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

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Ⓢ *Supporting Information*

ABSTRACT: This study investigated the flow of mercury (Hg) associated with zinc (Zn), lead (Pb), and copper (Cu) concentrates and provided new insights on the Hg emissions and recovery in both metals-production and wastes-disposal processes in China. Total Hg input from concentrates consumed in China reached 1005.4 t, of which 31.7% was dumped as discarded slags and 2.3% was stabilized (permanent storage). Approximately 202.1 t of Hg was directly emitted to air, water, and soil. More specifically, metals production processes emitted 100.4 t Hg to air. Wastes disposal processes contributed to an additional 47.8 t of atmospheric Hg emissions (which were ignored in most emission inventories) and 32.7 and 21.3 t of Hg to water and soil, respectively. At the same time, out of the 62.6 t of recovered Hg, 95.2% was reclaimed from acid slags. Interim storage of 398.9 t of Hg also highlights the significance of acid slags as potential Hg recovery sources due to the global ban on primary Hg production. The uncertainty ranges (confidence interval: 10%−90%) for Hg emissions to air, water, and soil and for Hg recovery were (−75%, 89%), (−96%, + 111%), (−120%, + 149%), and (−78%, 92%), respectively.
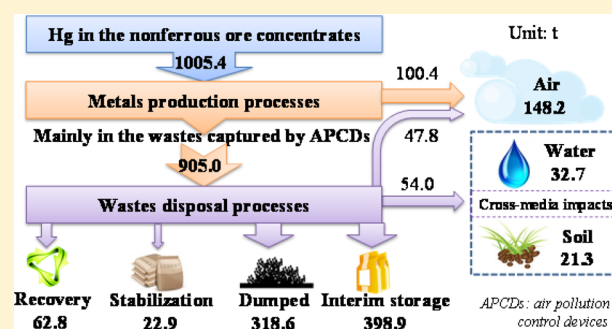
1. INTRODUCTION

Hg is a toxic pollutant that exists as Hg ores in the environment and is also associated with fossil fuels or metal ores in deposits. Fossil-fuels combustion and nonferrous-metals production have led to significant unintentional Hg emissions to the environments.[1,2] Coal-fired power plants and nonferrous metals smelting contributed to 100.0 and 97.4 t of China's atmospheric Hg emissions, respectively.[3] In the Minimata Convention on Mercury, both coal-fired power plants and nonferrous-metals-production processes (referred to as primary metals-production processes if not otherwise noted in the following text) were listed as regulated sources for emissions to air as well as releases to water and soil.[4]

Great efforts have been put forth to quantify historical and current atmospheric Hg emissions from nonferrous-metals production processes on global or regional scales. Emissions from Chinese nonferrous-metals production processes were estimated in the range of less than 54.5 to 320.5 t using average emission factors (Table S1).[5−11] Another estimation method is the technology-based method, which was used in most recent studies (Table S1).[1−3,12−15] All of these inventories provide insights on historical and present-day emissions from non-ferrous metal smelting to the atmosphere. However, it should be noted that atmospheric Hg emissions from Chinese nonferrous metals production processes only accounted for

13.4% of the Hg in the consumed concentrates in 2010.[1] The rest of Hg either remained in smelting slags (including furnace slags, leaching slags, and purification slags) or was captured from wastes by air pollution control devices (APCDs).[1,11,16,17] As discovered in recent field experiments in Chinese Zn smelters, disposal of these wastes represents an important atmospheric Hg emission source.[18] However, this component of emissions was not included in these inventories.[1−3,5−15] Additionally, neither emissions to water nor soil from whole-concentrates utilization processes (including both metals-production and wastes-disposal processes) were quantified. Previous Hg flow studies also indicate that these contributions are relevant to atmospheric Hg emissions.[19−22] Therefore, to effectively plan Hg pollution-control strategies, it is crucial to address the fates of Hg associated with nonferrous ore concentrates.[23−26]

The aim of this study was to quantify the fates of Hg associated with nonferrous ore concentrates by investigating Hg flow in both metals-production and wastes-disposal processes. The Hg flow information yielded from this study provided new

insights on where Hg emissions to air, water, and soil occurred during the whole-concentrates utilization processes. A comparison of the low rate of Hg recovery and the high percentage of interim storage highlighted the potential for Hg recovery in nonferrous metal smelters.

