for ore concentrates

We sampled the concentrates following the method of Stockpile Random Sampling (SRS) and Loader Random Sampling (LRS), which was detailed described in our previous paper (Zhang et al., 2012). Usually, at least three valid samples were collected in each mine for analysis. However, for samples with extreme values or mines with large production, additional samples were collected and analyzed. The collected samples were air dried to constant weight, and then pulverized into 80 meshes (200 μm in particular diameter). The dried samples were then analyzed following EPA method 3052 with F732-V Intelligent Mercury Analyzer. This analyzer with Cold Vapor Atomic Absorption Spectrophotometry (CVAAS) has a detection limit of 0.05 μg/L. For samples below this detection limit, Milestone DMA-80 Direct Mercury Analyzer (following EPA method 7473), with a detection limit of 0.02 ng was applied for analysis. Both the two instruments are calibrated using the dilutions of a 1000 μg/mL certified mercury standard (State Nonferrous Metals and Electronic Materials Analysis and Testing Center, P/N GSB04-1729-2004). All samples were analyzed in triplicate or more times to obtain parallel results and 5% uncertainty was allowed between parallel results.

3. Development of the ore concentrates transport matrixes

The ore concentrates transport matrix were developed based on the import and export of concentrates for each province in China in 2010. The total concentrates supply of each province are taken from the Yearbook of Nonferrous Metals Industry of China (2011). The transportation data $C_{su,k \rightarrow ij}$ between provinces were based on the trade between ore mineral plants and the 244 smelters in our survey. For smelters without trade information (mostly in small smelters with discontinued production), we assumed that local concentrates were consumed in these smelters. Based on the above information, linear equations were established and solved. The zinc, lead and copper concentrates transport matrixes are shown in **Table S2, S3, S4**.

Table. S2. Zinc concentrates trade matrix between regions

Zinc		Consumption (kt)																	
		RG	AH	FJ	GS	GD	GX	GZ	HN	HuN	IM	JX	LN	QH	SaX	SC	XJ	YN	ZJ
Supply (kt)	RG	SU	2	11	249	271	516	22	291	1260	389	2	402	99	580	254	1	916	35
	OC	1458			24	167	58		141	227	59		252		323	23		182	
	AH	13	2							12									
	CQ	24								5	4							15	
	FJ	147		11						137									
	GS	203			188						5					10			
	GD	194				103	10		2	24								55	
	GX	337					337												
	GZ	26						22		4									
	HB	38								3	16		3					15	
	HN	60							53	2								5	
	HuN	583								583									
	IM	750					67		30	165	284		70	11	65			58	
	JS	21					8			3			10						
	JX	55								48		2						5	
	JL	22								2			20						
	LN	47											47						
	QH	84												84					
	SaX	211			25					14					163			9	
	SC	367					10		63	13	20					211		50	
	XJ	26			12				1						13		1		
	XZ	26									1			5		10		10	
	YN	560					25			8					15			512	
	ZJ	46								11									35

Table. S3. Lead concentrates trade matrix between regions

Lead		Consumption(kt)													
		RE	AH	GS	GD	GX	HN	HuN	IM	JX	LN	NX	QH	SaX	YN
Supply (kt)	RE	SU	76	42	87	113	1151	640	108	68	28	6	58	104	379
	OC	881	63		8	52	568	55		13	9				113
	AH	13	13												
	FJ	75						23							50
	GS	48		42										5	
	GD	127			79			11		16					21
	GX	238				61	63	75							39
	GZ	23					23								
	HB	14						14							
	HN	70					70								
	HuN	283						283							
	IM	443					221	59	108			6		37	14
	JS	15					15								
	JX	47					8			39					
	JL	42					29	13							
	LN	19									19				
	QH	68											58	10	
	SaX	52												52	
	SX	13					13								
	SC	208					92	88							28
	XJ	12.48						12							
	XZ	28.08													28
	YN	107					21								86
	ZJ	369					29	8							

