**Atmospheric
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Update of mercury emissions from China's primary zinc, lead and copper smelters, 2000–2010

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Mercury emissions from China's primary zinc, lead and copper smelters, 2000–2010

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Abstract

China is the largest anthropogenic mercury emitter in the world, where primary nonferrous metal smelting process is regarded as one of the most significant emission sources. In this study, atmospheric mercury emissions from primary zinc, lead and copper smelters in China during 2000–2010 were estimated using a technology-based methodology with comprehensive consideration of mercury concentration in concentrates, smelting process, mercury removal efficiencies of air pollution control devices (APCDs) and installation rate of a certain type of APCD combination. Our study indicated that atmospheric mercury emission from nonferrous metal smelters in 2000, 2003, 2005, 2007 and 2010 was 67.6, 100.1 86.7 80.6 and 72.5 t, respectively. In 2010, the mercury in metal concentrates consumed by primary zinc, lead and copper smelters were 543 t. The mercury emitted into atmosphere, fly ash, other solids, waste water and acid was 72.5, 61.5, 2.0, 3774 and 27.2 t, respectively. Mercury retrieved directly from flue gas as byproduct of nonferrous metal smelting was about 2.4 t. The amounts of mercury emitted into atmosphere were 39.4, 30.6 and 2.5 t from primary zinc, lead and copper smelters, respectively. The largest amount of mercury was emitted from Gansu province, followed by Henan, Yunnan, Hunan, Inner Mongolia and Shaanxi provinces. The average mercury removal efficiency was 90.5 %, 71.2 % and 91.8 % in zinc, lead, and copper smelters, respectively.

1 Introduction

Researches on atmospheric mercury emission from major sources have been intensively carried out in the past several years due to worldwide concern on mercury contamination (Strode et al., 2009; Li et al., 2009, 2012; Lin et al., 2010; Wu et al., 2010; Tian et al., 2010; Kocman et al., 2011; Fukuda et al., 2011). Nonferrous metal smelting process is believed to be one of the most significant anthropogenic mercury emission sources in the world. Global atmospheric mercury emission from nonferrous metal

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smelters reached 310 t, of which about 203 t was emitted from China in 2007 (Streets et al., 2005; Wu et al., 2006; Hylander and Herbert, 2008; Pirrone et al., 2010; Wang et al., 2010).

The main factors affecting atmospheric mercury emission from nonferrous metal smelters include mercury concentration in ore concentrate, smelting technology, the type of APCD combination applied and the installation rate of a certain type of APCD combination. Current inventories about atmospheric mercury emission from China's zinc, lead and copper smelters are subject to high uncertainty due to the following reasons.

First, mercury content in ore concentrates was reported over a wide range and there were few data about mercury concentration in Chinese concentrates. Global results about mercury content in concentrates from Brook Hunt indicated that the maximum are 6000, 325 and 1500 $g\,t^{-1}$ for zinc, lead and copper concentrates, respectively, while the minima are all less than 1 $g\,t^{-1}$ (Hylander and Herbert, 2008). However, no data about China's mines were collected in this report. Streets et al. (2005) reported that mercury concentration in Chinese zinc concentrates varied from less than 1 $g\,t^{-1}$ to more than 1000 $g\,t^{-1}$. Yin et al. (2012) pointed out that such wide range depended on the ore types and their geneses. Data about mercury concentration in Chinese lead and copper concentrates are scarce.

Second, results from field measurement about mercury removal efficiency of APCDs were limited in previous inventories. Mercury removal efficiencies were estimated on the basis of sulfur abatement technology. About 99 % of gaseous mercury was estimated to be removed from flue gas in copper smelters with double-contact sulfuric acid plants or in zinc/lead smelters with both double-contact plants and mercury removal tower. Mercury removal efficiency was regarded as 95 % for copper smelters with single-contact sulfuric acid plants or zinc/lead smelters with sulfuric acid plants (Hylander and Herbert, 2008). In order to reduce the uncertainty of mercury emission inventory, field measurements have been conducted in China's zinc, lead and copper smelters in the past several years. The total mercury removal efficiency for tested

smelters is from 99.2 % to 99.8 % (Li et al., 2010; Wang et al., 2010; Zhang et al., 2012).

Third, various smelting processes and APCDs are used in China's smelters and they have been improved in the past decade because of the stringent regulations for environmental protection. Therefore, the emission factors used in previous studies will not apply to current situation since installation rate of the types of APCD combinations in smelters have been undergoing change. Streets et al. (2005) adopted the average mercury emission factors of 86.6, 43.6 and 9.6 g t^{-1} for zinc, lead and copper, respectively, mainly based on the average mercury concentration in concentrates without consideration of APCDs. Hylander and Herbert (2008) estimated the emission factors of 16.61, 14.91 and 6.72 g t^{-1} for zinc, lead and copper smelters, respectively, in the global inventory of 2005 for China's nonferrous metal smelters. However, the increased installation rate of acid plants after 2005 indicates that these emission factors are not applicable to China.

In this paper, nationwide as well as imported concentrates have been sampled and analyzed for mercury content. Up-to-date mercury removal efficiencies in existing literatures have been summarized and applied in this study. Moreover, information on smelting technologies as well as APCDs has been investigated nationwide. A technology-based method with comprehensive consideration of the above factors is used to estimate atmospheric mercury emissions from primary zinc, lead and copper smelters in China during 2000–2010.

2 Methodology

Various smelting processes are used in China's nonferrous metal smelters. Zinc smelting processes include oxygen pressure leaching process (OPLP), electrolytic process (EP), imperial smelting process (ISP), retort zinc smelting process (RZSP) or electric zinc furnace (EZF). There is no atmospheric mercury emission from OPLP since it is a hydrometallurgical process and mercury in ore concentrate is released into water

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or solid waste. Lead smelting processes can be 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 include flash furnace smelting process (FFSP), rich-oxygen pool smelting process (RPSP), imperial furnace smelting process (IFSP), roasting-leaching-electrolyzing process (RLEP) as well as the old technologies that were forbidden by Chinese government such as electric furnace smelting process (EF) and revelatory furnace smelting process (RF).

2.1 Mercury input model

In all the above processes, although additives, such as quartz stone, limestone, also contain limited mercury, ore concentrate is the main source of mercury input. Mercury input Q for smelters with j technology in i province can be calculated using the following equation.

$$Q_{ij} = [\text{Hg}]_{\text{com},ij} C_{\text{com},ij} = \sum_k [\text{Hg}]_{\text{su},k \to ij} C_{\text{su},k \to ij} \qquad (1)$$

where, $[\text{Hg}]_{\text{com},ij}$ and $C_{\text{com},ij}$ are mercury content and amount of the ore concentrates consumed by j technology in i province; $[\text{Hg}]_{\text{su},k \to ij}$ and $C_{\text{su},k \to ij}$ are mercury content and supply of the ore concentrates produced in k province (or other countries) which are transported to i province and used by j technology for smelting. Concentrate supplies (see Table 1) are taken from the "Yearbook of nonferrous metals industry of China (2011)". Information about concentrates consumption is from our survey (see Table 1).