2. MATERIALS AND METHODS

2.1. Hg Flow Analysis. The Hg flow analysis approach was used to quantify the Hg distribution over its life cycle in a certain system. This approach enabled the identification and quantification of Hg into a study system, its temporal storage in the system, and its final fate. For the completion of the Hg flow analysis, four major steps were required, including goal and system definition, data acquisition, material balances, and modeling, and interpretation.[19,27] In this study, the target was designed to establish the flow of Hg associated with nonferrous ore concentrates (Zn, Pb, and Cu concentrates) in both metals-production and wastes-disposal processes. The geographical system boundary for our work was the political border of China. To construct the model and to confirm the flows within this model, we acquired information through literature review, expert judgment, best estimation, and direct interviews with the smelters. The data collected included Hg concentrations in ore concentrates, concentrates consumption, Hg removal efficiencies of pollution control devices (PCDs), application percentages of PCD combinations, and the raw materials and byproduct transportation matrix, among others. The collected data were subsequently used to construct the Hg flow model. To consider the probability distribution of the flow, we incorporated Monte Carlo simulations into the Hg model.[3] The Hg flow model consisted of three submodels, including an Hg input submodel, an Hg distribution submodel for the metals-production processes, and an Hg distribution submodel for the wastes-disposal processes (Figure 1). The probabilistic
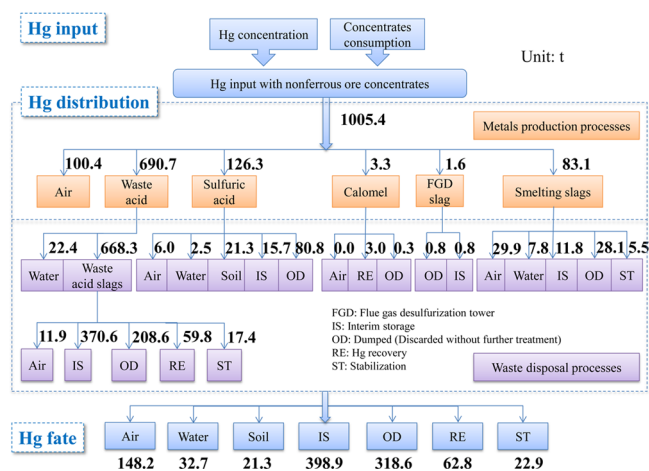
model are described in the section S1 of the Supporting Information, and a brief description is given in this section.

2.2.1. Hg Input Submodel. Hg input was calculated as the product of the Hg concentration in the consumed concentrates and the concentrates consumption. The Hg concentration in the consumed concentrates was converted from the Hg concentration in the produced and imported concentrates by introducing the concentrates import and interprovincial transport matrix. The key parameters of this submodel included the provincial concentrates consumption and Hg concentration in the produced and imported concentrates. The concentrates consumption data from the 244 smelters investigated in our previous study were updated by direct interviews with the smelters and expert consultations.[1] The concentrates consumption for smelters that lacked relative information was converted from metal production based on the metal contents of concentrates and the metal recovery rate. The provincial consumption of concentrates is shown in Table S2. The unique distribution curve with an uncertainty range of $\pm30\%$ was applied to represent the distribution function of concentrates consumption. The distribution function of the Hg concentration in the produced and imported concentrates was generated from the database described in our previous studies.[1,3] For provinces and imported countries with sufficient samples (no less than 15), the distribution functions were generated using the batch-fit function of the Crystal Ball software. The simulation results indicated that Hg concentration in these provinces fit the log-normal distribution. The key characteristics in the simulation curves included P10, P50, and P90 values. The meanings of these three values were explained by taking the simulation curve of the Hg concentration in the Zn concentrates produced in Hunan province as an example (69 samples). The P50 of 1.85 μg g^{-1} read from this figure indicated that a 50% probability that the actual result was no more than 1.85 μg g^{-1} (Figure 2).
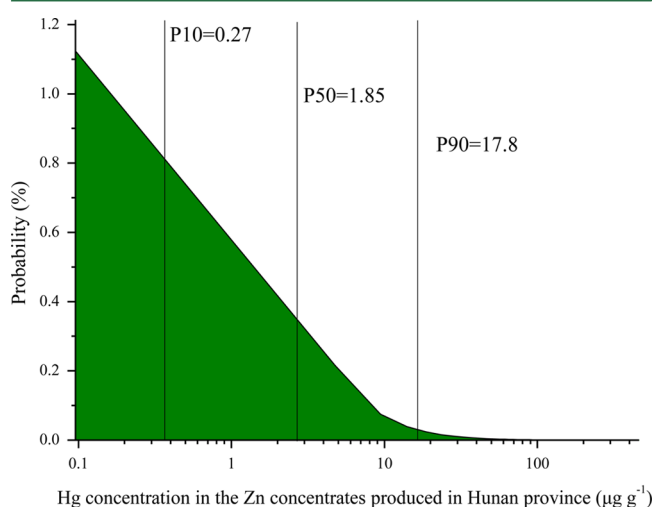

