Dear editor,

Thank you and the reviewer for your comments on our manuscript. We have revised our manuscript carefully based upon the reviewer's comments. We have attached a point-by-point list of our replies to the reviewer's comments. A native English speaker, Dr. Chuck Freed from US EPA, has helped us edited the manuscript to improve the language.

On behalf of our co-authors, I am resubmitting the revised manuscript to Atmospheric Physics and Chemistry. Please let me know if you have any questions.

Thank you very much for your considering our manuscript for publication. I am looking forward to hearing from you soon.

Sincerely yours,

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Response to the reviewers' comments

No.	Location	Comments and replies (Referee 1#)	
1	Section 2.1 Lines 18-22 (page 5)	C.	Please state how the samples were selected (i.e., sampling scheme) to demonstrate the representativeness of the samples.
		R.	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**. (**See supporting information of revised paper: Line 1-14, P2; Table S1**) 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. (**See supporting information of revised paper: Line 1-5, P4**)
2	Section 2.1 Lines 1-5 (page 6)	C.	1) Please specify the QAQC routines of the measurement. Has the instrument calibrated with internal standards or external references (such as those used by NIST)? 2) Also, based on Figure 1, the data is very much skewed and therefore using geometric mean is not representative of the Hg content. 3) The Hg content should be broken down into percentiles, which can be used for estimating data uncertainty.

		R.	1) 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). (**See supporting information of revised paper: Line 11-13, P4**). 2) Generally, both arithmetic mean and geometric mean can be used to reflect mercury content in ore concentrates. However, arithmetic mean is more vulnerable to extreme numbers, and therefore may not reflect the average of the specific phenomenon. The geometric mean is used for calculating dynamic average. It is less affected by the extreme values than the arithmetic mean, and it can reflect general level. We agree with the reviewer that mercury concentration data is truly skewed, but they meet the skewed distribution. Most concentrates have low mercury content, typically less than 10 g mercury t^{-1} copper concentrates, or 20 g mercury t^{-1} zinc / lead concentrates. However, the maximum values can reach 2534.06, 193.00 and 106.54 g t^{-1} for zinc, lead and copper concentrates, respectively. Thus, we think geometric mean is representative of the Hg content. (**See revised paper:Line6-9, P6; Table 1; Fig. S1.**) 3) We have broken down Hg content into percentiles for inventory uncertainty estimation in the revised manuscript. **(See supporting information of revised paper:Fig.S1).**
3	Section 2.1 Lines 16-17 (page 6).	C.	The Hg content in ore concentrates (Table 2) is not relevant for emission inventory estimate; it is the consumption that counts. Suggest deletion of this table for a better focus of the manuscript. Similarly, the data regarding ore supply from each province in Table 1 dilutes the focus (e.g., emission inventory estimate) in the manuscript.

		R.	The Hg content in ore concentrates is the basis for estimating Hg contents in the ore consumed. If we delete the table, it would be not possible to explain how the Hg content in ore consumed was calculated. To make the manuscript more focus, we have simplified Table2. In addition, the data on ore supplying each province were moved to the supporting information with concentrates trade matrixes. **(See revised paper: Table 1; supporting information: Line 16-24, Table S2, S3, S4)**
4	Section 2.1 Line 21 (page 6).	C.	1) How the weighted average was calculated? 2) Again, the Hg content in the ore consumption should be broken down into percentiles for uncertainty assessment.
		R.	1) The national weighted average was calculated according to E4. (**See revised paper: E4**) $$[Hg]=\frac{\sum_i\sum_j [Hg]_{com,ij} C_{com,ij}}{\sum_i\sum_j C_{com,ij}} \quad \text{(E4)}$$ where, $[Hg]_{com..,ij}$ and $C_{com..,ij}$ are mercury content and amount of the ore concentrates consumed by *j* technology in *i* province. 2) Hg content in the ore consumption was broken down into percentiles. (**See Fig.1 in the revised manuscript**)
5	Section 2.2 Lines 21-26 (page 7).	C.	1) Discussion regarding how the coefficients in Eq. E3-E7 were estimated should be provided. The data presented in Tables 5 and 6, although comprehensive, are not directly useful for emission inventory estimate efforts elsewhere. I recommended that the two tables be removed while keeping the references that detail the procedures of obtaining the values in the text.
		R.	The two tables were moved to the supporting information according to the comment of Referee 2#.The coefficients were estimated according to the following description. ➢ θ is the application percentage of a certain type of APCD