In order to get the mercury content in ore concentrates, 351 zinc concentrate samples, 190 lead concentrate samples and 174 copper concentrate samples were collected from 118 zinc mines, 83 lead mines and 55 copper mines, respectively. Besides, 39 zinc concentrate samples, 8 lead concentrate samples and 33 copper concentrate samples were also collected from imported concentrates. The imported zinc concentrate samples were from America, 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 F732-V Intelligent Mercury Analyzer with Cold Vapor Atomic Absorption Spectrophotometry (CVAAS), which has a detection limit of 0.05 μg l^{-1} was used to analyze mercury content in ore concentrates. For samples below the detection limit, Milestone DMA-80 Direct Mercury Analyzer, with a detection limit of 0.02 ng was applied for analysis. The geometric mean of mercury contents in all mines from one province was regarded as the mercury content of concentrates produced in this province.

The results indicate that most metal concentrates from Chinese mines have low mercury content, typically less than 10 g mercury t^{-1} copper concentrates, or 20 g mercury t^{-1} zinc/lead concentrates (see Fig. 1). However, the maximum of mercury contents are 2534.06, 193.00 and 106.54 g t^{-1} for zinc, lead and copper concentrates, respectively. The average mercury contents are 126.18, 33.08 and 3.16 g t^{-1} for zinc, lead and copper concentrates while geometric means are 9.74, 10.29 and 2.87 g t^{-1} for zinc, lead and copper concentrates, respectively. The distribution of mercury contents of concentrates from Chinese mines is quite close to global results from Brook Hunts in Hylander and Herbert's (2008) study. The geometric means of mercury content in ore concentrates imported from other countries are 9.04, 3.16 and 0.88 g t^{-1} for zinc, lead and copper concentrates, respectively.

The geometric means of mercury content in ore concentrates from different provinces show substantial differences (Table 2). For instance, the mercury content in zinc concentrates from Gansu province is 499.91 g t^{-1} while that from Xizang province is only 0.23 g t^{-1}. Mercury content in the concentrates consumed in each province is calculated according to the concentrates trade and transport among provinces (Table 3). Based on the mercury content and amount of concentrates consumed in each province, the national weighted average of mercury content of zinc, lead and copper concentrates consumed by China's smelters was 40.27, 20.03 and 2.25 g t^{-1}, respec-

tively, in 2010; while the corresponding results are 47.02, 16.81 and 2.82 g t^{-1}, respectively, in 2005.

2.2 Mercury emission model

Mercury in ore concentrates is released in the form of gaseous mercury during pyrometallurgical extraction process and parts of them are captured by APCDs and enter into waste water, acid or fly ash. Usually, pyrometallurgical extraction of nonferrous metals from concentrate requires dehydration, smelting/roasting, extraction and reclaiming/refining. Total atmospheric mercury emission from one smelter includes the sum of emission from the above four procedures. Mercury emission from smelting flue gas, excluding overflow flue gas, is called as primary flue gas emission (E_p). Mercury emission from dehydration, overflow, extraction and refining/reclaiming flue gas is regarded as other emission (E_o). The atmospheric mercury emission for smelters with j technology in i province can be calculated with the following equation.

$$E_{ij} = E_{p,ij} + E_{o,ij} \tag{2}$$

The mercury removal effect of APCDs has been proved in previous studies (Wang et al., 2010; Li et al., 2010; Zhang et al., 2012). Broadly speaking, APCDs for primary flue gas in most nonferrous metal smelters consist of dust collectors (DC) including cyclone dust collector, waste heat boiler, electrostatic precipitator and fabric filter (or their combination), flue gas scrubber (FGS), electrostatic demister (ESD), mercury reclaiming tower (MRT), and conversion and absorption tower (CAT). The CAT can be further divided into double conversion double absorption (DCDA) tower and single conversion single absorption tower (SCSA). The information of APCDs in most smelters is provided by Chinese nonferrous metal industry association or collected through literature research and field investigation. The proportion of metal production from smelters with different types of APCDs is given in Table 4. Combining the effect of APCDs and mercury flow diagram in smelters (Fig. 2), atmospheric mercury emission from primary flue

gas is calculated with the following equation.

$$E_{\mathrm{p},ij} = \sum_l \theta_{l,ij} Q_{ij} (1 - \gamma_{\mathrm{d},j}) \gamma_{\mathrm{s},j} (1 - \xi_{\mathrm{of},j})(1 - \eta_l) \tag{3}$$

where E is atmospheric Hg emission (kg); Q is mercury input (kg); p refers to primary smelting flue gas; i refers to province; j refers to technology; of refers to overflow flue gas; d refers to dehydration sector; l is the type of APCD combinations (Table 4); γ is mercury release rate (Table 5); ξ is mercury distribution coefficient (Table 5); θ is installation rate of a certain type of APCD combinations (Table 6); η is mercury removal efficiency of APCD (Table 7).

For most processes, dust collectors are widely installed for dehydration, overflow, extraction and refining/reclaiming flue gas. In several large smelters with advanced smelting processes, desulfurization devices are installed for flue gas control. No APCDs are installed for the other flue gas in the out-of-date processes such as AZSP, RZSP and EF/RF. Thus, different mercury removal efficiencies for other flue gas are given according to the smelting processes applied (see Table 5).

$$
\begin{aligned}
E_{\mathrm{o},ij} &= E_{\mathrm{d},ij} + E_{\mathrm{of},ij} + E_{\mathrm{e},ij} + E_{\mathrm{r},ij} \\
&= Q_{ij}\gamma_{\mathrm{d},j}(1 - \eta_{\mathrm{o},j}) \\
&\quad + Q_{ij}(1 - \gamma_{\mathrm{d},j})\gamma_{\mathrm{s},j}\xi_{\mathrm{of},j}(1 - \eta_{\mathrm{o},j}) \\
&\quad + Q_{ij}(1 - \gamma_{\mathrm{d},j})(1 - \gamma_{\mathrm{s},j} - \xi_{\mathrm{ss},j})\gamma_{\mathrm{e},j}(1 - \eta_{\mathrm{o},j}) \\
&\quad + Q_{ij}(1 - \gamma_{\mathrm{d},j})(1 - \gamma_{\mathrm{s},j} - \xi_{\mathrm{ss},j})(1 - \gamma_{\mathrm{e},j} - \xi_{\mathrm{se},j})\gamma_{\mathrm{r},j}(1 - \eta_{\mathrm{o},j})
\end{aligned}
\tag{4}
$$

where o refers to other flue gas; d, s, e, r refers to dehydration, smelting/roasting, extraction and refining/reclaiming, respectively. Q is mercury input (kg); ξ is called as distribution coefficient (Table 5); ξ_{ss} and ξ_{se} here refers to the proportion of mercury flows into the solids that are not sent to the next sector in smelting and extraction sector, respectively (Table 5); η_{o} is the mercury removal efficiency for other flue gases (see Table 5).

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Atmospheric mercury emission from i province is calculated by

$$E_i = \sum_j E_{ij} \tag{5}$$

Atmospheric mercury emission from j process is calculated by

$$E_j = \sum_i E_{ij} = EF_j \times M_j = EF_j \times \sum_i C_{\mathrm{com.,}ij} \times \alpha_j \times \varphi_j \tag{6}$$

Thus, the average emission factor for j process is

$$
\begin{aligned}
EF_j = {} & \frac{1}{\sum_i C_{\mathrm{com,}ij} \times \alpha_j \times \varphi_j} \times \Bigg[\sum_i \sum_l Q_{ij}(1-\gamma_{\mathrm{d,}j})\gamma_{\mathrm{s,}j}(1-\xi_{\mathrm{of,}j})\theta_{l,ij}(1-\eta_l) \\
& + \sum_i Q_{ij}\gamma_{\mathrm{d,}j}(1-\eta_{\mathrm{o,}j}) + \sum_i Q_{ij}(1-\gamma_{\mathrm{d,}j})\gamma_{\mathrm{s,}j}\xi_{\mathrm{of,}j}(1-\eta_{\mathrm{o,}j}) \\
& + \sum_i Q_{ij}(1-\gamma_{\mathrm{d,}j})(1-\gamma_{\mathrm{s,}j}-\xi_{\mathrm{ss,}j})\gamma_{\mathrm{e,}j}(1-\eta_{\mathrm{o,}j}) \\
& + \sum_i Q_{ij}(1-\gamma_{\mathrm{d,}j})(1-\gamma_{\mathrm{s,}j}-\xi_{\mathrm{ss,}j})(1-\gamma_{\mathrm{e,}j}-\xi_{\mathrm{se,}j})\gamma_{\mathrm{r,}j}(1-\eta_{\mathrm{o,}j}) \Bigg]
\end{aligned}
\tag{7}
$$
$$\tag{8}$$

where α is metal concentration (Table 5); φ is metal recovery rate of smelting process (Table 5).