Figure 1. Flow of Hg associated with the nonferrous ore concentrates.

Figure 2. Simulation curve of the Hg concentration in the Zn concentrates produced in the Hunan province.

approach addressed the uncertainty issue of Hg emissions and recovery by representing the uncertainty of the key parameters using probability density functions and propagating the uncertainty through the Hg flow model via the Monte Carlo technique. Crystal Ball software was used for the Monte Carlo simulations in this study.[28−32]

2.2. Description of the Hg Flow Model and Key Parameters. The detailed construction steps of the Hg flow

Generally, the P50 value was treated as the best estimated value. The P10 value and P90 value of this figure indicated respective probabilities of 10% and 90% that the actual results were no more than 0.27 and 17.8 μg g^{-1} (80% chance). For provinces and imported countries that lacked sufficient concentrate samples, we simply used the minimum, the average

Table 1. Hg Removal Efficiency of Pollution-Control Devices (%)

| pollution-control devices[a] | | Hg removal efficiency (%) | | | | number of PCDs tested |
		mean	minimum	maximum	SD	
air-pollution-control devices	DC + FGS + ESD + DCDA + DFGD	98.5	98.0	98.7	0.4[b]	1
	DC + FGS + ESD + SMR + DCDA	99.2	99.1	99.3	0.2	2
	DC + FGS + ESD + DCDA	97.4	87.0	99.9	2.4	6
	DC + FGS + ESD + SCSA	86.5	86.5	86.5	2.7[b]	1
	DC + FGS	41.0	27.0	55.0	20.0	4
	FGS + ESD	82.5	72.5	99.5	10.9	7
	DC	12.0	2.0	20.0	7.0	4
	FGS	33.0	17.0	49.0	23.0	2
	FF	38.3	3.8	56.1	23.4	7
	WFGD	52.0	52.0	52.0	3.4[b]	1
	SMR	88.0	88.0	88.0	3.5[b]	1
waste-acid-pollution control devices	SET + NEU + ASR	95.0	87.2	99.5	6.0	4
	SET + SUL + NEU + ASR	98.9	96.7	100.0	1.9	3

[a]DC: dust collectors; FGS: flue-gas scrubber; ESD: electrostatic demister; DCDA: acid plants with double contact and double absorption towers; DFGD: dry flue-gas desulfurization towers; SMR: specific Hg removal devices; SCSA: acid plants with single-contact and single-absorption towers; FF: fabric filter; WFGD: wet flue-gas desulfurization towers; SET: settling stage; NEU: neutralization stage; ASR: arsenic removal; SUL: sulfurization and coordination stage. [b]The standard deviation of Hg removal efficiency from several tests for one air-pollution-control devices was used.

mean, and the standard deviation of the concentration in the samples to generate the log-normal distribution function. The key characteristics of the distribution curve for Hg concentration in the produced and imported concentrates are listed in Table S3.