			combinations; information about θ is obtained from our investigation of 244 smelters and China's Nonferrous Metal Industry Association (**Table S3**). (**See revised paper: Line 20-22, P7; supporting information Table S5**) ➢ γ is the mercury release rate; the value of γ was based on our field experiments in Chinese smelters(Li et al., 2007; Wang et al., 2010; Zhang et al., 2012). Mercury release rates in various smelting processes, γ_s ,are at the range of 97.7% - 99.4%.γ_d , γ_e, γ_r is the mercury release rates in hydration, extraction and refining/reclaiming process, respectively. The value of these three parameters is shown in **Table S6**. (**See revised paper: Line 23-25, P7; Line 1, Line 16-17, P8; Supporting information Table S6**) ➢ η is the mercury removal efficiency of APCD, which was based on field experiment in Chinese smelters (Li et al., 2007; Wang et al., 2010; Zhang et al., 2012). The value of η is shown in **Table 3**. (**See revised paper: Line 7-9, P8; Table 3**) ➢ ξ is the distribution coefficient (**Table S6**). ξ_{of} refers to the proportion of gaseous mercury emitted into atmosphere as overflow flue gas. The value of distribution coefficient was calculated from the mercury mass balance of field experiment result (Li et al., 2007; Wang et al., 2010; Zhang et al., 2012). Mercury distribution rate for dehydration is 0.1% - 1.0% (**Table S6**). ξ_{ss} and ξ_{se} here refer to the proportion of mercury entering into the solid waste in the smelting and extraction sector, respectively. The value of ξ_{ss} and ξ_{se} is 0.02%-20.6%, and 2.4%-14.4%, respectively (**Table S6**). (**See revised paper: Line 4-6, P8; Line 13-15, P8**)

			➢ η_o is the mercury removal efficiency for other flue gases (**Table S6**). 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, flue gas desulfurization (FGD) devices are installed. No APCDs are installed for the flue gas from the out-of-date processes such as AZSP, RZSP and EF/RF. Therefore mercury removal efficiencies for other flue gas depend on the APCD applied. The mercury removal efficiencies of dust collector and FGD were 12.5% and 34.7%, respectively. (**Table S6**). (**See revised paper: Line 17-24, P8; supporting information, Table S6**) ➢ α is metal concentration and the values for zinc, lead and copper concentrates were 50.5%, 62.85 and 21.7%, respectively (**Table S6**) (CNMIA, 2011).(**See revised paper: Line 6-7, P9; supporting information, Table S6**) ➢ φ is metal recovery rate of smelting process. For most zinc smelting process, the metal recovery rate was 95.5% while for EP it was 94%. For the lead and copper smelting processes, the metal recovery rate was 96.8% and 97.8%, respectively (**Table S6**). (**See revised paper: Line 7-10, P9; supporting information, Table S6**)
6	Section 3 (Results and Discussion).	C.	The objective of this work is to provide reliable emission inventory updates so that the uncertainty in earlier data can be reduced. To this end, I was somewhat surprised by the fact that the authors missed two important aspects in such an evaluation: 1) there is no assessment of uncertainty of the data and the estimated emission values, 2) there is no assessment of possible emission speciation in this

			work. These two components should be supplemented in this section. 3) The text in Section 3.1 (including Fig. 3) seems to be off topic because it is not directly related to emission inventory estimate. 4) Finally, how the updated emissions from the non-ferrous smelters would influence the understanding of total Hg emissions in China should be discussed.
		R.	1) We agree with the reviewer that the uncertainty analysis is necessary and we have added this part in the revised manuscript. The uncertainty of this inventory was estimated by combining the coefficients of variation (CV, or the standard deviation divided by the mean) of the contributing factors according to the detailed methodology for uncertainty analysis described in Streets et al. (2003a). The relative 95% confidence intervals for emissions are calculated as 1.96 times CV. Thus, atmospheric mercury emission from zinc, lead and copper smelters was 39.4±31.5, 30.6±29.1, 2.5±1.1 t in 95% relative confidence and the uncertainty is ±80%, ±95% and ±45%, respectively. In previous studies, the uncertainty for these three sources reached 100%, 200% and 100%, respectively. The improvement in this study was contributed by better knowledge on the mercury content of ore concentrates and mercury removal efficiency of APCDs. However, more field experiments are still important to better understand the mercury fate in smelters. Besides, high uncertainties exist for the emissions from small-scale smelters. (**See revised paper: Line26-27, P11; Line1-10, P12**). 2)The mercury speciation profile was assumed to be 80% Hg^0, 15% Hg^{2+} and 5% Hg^p for nonferrous metal smelters in previous