Mercury captured by dust collector deposits on particles and remains in the fly ash. Some mercury is washed by water in the FGS or ESD while some of them flow into the sulfuric acid in the CAT. A limited part of them is recovered from flue gas in the form of calomel as byproduct. There is still trace amount of them remaining in other solids, including sludge and byproduct.

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3 Results and discussion

3.1 Mercury fate in China's nonferrous metal smelters

In 2010, total mercury input into China's primary nonferrous metal smelters with the consumption of ore concentrates in 2010 was 543 t, of which 74.8 %, 19.5 % and 5.7 % was input into zinc, lead and copper smelters, respectively. At the same time, with continuous expansion of smelter capacity and increased production, concentrates import become one way to solve China's shortage of concentrates. Thus, about 17.5 t of mercury from other countries entered into China's nonferrous metal smelters due to concentrates trade, in which 75.4 %, 15.9 % and 8.7 % went into zinc, lead and copper smelters in 2010. The amounts of mercury in the metal concentrates consumed by each province in 2010 were shown in Fig. 3. The mercury inputs in Gansu, Shaanxi and Yunnan province were much larger than that in other province due to the high mercury contents in their zinc concentrates.

The mercury in ore concentrate is either adsorbed on fly ash, dissolved in the waste water or acid, recovered as a byproduct, emitted into atmosphere or remained in other solids. The mercury emitted into atmosphere, fly ash, other solids (sludge or byproduct), water and acid in 2010 was 72.5, 61.5, 2.0, 377.4 and 27.2 t, respectively. The distribution of mercury output for zinc, lead and copper smelters is shown in Fig. 4. More than 50 % of mercury went into water in all of these three kinds of smelters. Mercury in fly ash and sulfuric acid was about 10 % and 5 %, respectively. Mercury in other solids was less than 2 %. There was no mercury recovered from flue gas in lead and copper smelters, while about 2.4 t of mercury was retrieved in the form of calomel as byproduct in zinc smelters. The percentage of mercury emitted into atmosphere in lead smelters was 29.0 %, much higher than that in zinc and copper smelters.

3.2   Provincial atmospheric mercury emission from primary smelters in 2010

In 2010, mercury emitted into atmosphere was about 72.5 t from China's primary non-ferrous metal smelters. Emission from primary zinc, lead and copper smelters was 39.4, 30.6 and 2.5 t, respectively. The largest mercury emitter was Gansu province, fol-

5   lowed by Henan, Yunnan, Hunan, Inner Mongolia and Shaanxi provinces. Summation of the emission from these six provinces accounted for 87.9 % of the national emission (Fig. 5).

China's zinc smelters emitted 39.4 t of mercury into atmosphere in 2010. Gansu, Yunnan, Shaanxi and Henan provinces were the top four emitters. For zinc smelters, sum-

10   mation of mercury emission from these four provinces accounted for 80.5 % of national amount. High mercury content of zinc concentrate consumed was the main reason for the high mercury emission in Gansu and Shaanxi province. For example, mercury concentration in the concentrates consumed by zinc smelters in Gansu province was as high as 403.4 g t^{-1}, which is about 10 times higher than the national average. Thus,

15   total mercury input into zinc smelters reached 181 t in Gansu province. If national average was used, this value would be only 18 t. High mercury emission in Yunnan and Henan is caused by the low installation rate of acid plants, which is only 79.3 % and 48.5 %, respectively.

Atmospheric mercury emission from lead smelters was about 30.6 t. Mercury emis-

20   sion from China's lead smelters was mainly from Henan, Hunan, Yunnan and Inner Mongolia. The emissions of these four provinces accounted for 89.6 % of total emission from lead smelters. Huge concentrates consumption, more than 60 % of national consumption, was the most important factor for the high mercury emission from lead smelters in Hunan and Henan. High mercury concentration in the concentrates con-

25   sumed in Inner Mongolia contributed to its high emission while low mercury removal efficiency led to the high emission in Yunnan's lead smelters.

Copper smelters emitted 2.5 t of mercury in 2010 and nearly half was emitted in Yunnan province. High mercury content of copper concentrates consumed in local smelters

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was the main reason for the large mercury emission in Yunnan province. Mercury content in the ore concentrates consumed by smelters in Yunnan province was $8.7\,\mathrm{g\,t}^{-1}$, about four times of the national average $(2.3\,\mathrm{g\,t}^{-1})$.

3.3 Atmospheric mercury emission from various smelting processes in 2010

In 2010, China's production of zinc, lead and copper from primary smelters reached 5033, 2794 and 2921 kt, respectively. For primary zinc smelters, about 2.5 % of refined zinc is produced with hydrometallurgical process. Refined zinc produced by EP, ISP, RZSP, EZF and others, accounted for 78.7 %, 7.1 %, 7.9 %, 1.3 % and 2.5 % of total zinc production, respectively. For primary lead smelters, the percentages of lead produced by RPSP, ISP, SMP and SPP were 47.3 %, 5.1 %, 20.2 % and 27.4 %, respectively. Refined copper produced by FFSP, RPSP, IFSP, RLEP and EF/RF, accounted for 34.2 %, 52.4 %, 9.8 %, 0.2 % and 3.4 %, respectively.

For zinc smelters, most of mercury is emitted from smelters with EP. Mercury emission from RZSP, EZF, ISP and AZSP was 6.3 %, 2.4 %, 5.4 % and 14.4 %, respectively. For lead and copper smelters, more than half of mercury was emitted from smelters with out-of-date technologies (Fig. 6). Besides, the average mercury removal efficiency of air pollution control devices in zinc, lead and copper smelters was 90.5 %, 71.2 % and 91.8 %. The mercury emissions can be further reduced by improving the mercury removal efficiencies of current APCDs or installing mercury reclaiming tower.

3.4 Historical changes of mercury emission from primary nonferrous metal smelters

According to our estimation, atmospheric mercury emission from nonferrous metal smelters in 2000, 2003, 2005, 2007 and 2010 was 67.6, 100.1, 86.7, 80.6, and 72.5 t, respectively. At the same time, the refined metal production from primary smelters has been increasing from 3909 kt to 4958, 6460, 8190 and 10749 kt, respectively (see Figs. 7, 8). The increased application of acid plants was the main reason for atmo-

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spheric mercury abatement in the past decade. Broadly speaking, the suitability of flue gas for making acid depends on its SO_2 concentration, which is determined by smelting process. Flue gas from process such as IFSP or SMP has a SO_2 concentration lower than 3.5 % and cannot be used to produce sulfuric acid. In that case, other flue gas desulfurization technologies such as ammonia absorption are applied. Flue gas produced from pool smelting process, such as RPSP, usually has a SO_2 concentration higher than 3.5 % and can be used to produce sulfuric acid.