2.2.2. Hg Distribution Submodel for the Metals Production Processes. The metals production processes generally consisted of four stages, including dehydration, initial oxidation (roasting or smelting of concentrates), extraction, and refining (Supporting Information section S1 and Figure S1). For each stage that used pyrometallurgical methods, the main parameters used to construct the model contained the Hg input with the raw materials to each stage, Hg release from the raw materials to the flue gas in the furnace, and Hg removal by the APCDs. The Hg input to the dehydration stage was equal to the Hg in the consumed concentrates. The raw materials for the other three stages were the products in the former stage, and the Hg in these products was calculated based on Hg mass-balance methods. The Hg released rate in the furnace of each stage was collected based on the field experiment results, as shown in Table S4.[11,16−18,33] The initial oxidation stage was the largest release point for air pollutants, including Hg. Thus, flue gas is typically subjected to a thorough dedusting, and additional Hg removal is required in certain cases prior to thorough desulfurization. Desulfurization technology generally included acid plants or acid plants plus flue gas desulfurization (FGD) towers.[34−36] These APCDs were combined into eight types in 2012. The application percentages for each type of APCD combination are shown in Table S5. The application percentage was assumed to be a unique distribution with an uncertainty on the order of ±5%. It was assumed that the dust collectors were used for flue gas from the dehydration, extraction, and refining stages. The Hg removal efficiencies of the APCDs from field experiments are summarized in Table 1.[11,16−18,33] Distribution fitting of the Hg removal efficiency could not be performed due to an insufficient number of tests available to generate the distribution curve, and thus, we assumed that the Hg removal efficiencies of the APCDs fit the normal distributions. The standard deviation of the Hg removal efficiency was used to generate the uncertainty. Most Hg in the flue gas was captured in the wastes and byproducts (including

furnace slags, waste acid, sulfuric acid, calomel, and FGD slags), which were generally treated in the wastes disposal processes.[37−40] In the Zn electrical process, dust and calcine byproducts from the initial oxidation stage were used to extract ZnO via a hydrometallurgical method (leaching, purification, and electrolysis).[18] Thus, the Hg was distributed into various types of purification slags, leaching slags, and waste acid (leachate); see Table S6. These wastes were also treated in the wastes-disposal processes.

2.2.3. Hg Distribution Submodel for the Wastes Disposal Processes. This submodel focused primarily on the Hg distribution in waste acid disposal (including calomel disposal), sulfuric acid disposal, smelting slags disposal, and FGD slags disposal processes (Supporting Information section S2 and Figure S1). In this work, smelting slags here included purification slags, leaching slag, and furnace slags. The disposal methods and assumptions applied for these wastes and byproducts are summarized in Table S7. The fates of the Hg in these wastes and byproducts were classified into seven types, including emissions to air, water, or soil, dumped (discarded without further disposal), interim storage, recovery, and stabilization. The term "dumped" implies that Hg was stored in the wastes, which were discarded without further treatment due to technology limitations or low economic profit. The dumped wastes were generally discarded in open pits or crude landfill sites lacking proper environmental pollution control. In this study, we did not consider Hg re-emission from these dumped wastes because limited documentations were available. However, the potential environmental risk urges for attention in future studies. The term "interim storage" means that Hg was stored in wastes and byproducts that might be used in the upcoming years. The Hg in the temporarily stored wastes and byproduct was thought to have little environmental impact during the storage period. However, once the wastes or byproduct are reused, the Hg contained will re-enter into industrial activity and could cause potential environmental impacts. The term "stabilization" means that the wastes were properly treated, such as cement solidification of arsenic slags (one type of acid slags produced in Cu smelters). It is next-to-impossible that Hg would be released into the environmental compartments in the normal conditions.