			estimate(Pacyna et al., 2002). The field experiments in Chinese nonferrous smelters provided a different speciation profile (Wang et al., 2010; Zhang et al., 2012). In this study, the median of the results from field experiments was used to estimate mercury speciation emissions. For zinc smelters, the percentage of Hg^{2+}, Hg^{0} and Hg^{p} in flue gas emitted to the atmosphere was65%, 30% and 5%, respectively. The Hg^{2+}, Hg^{0} and Hg^{p} emissions from zinc smelters were 25.6, 11.8 and 1.97 t, respectively. Using the same speciation profile, the Hg^{2+}, Hg^{0} and Hg^{p} emissions from lead smelters were 11.64, 17.74 and 1.53 t, respectively, and those for copper smelters were1.19, 1.16 and 0.12 t, respectively.(**See revised paper: Line 28-29, P10; Line1-8, P11**) 3)We followed the reviewer's comment and deleted the discussion about mercury fate in the revised manuscript. 4) Feng et al. (2009) summarized previous studies and pointed out that the average emission factors were 5.4-155 g t^{-1} Zn, 43.6 g t^{-1}Pb, 9.6 g t^{-1} Cu, respectively. If these three emission factors were adopted for emission estimation as that in Pirrone et al. (2010), the atmospheric mercury emission from nonferrous metal smelters in 2010 will reach 558 t. This indicated that atmospheric mercury emissions in China in 2010 will be overestimated by 400 t. **(See revised paper: Line13-18, P13**)
7	Figures.	C.	There are excessive figures in the manuscript. Most of the figures are simple bar charts that can be presented in a much more succinct fashion. 1) Figure 1's X Axis is not shown at correct scale because the data ranges are not consistent in the bins. The three subplots can be combined into three box-and-whisker plots in one single figure.

			2) Figures 3 and 5have many provinces that have no data and the scale in the Y Axis makes it difficult to read the input/emission quantity. Suggest the provinces without data be removed and change the Y Axis to log scale. 3) Figure 7 and 8 can be combined into one using a secondary Y Axis. 4) Figure 9 (and the associated discussion on page 12) is a distraction from the points the manuscript attempts to address - suggest deletion.
		R.	1) The three subplots were combined into three box-and-whisker plots in one single figure. (**See revised paper: Fig. 1.**) 2) Figure 3 was deleted and Figure 5 was revised. Figure 5 is better presented as 3 maps for zinc, lead, and copper smelting according to the advice of Referee 2#. (**See revised paper: Fig. 3.**) 3) Figure 7 and 8 was combined into one figure using a secondary Y Axis. (**See revised paper: Fig. 5.**) 4) Figure 9 (and the associated discussion on page 12) was deleted.
8		C	Although the English writing does not significantly impair the technical delivery, there are many places in the manuscript where the text is redundant, lacks clarity or has grammatical errors. A thorough editorial revision should be made after the technical revision.
		R	The revised manuscript was carefully edited by a native English speaker to improve the English.

No.	**Location**	**Comments and replies (Referee 2#)**

1	P. 18208 l.24 -18209 l. 3	C.	Provide context for this emission of 203 t – what % of the total anthropogenic emissions is this?
		R.	Atmospheric mercury emission from Chinese nonferrous metal smelters was estimated to be 9% of the total global anthropogenic emission. (**See revised paper: Line 8-10, P3**)
2	P. 18209 l.12	C.	What is "Brook Hunt"? Define or omit.
		R.	Brook Hunt is a corporation researching products and providing consulting services to support business analysis and decision making in the metals and energy industries.). (**See revised paper: Line 19, P3**)
3	P. 18209 l.20 – 18210 l.2	C.	It is not clear how this paragraph presents a reason for high uncertainty in past budgets (previously 99% was assumed, measured values are99.2-99.8%?). It may just be an issue of unclear language that needs to be explained better.
		R.	This paragraph was revised to clearly explain the reason of high uncertainty: Secondly, in most previous studies, an average emission factor was used to estimate emissions, which did not consider the removal effect of APCDs. Hylander and Herbert (2008) pointed out the synergic effect of APCDs but the mercury removal efficiencies in their paper were estimated on the basis of sulfur abatement technology. About 95% of gaseous mercury was removed from flue gas in zinc/lead smelters with sulfuric acid plants and no mercury removal tower (Hylander and Herbert, 2008). However, such kind of assumption neglected the different removal efficiencies of various types of sulfuric acid plant. Field measurements conducted in China's zinc, lead and copper smelters indicated the total mercury removal efficiency for