In 2000, only 18 sulfuric acid plants were installed in China's zinc, lead and copper smelters due to the smelting processes (Lin, 2001). The installation rate of acid plants slightly increased from 60.9 %, 30.7 % and 61.0 % in 2000 into 63.9 %, 34.2 % and 61.0 % in 2003 for zinc, lead and copper smelters, respectively. This improvement was mainly caused by the increased metal production in large smelters. However, such effect was weakened by the establishment of many small-scale smelters with poorly or limited air pollution control devices. Thus, the total atmospheric mercury emission from zinc, lead and copper smelters increased from 67.6 t in 2000 to 100.1 t in 2003. With increasing attention to environmental protection, many smelters with heavy pollution were phased out or reconstructed to reduce air pollution. In 2005, about 69 sulfuric acid plants were established in primary zinc, lead and copper smelters (Guo and Huang, 2007). The percentage of zinc, lead and copper production from smelters with acid plants in 2005 increased to 76.3 %, 43.7 % and 70.5 %, respectively (Fig. 9). In order to standardize nonferrous metal industry, the construction conditions for copper and zinc/lead smelters were successively promulgated in 2006 and 2007, where both best available technologies and double contact sulfuric acid plants are required for new smelters (NDRC, 2006, 2007). Moreover, the elimination of old technologies were enforced after 2008 and about $400\,kt\,yr^{-1}$ of zinc out-of-date productivities, $600\,kt\,yr^{-1}$ of lead out-of-date productivities and $300\,kt\,yr^{-1}$ of copper out-of-date productivities were required to be phased out before the end of 2011, which also had positive impact on mercury emission in China (The State Council, 2010). In 2010, the percentage of zinc,

lead and copper production from smelters with acid plants reached 87.8 %, 65.5 % and 95.6 %.

Mercury emission was further reduced after 2010 because of "the 12th five year national plan for comprehensive prevention and control of heavy metal pollution". In this plan, China has set up a target that in 2015, the mercury emission in key areas will be reduced 15 % on the basis of 2007 emission level and mercury emission in other areas will be kept at the emission level of 2007.

3.5 Comparison with previous studies

In previous mercury emission inventory studies, emission factor method was used and the difference of mercury emissions was mainly caused by the uncertainty of emission factor (Tables 8, 9). In earlier estimates, the mercury emission factor for China's nonferrous metal smelters was regarded as same as that for other countries (Nriagu et al., 1988; Pacyna et al., 1996). Pirrone et al. (1996) assumed the mercury emission factors for zinc and lead smelters in developing continents to be 25 and 3 $\mathrm{g\,t}^{-1}$ metal produced, respectively. But there were no data for developing countries including China. Wu et al. (2006) and Wang et al. (2006) analyzed the mercury content in concentrates and estimated the mercury emission factor to be 86.6, 43.6 and 9.6 $\mathrm{g\,t}^{-1}$ for zinc, lead and copper smelters, respectively. However, these values were proved to be overestimated since the synergistic mercury removal effect of APCDs was not considered (Feng et al., 2004; Li et al.,2010; Wang et al., 2010; Zhang et al., 2012).

According to the research by Hylander and Herbert (2008), the total atmospheric mercury emission from China's zinc, lead and copper smelters reached to 83 t in 2005, which is similar to our estimation. However, the emissions from each of the three sectors in these two inventories are quite different (Fig. 8), which is mainly caused by the difference of mercury content in ore concentrates consumed by smelters. National average of mercury content in zinc, lead and copper concentrates consumed by smelters reached to 47.02, 16.81 and 2.82 $\mathrm{g\,t}^{-1}$, respectively. However, global mercury concentration of 10, 9 and 3.5 $\mathrm{g\,t}^{-1}$ for zinc, lead and copper concentrates was used in the

former study. Besides, the application rate of acid plants in 2005 was about 76.3 %, 43.7 % and 70.5 % for zinc, lead and copper smelters (Fig. 9), which was also higher than Hylander and Herbert's estimation. Even in some zinc or lead smelters without acid plant, FGS or other desulfurization devices are installed for air pollution control, of which mercury removal efficiency is higher than 10 %.

4   Conclusion

In this paper, we have presented an updated estimate of mercury emissions from non-ferrous metal smelters using a detailed technology-based methodology specifically for China. We estimate that the mercury emission from zinc, lead and copper smelters in China increased by 48.1 %, from 67.6 t in 2000 to 100.1 t in 2003. After 2003, the mercury emission decreased 27.6 %, from 100.1 t in 2003 to 72.5 t in 2010 although the production of zinc, lead and copper increased 116.7 % at the same period. The mercury reduction is mainly because of the improvement of smelting process and the increase of application rate of acid plants, from 60.9 %, 30.7 % and 61.0 % in 2003 to 87.8 %, 65.5 % and 95.6 % in 2010 for zinc, lead and copper smelters, respectively.

In 2010, the mercury emitted into atmosphere, fly ash, waste water, sulfuric acid, and other solids (sludge or byproduct), was 72.5, 61.5, 377.4, 27.2 and 2.0 t, respectively. Mercury retrieved directly from flue gas as byproduct of nonferrous metal smelting was 2.4 t. The amounts of mercury emitted into atmosphere were 39.4, 30.6 and 2.5 t from primary zinc, lead and copper smelters. The average mercury removal efficiency of air pollution control devices in zinc, lead and copper smelters was 91 %, 71 % and 92 %, respectively.

With the deepening understanding of mercury fate in nonferrous metal smelters, atmospheric mercury emission estimates based on techniques and mercury abatement devices lower the estimation uncertainty. However, mercury removal efficiency is still in wide range according to current studies and mercury removal mechanism of APCDs is still unclear.

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Table 1. Supply and consumption of ore concentrates.

Region	Concentrates supply (metallic, kt)			Concentrates consumption (metallic, kt)		
	Zinc	Lead	Copper	Zinc	Lead	Copper
Anhui	13.46	13.04	128.21	1.91	76.16	526.68
Beijing						
Chongqing	23.89					
Fujian	147.45	74.77	10.04	10.82		
Gansu	202.93	47.59	73.74	248.99	42.31	593.78
Guangdong	193.97	126.76	9.22	270.89	87.05	
Guangxi	337.43	237.94	7.42	515.75	112.92	27.19
Guizhou	25.61	23.45		21.98		
Hainan						
Hebei	37.60	13.69	1.72			13.30
Heilongjiang			2.14			
Henan	59.90	69.52	7.27	290.80	1153.12	5.13
Hong Kong						
Hubei			54.59			203.51
Hunan	583.02	283.40	6.44	1260.13	640.48	12.41
Inner Mongolia	750.11	443.48	170.29	389.42	107.50	236.60
Jiangsu	21.09	15.09	1.39			
Jiangxi	55.17	46.97	207.54	1.95	67.88	497.35
Jilin	22.04	41.90	16.30			12.27
Liaoning	47.39	18.88	11.56	402.47	27.80	24.75
Macao						
Ningxia					6.10	
Qinghai	83.66	67.55	39.77	99.36	57.91	
Shaanxi	211.26	52.19	8.64	579.53	103.62	3.07
Shandong			10.11			261.23
Shanghai						
Shanxi		12.69	27.19	0.43		68.11
Sichuan	367.12	208.23	71.89	254.07		41.33
Taiwan						
Tianjin						
Xinjiang	26.10	12.48	74.15	1.44		
Xizang	26.33	28.08	5.40			7.36
Yunnan	560.22	106.73	201.34	916.17	379.37	307.69
Zhejiang	46.43	36.89	9.49	35.41		46.80
National	3842.18	1981.33	1155.83	5301.52	2862.21	2888.56
Other countries	1459.34	880.88	1732.73			

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Table 2. Mercury content in ore concentrates produced in each province or from other countries.