Hg distribution in the waste-acid-disposal processes was calculated by considering the Hg amount in the waste acid, the application percentage of different types of waste acid pollution control device (WAPCD) combinations, and the Hg removal efficiency of each combination. A total of two major types of WAPCD combinations were used. Type 1 involved the combination of settling (SET) + neutralization (NEU) + arsenic removal (ASR). Type 2 added one additional stage in addition to type 1 (i.e., the sulfurization and coordination (SUL) stage (Figure S2)). The application percentages of different types of WAPCD combinations for the smelters were primarily based on direct interview with the smelters and the assumption that the sulfuric and coordination stage was used in large-scale smelters (Supporting Information section S2). The application percentage was assumed to be a unique distribution, with uncertainty on the order of ±30%. The Hg removal efficiencies of the WAPCD combinations are shown in Table 1. The normal distribution function was also applied for the WAPCDs. The Hg distribution in the sulfuric acid disposal processes was calculated based on the distribution factor method, including Hg distribution by different sulfuric acid users (Table S8) and Hg distribution among different fates in certain users (Table S9). The various sulfuric acid users were classified into five groups, including smelters, fertilizer plants, chemical plants, concentrating mills, and dealers. The Hg distribution in different sulfuric acid users was calculated based on the Hg amount in the sulfuric acid and the transportation matrix of the sulfuric acid. The transportation matrix of the sulfuric acid was developed from the transaction details provided by the investigated smelters.[1] For smelters lacking relative information, the sulfuric acid transportation matrix for other smelters located in the same province was simply applied. The uncertainty of the sulfuric acid transportation matrix was assigned on the order of ±30%, and the unique distribution function was applied. The initial Hg distribution of different fates for certain sulfuric acid users generated primarily from the Hg distribution in both metals-production and waste-acid-disposal processes according to the technology applied by the user. These distribution factors were subsequently modified based on expert opinion and available information. The Hg distributions in the fertilizer plants were calculated based on on-site tests.[41,42] In the leaching-slags disposal processes, the Hg removal efficiency of the APCDs was also assumed to fit the normal distributions. The standard deviation of the Hg removal efficiency was used to generate the uncertainty.

The detailed construction steps of the Hg distribution submodel for the wastes disposal processes are described in the Supporting Information section S2. Uncertainties associated with other parameters were not introduced directly into the calculations. To partially accounted for this, we somewhat increased the uncertainties associated with ore consumption and the transportation matrix of sulfuric acid.

2.3. Uncertainty Analysis. The P10 and P90 values of the distribution curve were assigned as the lower limit and upper limit of the simulation results. The (P50 − P10)/P50 and (P90 − P50)/P50 values were the lower limit and upper limit of the uncertainty range with a confidence degree of 80%. This information was used to highlight the data uncertainty and to identify priority research areas.

3. RESULTS AND DISCUSSIONS

3.1. Hg Input and Fates Analysis. In 2012, the Hg inputs associated with consumed Zn, Pb, and Cu concentrates reached

768.9, 167.7, and 68.8 t, respectively (the P50 value from the simulation curve is used to discuss the results in Section 3.1−3.3). The Hg inputs caused by consumption of imported Zn, Pb, and Cu concentrates were approximately 8.8, 5.1, and 4.0 t, respectively. The Hg in the imported concentrates accounted for 1.8% of the total Hg input.

The Hg fates are shown in Figure 1. The Hg emissions to the environmental compartments reached 202.1 t, of which the emissions from the entire utilization processes of Zn, Pb, and Cu concentrates contributed 67.8%, 26.5%, and 5.7%, respectively. The Hg stored in the dumped slags was 318.6 t, which accounted for 31.7% of the total Hg input. This component of Hg was believed to impact high ecological and human-health risks because few environmental protection measures exist to prevent the release of pollutants from these wastes. Moreover, nonferrous metals production has exists for several decades in China, and this trend might continue due to the sharply increasing production of primary Zn, Pb, and Cu.[43] Thus, the large amount of slags dumped on the land might further threaten ecological safety unless proper control measures are taken. The amount of Hg temporarily stored in wastes and by-products reached 398.9 t. This component of Hg could be reinput to industrial activities when the wastes or by-products are reutilized. Recovered Hg was 62.8 t, which accounted for up 6.3% of the Hg in the consumed concentrates. Stabilized Hg was only 22.9 t, which was approximately 2.3% of total Hg input.

3.2. Hg Distribution in the Flow. The Hg in the concentrates followed different distribution pathways before encountering to their final fates (see Figure 1). In the metals production processes, approximately 100.4 t of Hg in the concentrates was emitted to the air. The remaining Hg was distributed in different intermediated wastes and byproducts, including waste acid, calomel, sulfuric acid, FGD slags, and smelting slags. Waste acid was the most significant Hg-containing waste. The amount of Hg was 688.2 t in the waste acid, which is 68.5% of the total Hg input. Thus, attention should be focused on the disposal of waste acid because it can determine the Hg distribution in the wastes disposal processes to a great extent. The amount of Hg released in calomel, sulfuric acid, and FGD slags were 3.3, 126.3, and 1.6 t, respectively. The Hg released into smelting slags was 83.1 t.