			zinc/lead smelters with double-contact sulfuric acid plants and no mercury removal tower is over 99% while mercury removal efficiency is only 89% for Zn/Pb smelters with single-contact sulfuric acid plants (Li et al., 2010; Wang et al., 2010; Zhang et al., 2012). In this study, these updated removal efficiencies of APCDs will be used for emission estimation. (**See revised paper: Line 27-28, P3; Line 1-11, P4**)
4	P. 18210 Section 2 (Methodology)	C.	There is no mention of AZSP in this list of smelting processes, though it is later mentioned in the text and Table 5.
		R.	We are sorry for the omission. The sentence has been revised as follows. Zinc smelting processes include oxygen pressure leaching process (OPLP), electrolytic process (EP), imperial smelting process (ISP), retort zinc smelting process (RZSP), electric zinc furnace (EZF) or artisanal zinc smelting process (AZSP). (**See revised paper: Line 2-5, P5**)
5	P. 18212 l. 10	C.	Refer reader to Table 2 here.
		R.	The sentence has been revised as follows. Most concentrates have low mercury content, typically less than 10 g mercury t^{-1} copper concentrates, or 20 g mercury t^{-1} zinc / lead concentrates (see **Table 1, Fig. S1)**. (**See revised paper: Line 7-9, P6; supporting information: Fig. S1**)
6	P. 18212 ll.24-26	C.	Provide reference for the "trade and transport among provinces" Information
		R.	The transport matrix of ore concentrates was developed according to the import and export of concentrates for each province in China in 2010. The concentrates supply of each province are taken from the Yearbook of Nonferrous Metals Industry of China (CNMIA,2011). The transport between provinces ($C_{su,k\rightarrow ij}$) was

			based on the trade between the ore plants and the 244 nonferrous metal smelters in our survey. For smelters without trade information (mostly in small smelters with discontinued production), we assumed that local concentrates were used. Based on the above information, linear equations were established and solved. The zinc, lead and copper concentrates transport matrixes are given in **Table S2, S3,S4**. (**See supporting information of revised paper: Line 17-24, P4; Table S2, S3, S4**)
7	p. 18213 ll. 20-22	C.	Change to, "The CAT may be a double conversion……tower or a single conversion……tower."
		R.	The sentence has been revised as follows. The CAT may be a double conversion double absorption (DCDA) tower or a single conversion single absorption tower (SCSA). (**See revised paper: Line 8-9, P7**)
8	18213 ll. 22-24:	C	Please provide more details about how the information was obtained or reference appropriately.
		R	In this study, we grouped the APCDs used in smelters into 7 types (**Table 2**). The information about the type of APCD combinations used in most smelters is based on our investigation in 244 nonferrous metal smelters. For those smelters without APCD information but with acid plant, we assumed that the type 1 of APCD combinations (DC+FGS+ESD+DCDA) was adapted. For smelters without any information about acid production or APCDs, type 7 (None APCDs) were adapted. (**See revised paper: Line 9-15, P7**)
9	18215 l. 13	C.	What is the fate of the fly ash? Is it all collected or is a fraction released as atmospheric particulate (if so, do you have an estimate of the emission of particulate mercury to the atmosphere)?
		R.	A fraction of the fly ash was released as atmospheric particulate,