Table. S4. Copper concentrates trade matrix between regions

Copper		Consumption(kt)																		
		RE	AH	GS	GX	HB	HN	HB	HN	IM	JX	JL	LN	SaX	SD	SX	SC	XZ	YN	ZJ
Supply (kt)	RE	SU	527	594	27	13	5	204	12	237	497	12	25	3	261	68	41	7	308	47
	OC	1733	312	405				103	6	160	295		10		261	39			120	21
	AH	128	128																	
	FJ	10																		10
	GS	74		71			2													
	GD	9			9															
	GX	7			7															
	HB	2				2														
	HlJ	2	1			1														
	HN	7				2	2													3
	HuB	55						55												
	HN	6							6											
	IM	170	49					40		76			4			2				
	JS	1																		1
	JX	208			5						202									
	JL	16	4									12								
	LN	12				1							11							
	QH	40		40																
	SaX	9						6						3						
	SD	10				8														2
	SX	27														27				
	SC	72	27	3			1										41			
	XJ	74		67														7		
	XZ	5	5																	
	YN	201		8	5														188	

	ZJ	9																		9

RE: Region; SU: Summation value; OC: Other countries; AH: Anhui; CQ: Chongqing; FJ: Fujian; GS: Gansu; GD: Guangdong; GX: Guangxi; GZ: Guizhou; HB: Hebei;

HlJ: Heilongjiang; HN: Henan; HuB: Hubei; IM: Inner Mongolia; JS: Jiangsu; JX: Jiangxi; JL: Jilin; LN: Liaoning; NX: Ningxia; XZ: Xizang; YN: Yunnan; ZJ: Zhejiang;

Table. S5. Application percentage of a certain type of APCD combinations in each province

Installation proportion of certain type of APCDs, θ (%)[a]																							
Process (i)		Zinc-EP						Zinc-EZF			Zinc-RZSP		Lead-SMP				Lead-SPP				Copper-FFSP			
The type of APCDs (l)		1	2	3	4	5	7	1	5	6	1	5	1	3	4	5	4	5	6	7	1	3	4	5
Province (j)	Anhui																	100						
	Fujian	100																						
	Gansu	95					5	100												100				
	Guangdong	100																						
	Guangxi	90			9	1							38	53	3	7	63	14	23			100		
	Guizhou	87	0	0	0	0	13	0	75	25	0	1												
	Hebei																				83	17		
	Heilongjiang																							
	Henan	49					51						66	34						100				
	Hong Kong																							
	Hubei																						100	
	Hunan	55	45								100		89			11				100	100			
	Inner Mongolia	100													100					100		100		
	Jiangxi						100																	
	Jilin																						100	
	Liaoning	100									95	5				100		1			59	41		
	Ningxia														0	100								

	Qinghai	37		63										100										
	Shaanxi	97		1		2			100		100		100							100			100	
	Shanxi						100															100		
	Sichuan	100									100										100			
	Xinjiang																							100
	Yunnan	87		1		12		79	21			100			100					100	100			
	Zhejiang	100																			100			

$0 \le \theta \le 1, \sum_{l} \theta_{l,ij} = 1$

Table. S6. Parameters for mercury release rate, distribution coefficient, removal efficiency, metal concentration and recovery rate