In the wastes-disposal processes, disposal of waste acid could remove 96.8% of Hg from the waste acid to the acid slags (see Figure 1). The total Hg removal efficiencies of the WAPCDs in Zn, Pb, and Cu smelters were 96.7%, 96.7%, and 97.7%, respectively. Thus, approximately 22.3 t of Hg in the waste acid was emitted to water, of which approximately 81.3% was emitted from Zn smelters. The remainder of the 668.3 t Hg was released into acid slags, mainly settling slag, sulfurization and coordination slag, neutralization slag, and arsenic slag (Figure S2). Approximately 370.6 t of the Hg in acid slags was temporarily stored in the smelters in the form of settling slag, arsenic slag, and sulfuric and coordination slag, and these materials accounted for 92.9% of the total interim storage of Hg. The high percentage of Hg interim storage represents the conjunction of technical, economic, and managerial factors. Only acid slag with Hg concentration greater than 1% could be disposed by the qualified Hg recovery companies, which are limited by current Hg recovery technology. Even so, these companies dominated the trade price because only two companies are qualified to recover Hg from the acid slags in China. Thus, smelters unsatisfied with the price chose to store

the slags. For smelters that produced slags with lower Hg concentration, the strict limitations on transportation of hazardous wastes and the fact of inadequately controlled landfill sites resulted in preservation of the acid slags by the smelters. Approximately 208.5 t of Hg in the acid slags was discarded, mainly as neutralization slag. Disposal of neutralization slag is one of the most difficult problems in Chinese nonferrous metal smelters due to its large production and low concentration of valuable metals. Certain smelters roasted the acid slags to reduce the amount of dumped and temporarily stored slags. However, this process further led to 11.9 t of atmospheric Hg emissions. The total Hg reclaimed from the acid slags was only 59.8 t, and most of this amount was reclaimed from slags produced in Zn smelters. Stabilized Hg was 17.4 t, accounting for 2.5% of the Hg in waste acid. This observation also indicated that wastes management was an arduous task for Chinese nonferrous metal smelters.

The Hg reclaimed from calomel in China was only 3.0 t, which accounted for approximately 0.9% of global Hg recovered from flue gas in 2005.[2] This percentage might be lower considering the increasing number of specific Hg recovery towers installed after 2005. Compared with the low Hg recovery percentage, production in China has resulted in 39% of refined Zn, 44% of refined Pb, and 29% of refined Cu out of the corresponding global production in 2012,[43] respectively. These results indicate a large Hg recovery potential from flue gas in Chinese nonferrous metal smelters.

The use of sulfuric acid led to 80.8 t of Hg dumped as slags and 15.7 t of Hg temporarily stored (see Figure 1). Approximately 6.0, 2.5, and 21.3 t of Hg were directly emitted to air, water, and soil, respectively. Smelters were the main points for atmospheric Hg emissions in sulfuric acid utilization processes. In the smelters, sulfuric acid was used as an acid solvent. A certain amount of the Hg in sulfuric acid was released with other insoluble matter as slags, and further high-temperature treatment of the slags led to the atmospheric Hg emissions.[18] Sulfuric acid used as a raw material for fertilizer production transferred 21.3 t of Hg to the fertilizer. We assumed that the Hg in the fertilizer was released to the soil, and the Hg re-emission from the soil was not considered in this study. Thus, the sulfuric acid from Zn, Pb, and Cu smelters contributed to release of 11.4, 5.1, and 4.8 t of Hg into soil, respectively (see Figure S4—S6). It should be noted that the actual Hg emissions to the soil in the form of fertilizer were underestimated because the Hg derived from phosphate ore concentrates (another raw materials used for fertilizer) was not considered in this study.[42]

Disposal of the smelting slags led to 29.9 t of Hg emissions to the air (see Figure 1). These atmospheric Hg emissions derived mainly from high-temperature treatment of leaching slags produced in the Zn smelters, which contributed to 23.3 t of atmospheric Hg emissions. The Hg amount emitted to water and discarded in dumped slags were 7.8 and 28.1 t. This portion of Hg originated primarily from hydrometallurgical processing of various types of purification slags. Approximately 11.8 t of Hg was temporarily stored in the products. The amount of stabilized Hg was approximately 5.5 t. This portion of Hg was primarily stabilized in building materials, such as use of furnace slags (mainly in the form of ferrous residues) in roadbeds. The remainder of Hg in the FGD slags was primarily dumped, and only a tiny amount was temporarily stored in chemical plants as ammonia sulfate slag or sodium sulfate slag.