			but it took up less than 1% according to the removal efficiency of dust collector. We have estimated the emission of particulate mercury to the atmosphere and the fly ash in this study refers to the part collected. The discussion of the fate of fly ash was deleted since it distracted the focus of our study according to the advices from referee1#.
10	18214 ll. 15-17	C.	Change to, "Some mercury is washed: : :while some flows into the sulphuric acid…….A limited fraction is recovered……There is still a trace amount of mercury remaining……."
		R.	Such description is better but this part has been deleted according to the reviewer's comments.
11	18216 l. 4	C.	Is this "consumption of CHINESE ore concentrates in 2010"?
		R.	This is the consumption of all ore concentrates input into Chinese nonferrous metal smelters. We have deleted the discussion of this part in the revised manuscript.
12	18218-18220 (Section 3.4)	C.	As mentioned by Referee #1, this section can be condensed a great deal. I suggest changing the last line on p. 18218 to, "The increased application of acid plants, particularly after 2003, was the main reason for atmospheric mercury abatement in the past decade." The remainder of this section can be removed along with Fig. 9, or condensed to 2-3 sentences summarizing regulation changes and perhaps including only the 2000 and 2010 numbers for percent of smelters with acid plants
		R.	This section have been removed along with Fig. 9.
13	18220-21 (second paragraph of 3.5):	C.	Clarify that the similar results of Hylander and Herbert are coincidental due to lower estimated ore mercury concentrations but also lower application rates for acid plants. This was not obvious as written. Remove final sentence.
		R.	We agree that the similar results of Hylander and Herbert are

			coincidental due to lower estimated ore mercury concentrations but also lower application percentages for acid plants. We have corrected this part as follows. However, such similar results are coincidental due to their lower estimated ore mercury concentrations but also lower application percentages for acid plants. The weighted national average of mercury content in zinc, lead and copper concentrates consumed by smelters reached 47.02, 16.81 and 2.82 g t^{-1}, respectively. However, global mercury concentration of 10, 9 and 3.5 g t^{-1} for zinc, lead and copper concentrates was used in the former study. Thus, if we assumed concentrate consumption was the same in these two studies, the mercury input into Chinese nonferrous metal smelters was estimated to be higher than that given by Hylander and Herbert (2008). However, the application percentage of acid plants in 2005 was about 76.3%, 43.7% and 70.5% for zinc, lead and copper smelters, which was also higher than their estimation. According to E4, atmospheric mercury emissions from nonferrous metal smelters would increase if the mercury input increase and the application percentage of acid plants decrease. This indicates that the lower estimation of mercury input in Hylander and Herbert (2008) was offset by their lower estimation of application percentage of acid plants. (**See revised paper: Line 21-28, P13; Line 1-6, P14**)
14	18221 l.25-27	C.	what is the “wide range”? Can you include uncertainties on the 91%,71% and 92% listed in the previous paragraph?
		R.	The average mercury removal efficiency of air pollution control devices in zinc, lead and copper smelters was 90.5±52.5%, 71.2±63.7% and 91.8±40.7%, respectively. (**See revised paper: Line 21-23, P11**)

15	TABLES AND FIGURES:	C.	I recommend that Tables 1 and 6 and Figures 3, 8, and 9 be moved to supplementary material, and possibly Table 7 as well. Also, I agree with Referee#1 that you should remove provinces with no data (you can mention in a footnote that there were no data in those if you like).
		R.	The table and figures were moved to supporting information. Provinces with no data were removed. (**See supporting information of revised paper: Table S1; Table S3; See revised paper: Fig.3**)
16	TABLES AND FIGURES:	C.	Table 3: Are these geometric means as in Table 2? Can you provide some measure of uncertainty (e.g. standard deviation based on propagation of the largest errors)?
		R	Yes, these are geometric means. Standard deviation was given as the indicator of uncertainty. (**See revised paper: Table 3**)
17	TABLES AND FIGURES:	C.	Table 5: Include entries in all rows in column 1 (Metal); it is not clear which processes are for which metal otherwise. In column 2, is "RE" supposed to be "RLEP"?
		R.	Table 5 has been corrected. RE is an old copper smelting technology forbidden by Chinese government, which is different from RLEP. (**See revised paper: Table 5**)
18	TABLES AND FIGURES:	C.	Table 7：Does "This study" simply refer to an average of the literature values shown in this table? This needs to be discussed in the text (or in the supplement if you move this table).
		R.	Yes, the geometric mean of previous field measurement results was adapted in this study. (**See revised paper: Table 5**)
19	TABLES AND FIGURES:	C	Figure 5 would be better presented as a map (or 3 maps for zinc, lead, and copper smelting), colour-coding each province by mercury emissions. This would help non-Chinese readers quickly understand the regions of high mercury release.

		R	We have presented the results as 3 maps for zinc, lead, and copper smelting. (**See revised paper: Fig. 3**)