Region	Zinc Geometric mean (gt^{-1})	Standard deviation (gt^{-1})	Number of mines	Lead Geometric mean (gt^{-1})	Standard deviation (gt^{-1})	Number of mines	Copper Geometric mean (gt^{-1})	Standard deviation (gt^{-1})	Number of mines
Anhui	4.1		1	14.66	64.85	2	0.34	1.06	4
Choingqing				114.91		1			
Fujian	0.54	1.77	11	12.63	26.37	4			
Gansu	499.91	511.8	9	10.77	3.54	3	2.86	14.03	4
Guangdong	72.16	144.36	3	43.75	50.15	3	0.05		1
Guangxi	9.34	48.31	9	10.13	55.59	12	0.62	1.1	3
Heilongjiang				25.67		1			
Henan	4.96	4.4	4	2.25	17.98	7			
Hubei				6.86		1	0.99	3.2	6
Hunan	4.72	21.86	26	1.31	2.07	11			
Inner Mongolia	2.16	9.21	6	62.21	27.89	4	1.84	0.24	2
Jiangsu	13.29	18.73	2	18.61	19.02	3	0.06		1
Jiangxi	1.47	3.15	10	19.51		1	4.66	16.5	7
Jilin				55.58	0.82	2			
Liaoning				61.04	29.92	6			
Qinghai				0.6	1.31	3	1.77		1
Shaanxi	240.77	701.15	12	45.14	45.83	3			
Shandong				4.92		1	1.5		1
Shanghai									
Shanxi				52.17		1	0.14	0.17	3
Sichuan	45.55	54.64	10	26.46	54.22	5	2.15	1.93	3
Xinjiang	16.86	54.62	3				2.02	19.57	7
Xizang	0.23		1	0.02		1			
Yunnan	10.98	30.98	6	21.54	2.28	3	13.68	41.42	12
Zhejiang	0.88	1.95	5	20.96	50.66	5			
National average	9.74	343.38	118	10.29	40.25	83	2.87	1.49	55
Other countries	9.04	59.80	10	3.16	2.60	3	0.88	2.85	9

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Table 3. Mercury content in ore concentrates consumed by each province in 2010.

| Province | Mercury content ($g\,t^{-1}$) | | | Province | Mercury content ($g\,t^{-1}$) | | |
	Zinc	Lead	Copper		Zinc	Lead	Copper
Anhui	–	5.13	13.03	Jiangxi	1.47	22.06	9.81
Beijing	4.10	–	–	Jilin	–	–	55.58
Chongqing	–	–	–	Liaoning	8.07	42.47	37.85
Fujian	–	–	–	Macao	–	–	–
Gansu	0.54	10.77	5.06	Ningxia	–	62.21	–
Guangdong	403.39	39.91	–	Qinghai	8.44	0.60	–
Guangxi	33.15	6.92	25.56	Shaanxi	73.61	45.26	45.14
Guizhou	10.43	–	–	Shandong	–	–	3.16
Hainan	9.74	–	–	Shanghai	–	–	–
Hebei	–	–	9.11	Shanxi	9.04	–	24.06
Heilongjiang	–	–	–	Sichuan	58.35	–	26.46
Henan	–	19.78	10.22	Taiwan	–	–	–
Hong Kong	16.06	–	–	Tianjin	–	–	–
Hubei	–	–	16.91	Xinjiang	16.86	–	–
Hunan	–	14.33	2.20	Xizang	–	–	10.29
Inner Mongolia	8.98	62.21	22.18	Yunnan	17.66	15.21	14.38
Jiangsu	12.09	–	–	Zhejiang	0.88	–	9.26
National	40.27	20.03	2.25				

Table 4. The proportion of metal production from smelters with different types of APCDs.

APCDs	Type of APCDs combination (/)	Zinc		Lead		Copper	
		Production (kt)	Percentage (%)	Production (kt)	Percentage (%)	Production (kt)	Percentage (%)
DC + FGS + ESD + DCDA	1	3841.05	76.31	1720.57	61.58	2721.28	93.15
DC + FGS + ESD + MRT + DCDA	2	508.04	10.09	0.00	0.00	0.00	0.00
DC + FGS + ESD + SCSA	3	69.52	1.38	108.35	3.88	81.40	2.79
DC + FGS	4	37.24	0.74	179.67	6.43	18.09	0.62
DC	5	172.07	3.42	37.52	1.34	2.44	0.08
FGS	6	1.68	0.03	3.16	0.11	0.00	0.00
None*	7	275.10	5.47	744.68	26.65	98.12	3.36

* Smelters without detailed APCDs' information are treated as no APCDs.

Table 5. 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]	99.9[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]	99.9[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.

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Table 6. Installation rate of a certain type of APCD combinations in each province.

Process (i)	Zinc-EP						Zinc-EZF			Zinc-RZSP		Lead-SMP				Lead-SPP				Copper-FFSP			
The type of APCDs (l) Province (j)	1	2	3	4	5	7	1	5	6	1	5	1	3	4	5	4	5	6	7	1	3	4	5
Anhui																0	100	0	0				
Beijing																							
Chongqing																							
Fujian	100	0	0	0	0	0																	
Gansu	95	0	0	0	0	5	100	0	0							0	0	0	100				
Guangdong	100	0	0	0	0	0																	
Guangxi	90	0	0	9	1	0						38	53	3	7	63	14	23	0	0	100	0	0
Guizhou	87	0	0	0	0	13	0	75	25	0	1												
Hainan																							
Hebei																				83	17	0	0
Heilongjiang																							
Henan	49	0	0	0	0	51						66	34	0	0	0	0	0	100				
Hong Kong																							
Hubei																				0	0	100	0
Hunan	55	45	0	0	0	0				100	0	89	0	0	11	0	0	0	100	100	0	0	0
Inner Mongolia	100	0	0	0	0	0						0	0	100	0	0	0	0	100	0	100	0	0
Jiangsu																							
Jiangxi	0	0	0	0	0	100																	
Jilin																				0	0	100	0
Liaoning	100	0	0	0	0	0				95	5	0	0	0	100	0	1	0	0	59	41	0	0
Macao																							
Ningxia												0	0	0	100								
Qinghai	37	0	63	0	0	0						0	100	0	0								
Shaanxi	97	0	1	0	2	0	0	100	0	100	0	100	0	0	0	0	0	0	100	0	0	100	0
Shandong																							
Shanghai																							
Shanxi	0	0	0	0	0	100														0	100	0	0
Sichuan	100	0	0	0	0	0				100	0									100	0	0	0
Taiwan																							
Tianjin																							
Xinjiang																				0	0	0	100
Xizang																							
Yunnan	87	0	1	0	12	0	79	21	0	0	100	0	0	100	0	0	0	0	100	100	0	0	0
Zhejiang	100	0	0	0	0	0														100	0	0	0

Installation proportion of certain type of APCDs, θ (%)[*]

[*] $0 \leq \theta \leq 1$, $\sum_{l} \theta_{l,ij} = 1$

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Table 7. Mercury removal efficiency of APCD.