3.3. Implications on Hg Emissions and Recovery.

Studies on the emissions of Hg associated with nonferrous ore concentrates have generally focused on metals production processes.[1,5,7,14] In this study, approximately 100.4 t of Hg was emitted to air in the metals-production processes, accounting for 10.0% of the total Hg input from ore concentrates. The atmospheric Hg emissions from Zn, Pb, and Cu production processes contributed to 55.5, 41.5, and 3.4 t, respectively, where the total Hg removal efficiencies of APCDs were 92.8%, 75.3%, and 95.1%, respectively. The APCDs trapped most of the Hg in flue gas into various wastes and by-products. However, by tracking the Hg flow in the wastes disposal processes, our study found that an additional 101.8 t of Hg was emitted to the environmental compartments. Thus, Hg emissions from wastes disposal processes require attention because their emissions were comparable with that from metals-production processes. In addition, the Hg emission pathways were more diverse in the wastes-disposal processes (Figure 3).
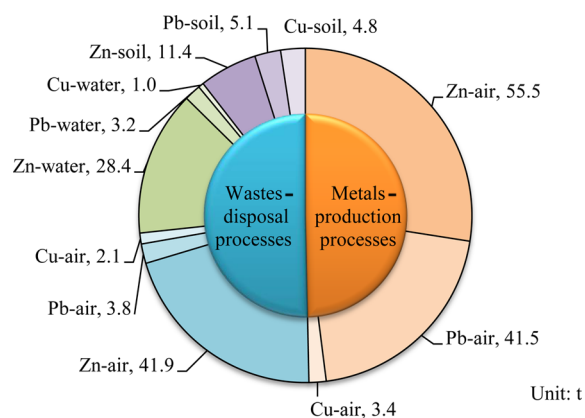

Figure 3. Hg emissions to air, water, and soil from different processes.

Approximately 47.8, 32.7, and 21.3 t of Hg were emitted to air, water, and soil, respectively, which indicates that if not safely managed, control of the atmospheric Hg emissions in the metals-production processes could lead to Hg re-emissions to the air from the wastes-disposal processes. This portion of the emissions was ignored in most atmospheric Hg emission inventories.[1−3,5−7,13,15] Moreover, disposal of the wastes and byproducts also could produce unintentional Hg emissions to water and soil, activities that are referred to as cross-media pollution effects. Therefore, end-of-pipe measures used to control Hg emissions from wastes disposal processes could become increasingly complicated due to more diverse emission points and lower Hg concentration in the wastes, liquid, or gas. Because the Hg flow identified the integrated effects of the APCDs traditionally used to reduce the atmospheric Hg emissions in the metals-production processes, efforts also should be focused on reducing atmospheric Hg emissions from the wastes-disposal processes as well as avoiding cross-media pollution effects. Additional control strategies targeted at reducing the amount of Hg in intermediated wastes should be taken into consideration before further utilization of these wastes (e.g., waste acid and sulfuric acid). In the long term, the best way to achieve the greatest reduction of Hg emissions is to produce refined metals using the scrap metals instead of ore concentrates. However, this measure generally requires a relatively good recycling system, mature recycling technology, and standardized management. Chinese government has started

to promoted nonferrous metals production from the scrap metals.[44−46]

Another implication was focused on Hg recovery in nonferrous metal smelters. Generally, flue gas from initial oxidation stage contained the largest amount of mobile Hg. Thus, specific Hg removal technology for flue gas was applied at this stage, and slags with high Hg concentration were produced, such as calomel. Therefore, the estimation of the amount of global Hg recovered was mainly based on the amount of Hg reclaimed from the slags produced by the specific Hg removal technologies.[2] Nevertheless, this estimation method underestimated the possible Hg recovery in nonferrous metal smelters. As indicated from our studies, approximately 59.8 t of Hg was recovered from the waste-acid slags, whereas only 3 t of Hg was reclaimed from the calomel. In addition, the Hg reclaimed from the acid slags only accounted for 9.0% of the total Hg in acid slags, whereas approximately 55.5% of the Hg in the acid slags was generally tightly sealed in the hazardous waste repositories of the smelters. This scenario indicates a great potential to reclaim Hg from acid slags in China. The recovery of Hg is often an issue in which a number of factors must be considered, such as market perspectives for Hg and available technologies. With increasing awareness of the environmental and health effects of Hg, it was expected that primary Hg production would continuously decrease, and the demand for Hg would be increasingly met by the recycling of Hg. For the further promotion of Hg recovery in China, many new measures should be applied, such as the development of new Hg recovery technologies that focus on low Hg-containing wastes.