Metal	Process	Mercury release rate				distribution coefficient			mercury removal efficiency	metal content	metal recovery rate
		γ_d (%)	γ_s (%)	γ_e (%)	γ_r (%)	ξ_{of} (%)	ξ_{ss} (%)	ξ_{se} (%)	η_o (%)	α (%)	φ (%)
Zinc	EP	0.80[a]	99.4[a,c]	0.00	87.2[a,b]	0.55[d]	0.00	0.00	12.5[a,b]	50.5[g]	94.0[g]
	EZF	0.45[d]	99.4[d]	59.1[d]	0.00	0.55[d]	0.00	0.00	12.5[a,b]	50.5[g]	95.5[g]
	RZSP	0.45[d]	99.4[d]	59.1[d]	0.00	0.55[d]	0.00	0.00	0.00	50.5[g]	95.5[g]
	ISP	0.10[b]	99.1[b]	59.1[d]	0.00	1.00[b]	0.00	0.00	12.5[a,b]	50.5[g]	95.5[g]
	AZSP	0.00	99.4[d]	59.1[d]	0.00	0.55[d]	0.00	0.00	0.00	50.5[g]	95.5[g]
Lead	RPSP	0.00	98.9[b]	60.1[b]	93.7[b]	0.55[d]	0.02[b]	2.40[b]	34.7[b]	62.8[g]	96.8[g]
	SMP	0.10[d]	98.7[b]	58.0[b]	0.00	0.55[d]	0.00	14.4[b]	12.5[a,b]	62.8[g]	96.8[g]
	ISP	0.10[b]	99.1[b]	59.1[d]	0.00	1.00[b]	0.00	0.00[b]	12.5[a,b]	62.8[g]	96.8[g]
	SPP	0.00	98.8[d]	59.1[d]	0.00	0.55[b]	20.6	14.4[d]	0.00	62.8[g]	96.8[g]
Copper	FFSP	0.90[b]	97.7[b]	0.00[e]	0.00	0.55[d]	0.80[b,f]	0.00	34.7	21.7[g]	97.8[g]
	RPSP	0.00	98.1[b]	0.00[e]	90.0[d]	0.10[b]	1.80[b,f]	0.00	12.5[a,b]	21.7[g]	97.8[g]
	RE	0.00	97.9[d]	0.00[e]	0.00	0.55[d]	1.30[d]	0.00	12.5[a,b]	21.7[g]	97.8[g]
	IFSP	0.45[d]	97.9[d]	0.00[e]	90.0[d]	0.55[d]	1.30[d]	0.00	12.5[a,b]	21.7[g]	97.8[g]
	EF/RF	0.00	97.9[d]	0.00[e]	0.00	0.55[d]	1.30[d]	0.00	0.00	21.7[g]	97.8[g]

a. Wang et al., 2010

b. Zhang et al., 2012

c. Li et al., 2007

d. Estimated value

e. Smelting flue gas is mixed with extraction flue gas as primary flue gas in copper smelters. Smelting and extraction sector are regarded as one sector. Mercury release rate for primary flue gas includes that released from extraction process.

f. Include mercury in extraction slag.

g. The editorial board of Chinese nonferrous metal industry association, 2011

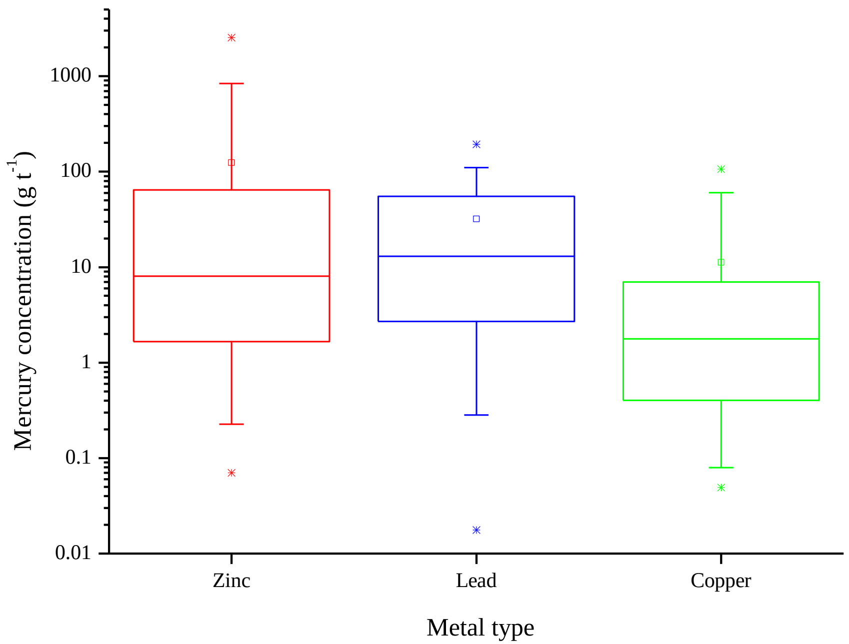

Fig. S1. Mercury concentration of concentrates from Chinese mines