Reference APCD	Zhang et al. (2012), η (%)						Wang et al. (2010), η (%)	Li et al. (2010), η (%)	This study Average η (%)	Standard deviation
	Smelter 1	Smelter 2	Smelter 3	Smelter 4	Smelter 5	Smelter 6				
DC	20.0	13.9	13.8	–	2.4	–	–	–	12.5	7.3
FGS	66.6	–	–	–	–	–	17.4	–	42.0	34.8
ESD	32.2	–	–	–	–	–	30.3	–	31.3	1.3
FGS + ESD	88.2	99.0	99.3	80.5	76.2	97.5			90.1	10.1
RT	–	–	–	–	–	–	87.5	91.4	89.5	2.8
DCDA	99.2	80.0	30.4	90.9		28.0	97.4	–	71.0	33.1
SCDA	–	–	–	–	52.3	–	–	–	52.3	–

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Table 8. Atmospheric mercury emission estimation from China's zinc, lead and copper smelters during 2000–2010.

Estimation year	Atmospheric mercury emission (t)				Reference
	Zinc	Lead	Copper	Total	
2000	161.4	48.0	12.7	222.1	Wu et al. (2006)
2000	44.23	17.99	5.40	67.63	This study
2001	173.0	54.3	13.7	241.0	Wu et al. (2006)
2002	178.5	57.8	14.8	251.1	Wu et al. (2006)
2002	80.7	–	–	–	Li et al. (2010)
2003	187.6	70.7	17.6	275.9	Wu et al. (2006)
2003	84.6	–	–	–	Li et al. (2010)
2003	73.08	20.88	6.11	100.08	This study
2004	97.1	–	–	–	Li et al. (2010)
2005	37.59	29.75	15.84	83.19	Hylander and Herbert (2008)
2005	97.4	–	–	–	Li et al. (2010)
2005	56.98	25.14	4.57	86.69	This study
2006	104.2	–	–	–	Li et al. (2010)
2006	107.7	–	–	–	Yin et al. (2012)
2007	–	–	–	203	Pirrone (2010)
2007	46.17	30.53	3.93	80.63	This study
2010	39.4	30.6	2.5	72.5	This study

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Table 9. Comparison of mercury emission factors for China's primary zinc, lead and copper smelters.

Metal	Smelting Process	Mercury emission factor ($g\,t^{-1}$)											
		A[a]	B[a]	C[a]	D[a]	E[a]	F[a]	G[a]	H[a]	I[a]	J[a]	K[a]	L[a]
Zinc	–[b]	8–45	25	20		86.6	7.5–8	16.61	7				7.82
	EP with MRT									5.7	0.5		0.59
	EP without MRT									31		0.57	9.75
	RZSP									34			6.16
	EZF												13.80
	ISP									122		2.98	6.02
	AZSP				79/155					75			45.75
Lead	–[b]	2–4	3	3		43.6	3	14.91	3				10.97
	RPSP											1.00	1.19
	SMP											0.49	10.16
	SPP												29.35
	ISP												6.07
Copper	–[b]			10		9.6	5–6	6.72	5				0.85
	FFSP											0.23	7.91
	RPSP											0.09	0.28
	IFSP												1.07
	EF/RF												14.96
	RLEP												0.38