3.4. Uncertainty Analysis. The P10, P50, and P90 values for Hg emissions and Hg recovery were obtained from the simulation curves (see Table S13), and the calculated uncertainty ranges are shown in Figure 4. The uncertainty
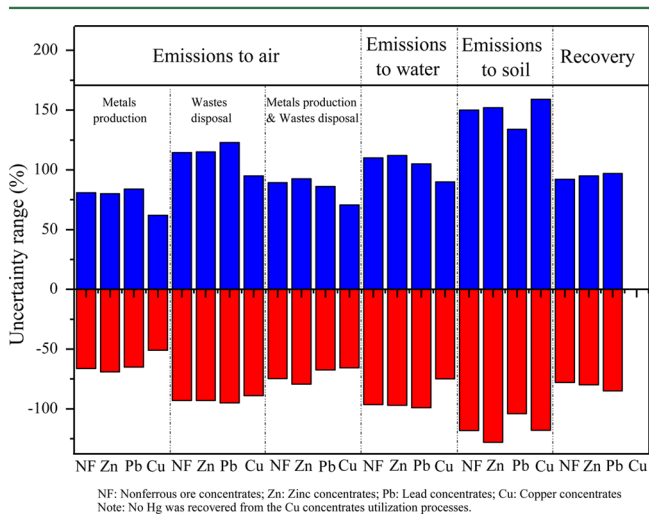

NF: Nonferrous ore concentrates; Zn: Zinc concentrates; Pb: Lead concentrates; Cu: Copper concentrates
Note: No Hg was recovered from the Cu concentrates utilization processes.

Figure 4. Uncertainty ranges of Hg emissions and recovery.

range for the atmospheric Hg emissions was (−75%, 89%) with 80% confidence. In the metals-production processes, the uncertainty range for atmospheric Hg emissions was (−67%, + 81%), for which the uncertainty ranges were (−69%, 80%), (−65%, 84%), and (−51%, 62%) for the Zn, Pb, and Cu production processes, respectively. The uncertainties mainly originated from the uncertainty of Hg concentrations in the concentrates. For example, the Hg concentration in Zn

concentrates from Gansu, Shaanxi, and Yunnan province contributed 31%, 22%, and 18% to the uncertainties of atmospheric Hg emissions from Zn production processes, respectively. The uncertainty range for atmospheric Hg emissions from wastes disposal processes was (−93%, + 115%). This uncertainty was primarily caused by the uncertainty of Hg input into the waste disposal processes and the Hg removal efficiency of the WAPCDs. The uncertainty ranges for Hg emissions to water, soil, and Hg recovery were (−96%, + 111%), (−120%, + 149%) and (−78%, 92%), respectively. Thus, the Hg emissions to soil showed the largest uncertainty, which was mostly due to the Hg amount in sulfuric acid and the uncertainty of the sulfuric acid transportation matrix.

The uncertainty analysis indicated that additional information on the wastes disposal processes was required to constrain the Hg flow. For instance, the application percentages of different types of WAPCD combinations require further investigation. Currently, a large amount of Hg is temporarily stored, and to evaluate the potential impacts of this stored Hg, a dynamic Hg flow should be established. In addition, Hg re-emissions from contaminated nonferrous metal production sites were not considered in this study and require further studies in the future.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information
The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b04934.

> S1, additional description of the atmospheric Hg emissions from nonferrous metal production processes (Table S1); S2, a detailed description of the Hg flow model (Tables S2−12, Figures S1−S3); S2, additional description of the flow of Hg associated with Zn, Pb, and Cu concentrates (Figures S4−S6); S3, the key parameters of the emission and recovery curve (Table S13). (PDF)

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Notes
The authors declare no competing financial interest.

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