[a] A: Nriagu et al. (1988); B: Pirrone et al. (1996); C: Pacyna et al. (2002); D: Feng et al. (2004); E: Streets et al. (2005); Wu et al. (2006); F: Pacyna et al. (2006); G: Hylanderand Herbert (2008); H: Pacyna et al. (2010); I: Li et al. (2010); J: Wang et al. (2010); K: Zhang et al. (2012); L: This study;
[b] Not specific value for each process.
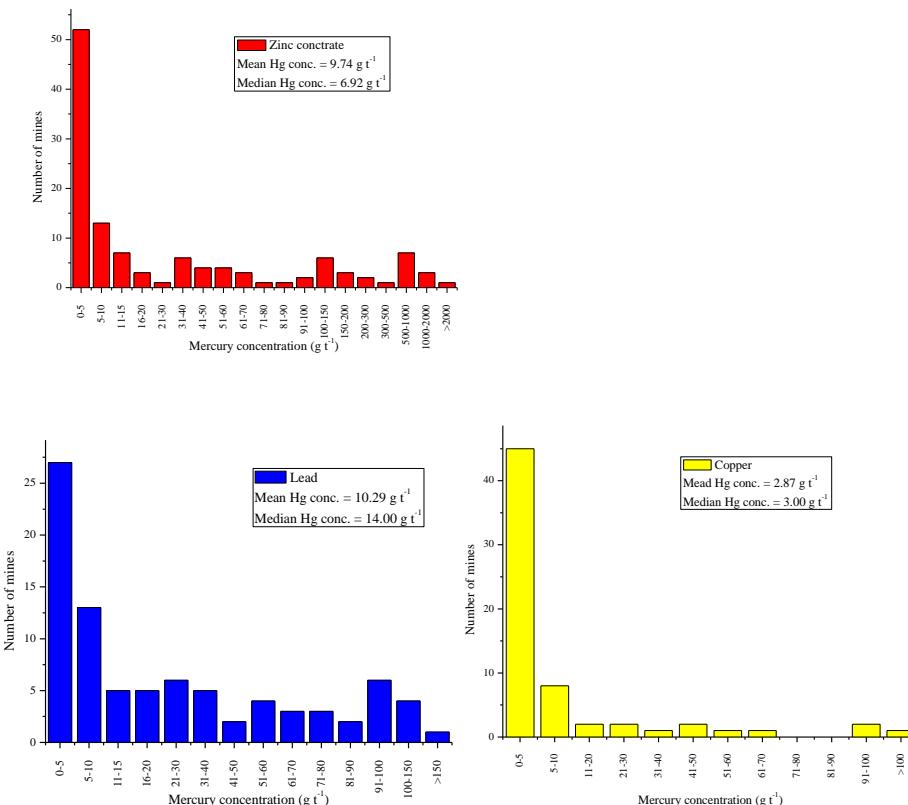
Fig. 1. Histograms showing number of Chinese mines in certain range of mercury content.
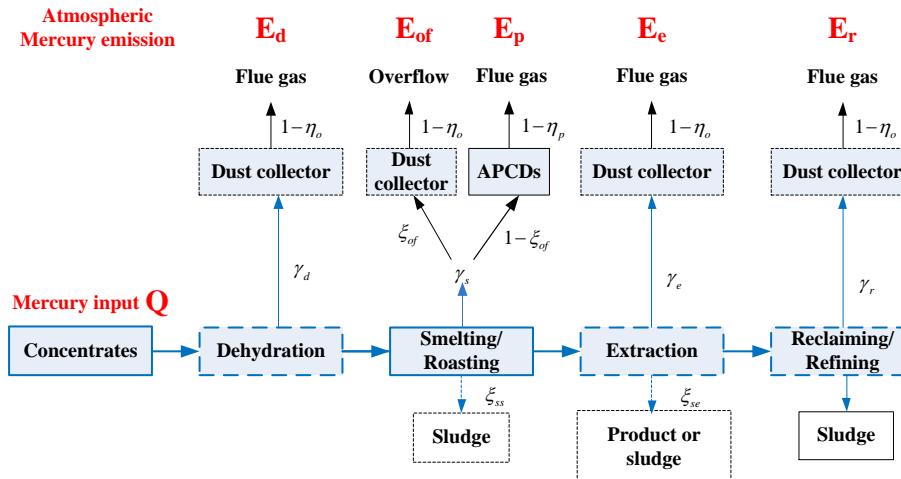
**Fig. 2.** Flow diagram for nonferrous metal smelters.
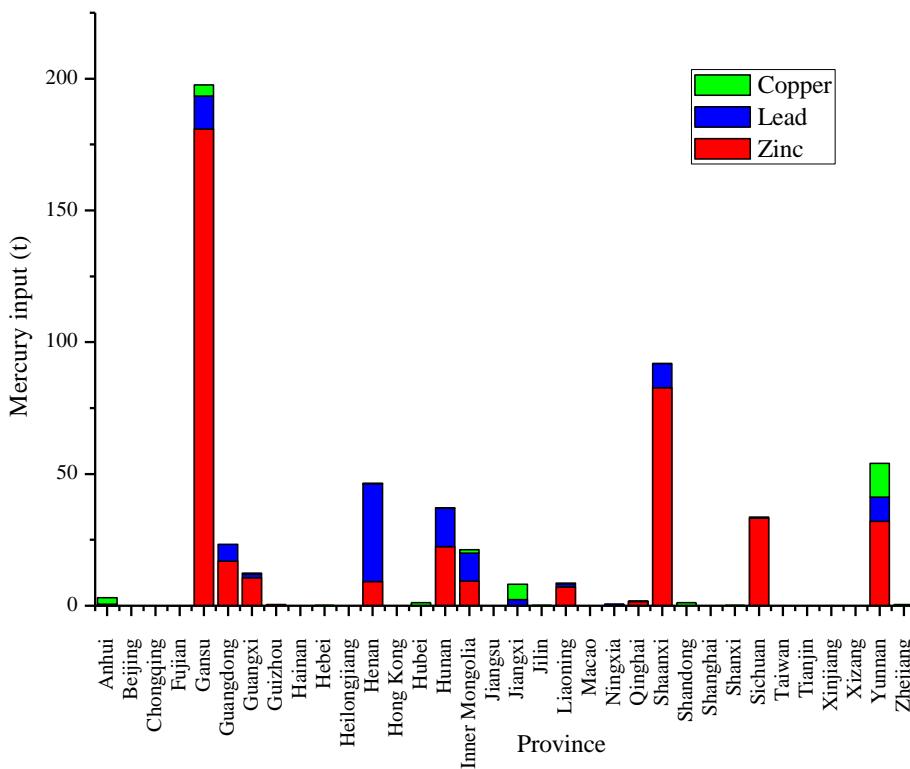
Fig. 3. Amount of mercury in the ore concentrates consumed by each province in 2010.
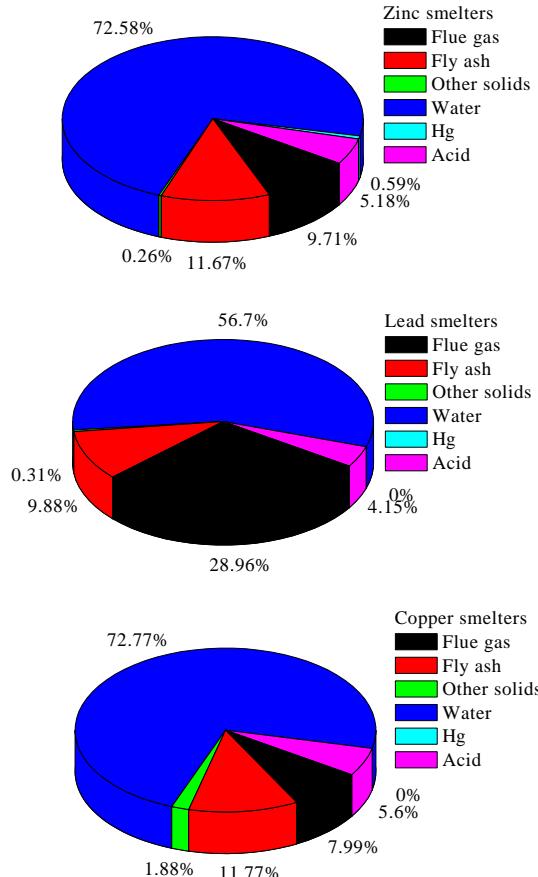
Fig. 4. Fate of mercury in China's zinc, lead and copper smelters.
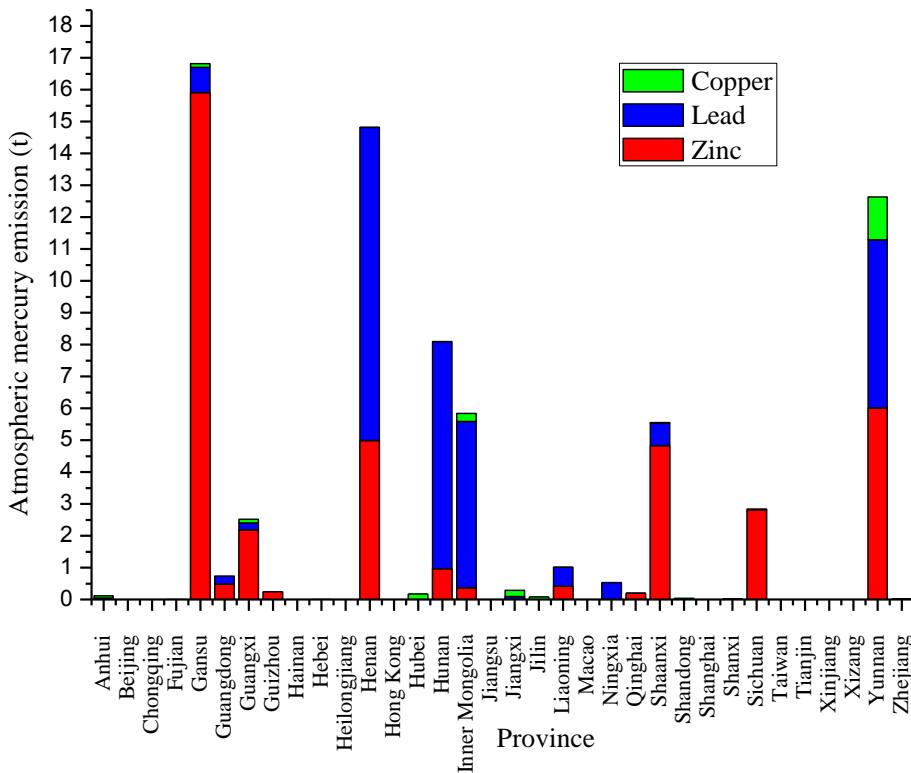
Fig. 5. Atmospheric mercury emission from zinc, lead and copper smelters by province, 2010.
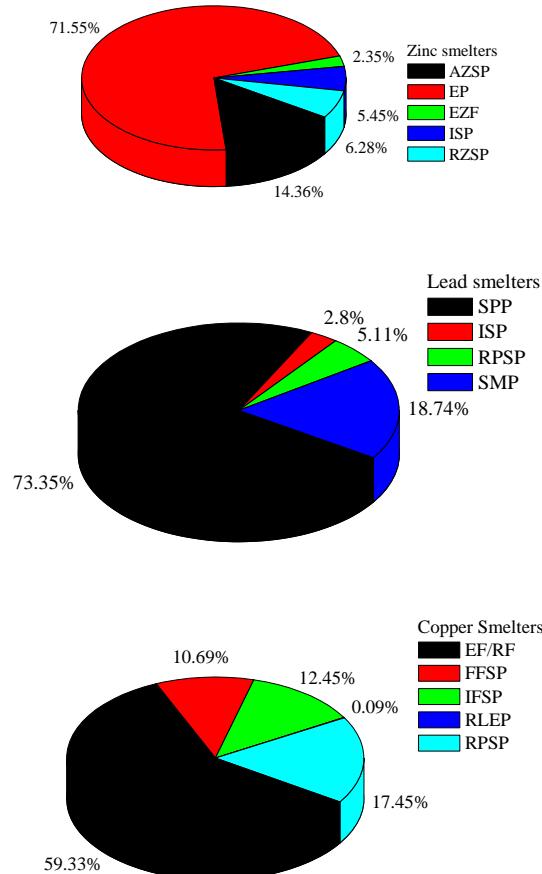
Fig. 6. Atmospheric mercury emission from zinc, lead and copper smelters by process, 2010.
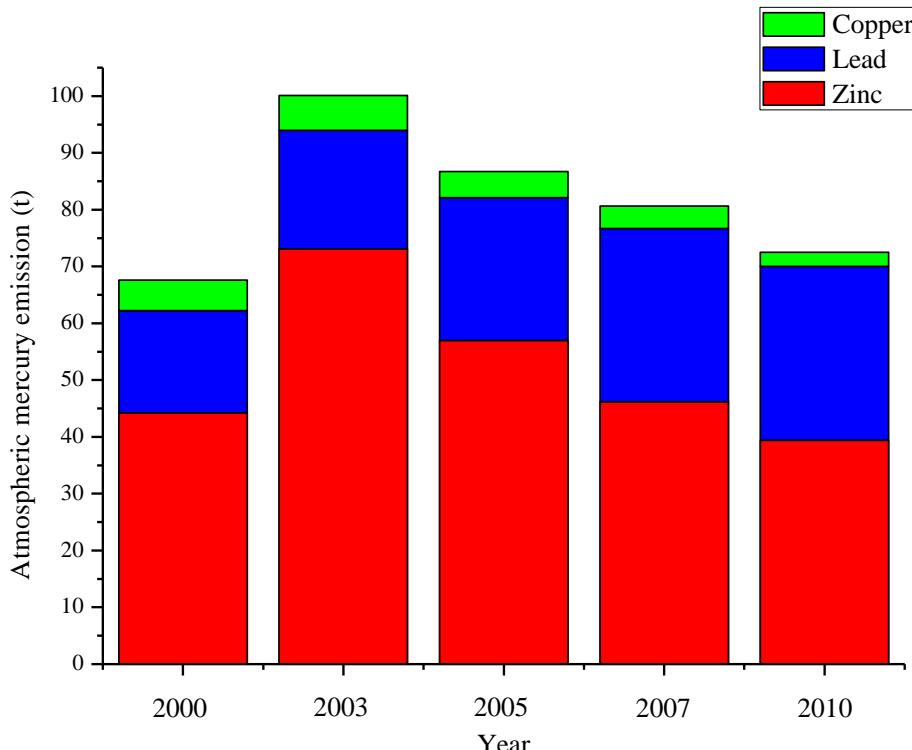
Fig. 7. Historical changes of atmospheric mercury emission from primary nonferrous metal smelters in China, 2000–2010.

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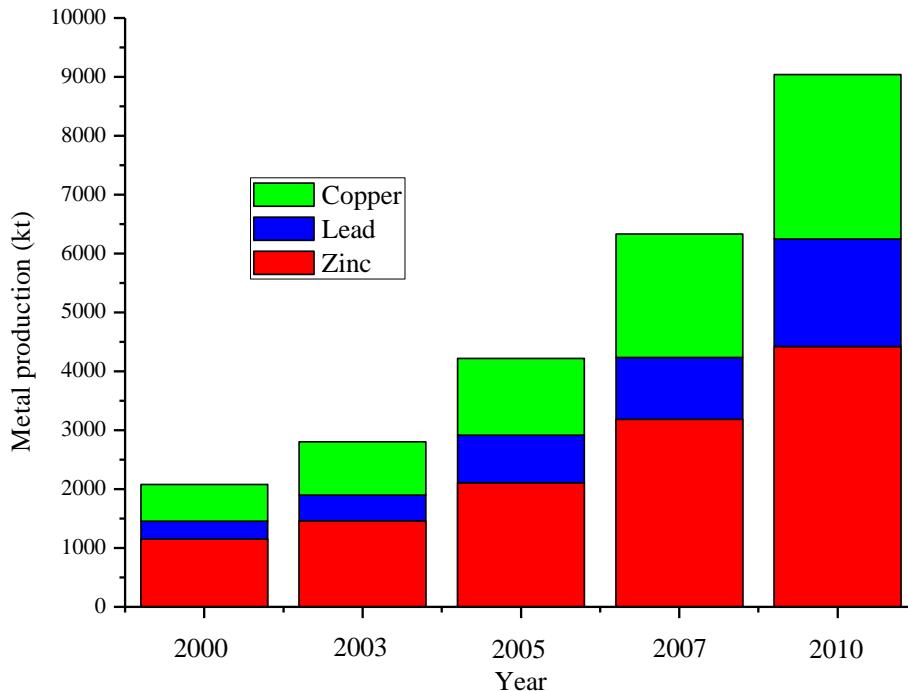
Fig. 8. Historical changes of metal production from primary nonferrous metal smelters in China, 2000–2010.
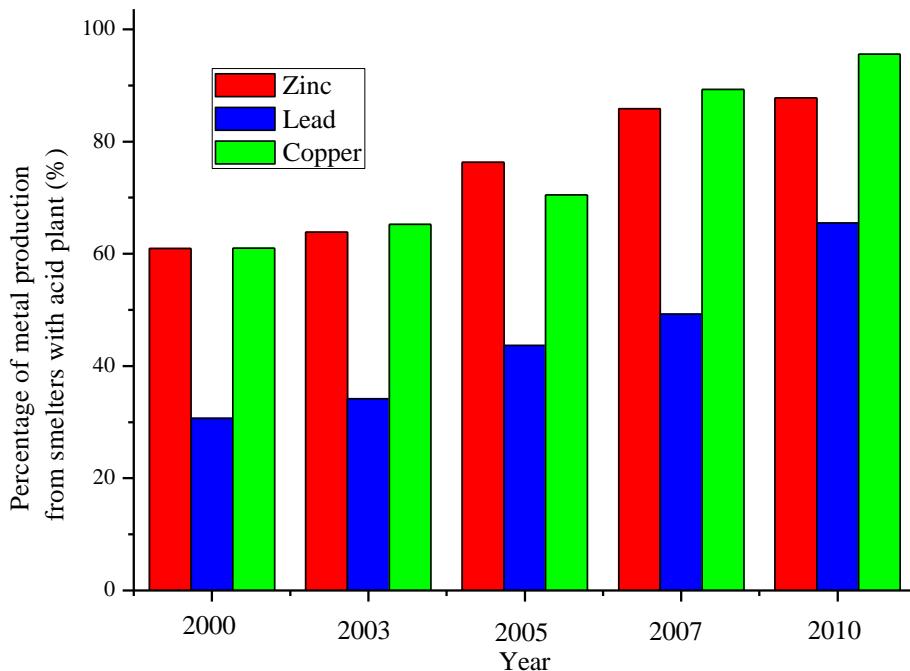
Fig. 9. Historical changes of the percentage of metal production from smelters with acid plants, 2000–2010.