Main revisions and response to reviewers' comments

Manuscript No.: acp-2014-751

Title: Evaluating the effects of China's pollution control on inter-annual trends and uncertainties of atmospheric mercury emissions

Authors: Yu Zhao*, Hui Zhong, Jie Zhang, Chris P. Nielsen

We thank very much for the valuable comments from all the reviewers, which help us improve the quality of our manuscript. Following is our point-by-point responses to the comments and corresponding revisions.

Reviewer #1

1. *This study is of great value to the scientific field of mercury emissions inventories. It uses a detailed, novel approach, to better constrain the anthropogenic emissions of mercury from China; one of the largest emitters of mercury from a global perspective. Hence, an improved emissions inventory is therefore very important. The work is very thorough and covers the most important industrial sectors. The focus on uncertainties, temporal and spatial variability (due to pollution controls) makes the updated inventory more reliable and applicable. However, there are a few areas that require some additional attention from the authors to make this work ready for publication. These are detailed below.*

Response and revisions:

We thank the reviewer for his/her comment and positive remark on the importance of the paper. The responses to all the comments from the reviewer and corresponding revisions are made and described as below.

2. *In section 2.3 "Uncertainty analysis", line 208—210, it is described that*

"Generally, normal distributions, are assumed…" However, some industrial sectors do not have normally distributed data; and it is wondered if the data is not normal, how is the data then treated? Please describe in the text.

Response and revisions:

We thank the reviewer for his/her comment. We have stressed that the normal distributions are assumed for the activity level data, i.e., fuel consumption and industrial and agricultural production, for all the sectors **(lines 199-204, page 10 of the revised manuscript)**. There is no exception.

We believe that what the reviewer means is that some emission factor data for industrial sectors do not have normal distribution, and we've described the details in Section 3 by source.

3. *Section 3.1 "Other industrial sources": why is the data for this sector assumed to be "log normally distributed", whereas other sectors are normally distributed? The authors may want to explain this in the ms.*

Response and revisions:

We thank the reviewer for his/her comment, and we believe this question is partly related with Question 2.

For emission factors (not for activity levels, which are discussed in Q2), normal distributions are applied for given parameters through data fitting, as shown in Table 1 and Figure S1 in the Supplement. That means there are sufficient data points supporting the statistical analysis. However, there are very limited data available for some emission factors or parameters with emission factor estimates for industrial sources, and large uncertainty or coefficient of variation (CV) may exist. In this case, lognormal distribution is believed a better choice for the probability density. In particular, the lognormal distribution is preferred for variables which must be positive and for which the CV is greater than 30%, and it is commonly used in previous

studies (Bond et al., 2004). Therefore, we follow those studies and assume lognormal distribution for emission factors/parameters for certain industrial sources. The CV is conservatively set at 100%, to reflect the big uncertainty. We have added the discussion and relative references in **lines 385-387, page 18 of the revised manuscript**.

Reference:

Bond, T. C., Streets, D. G., Yarber, K. F., Nelson, S. M., Woo, J. H., and Klimont, Z.: A technology-based global inventory of black and organic carbon emissions from combustion, J. Geophys. Res., 109, D14203, doi: 10.1029/2003jd003697, 2004.

4. *Section 3.1 "Non-ferrous metal smelting": why are the 10th and 90th percentiles used? The authors do not provide a rational. Thus, one can ask "why not use 2-standard deviations instead?" The authors may want to explain this in the ms.*

Response and revisions:

We thank the reviewer for his/her comment. We should acknowledge that usage of 10^{th} and 90^{th} percentiles is a subjective (and also a conservative) decision, attributed mainly to the data limit. The current assumption (uniform distribution, with the lowest and highest measurement values taken as the 10th and 90th percentile) provides a distribution with larger variation than the commonly used normal distribution with 2-standard deviations, thus bigger uncertainty of the emission factors is indicated, due to insufficient data from field tests. We have stated this in **lines 333-337, page 16 of the revised manuscript**.

5. *Section 3.2, line 447 states that the increasing Hg emissions from China (679t in 2005, to 750t in 2012, with a peak of 771t in 2011) can help explain the observed decrease in atmospheric Hg concentrations worldwide. This is of course an incorrect*

conclusion because both are 180 degrees counter each other. China's mercury emissions only changed relative to energy usage, not in absolute terms; the emissions grew less than China's economic growth, but increased nonetheless. Hence, the authors' results do not support the observed global trend, quite the contrary. The reviewer suggests the authors delete this section to avoid confusion. A discussion of this discrepancy is outside the scope of this paper.

Response and revisions:

We thank the reviewer and agree with his/her important comment. From our estimates, China's Hg emissions kept increasing from 2005 to 2011, but the growth rate is much smaller than that of energy consumption for the same period, and also much smaller than that of emissions from 2000 to 2003, compared with previous study (Wu et al., 2003). Therefore, we have followed the reviewer's suggestion and deleted the phrase "and may help to explain decreased observed Hg concentrations worldwide" **(line 441, page 20 of the revised manuscript)**.

6. *Recommended edits.*

Response and revisions:

We thank the reviewer for his/her very careful comments on language and editing. We have taken most of the reviewer's suggestion and have improved the language and accordingly in the revised manuscript. Below are the two exceptions:

(1) "Line 514-516: Reviewer wonders if it is maybe better to apply these abbreviations for the first time in each respective section of chapter 3!?"

Actually we have defined all of those abbreviations when they appear for the first time in Section 2.1 **(lines 114-121, page 6 of the revised manuscript)**. We follow the reviewer's suggestion and use only the abbreviations after that.

(2) "Please explain what "triangular" and "Weibull" data distributions are, as most

likely many readers will not have knowledge of these type of data types (reviewer included) ”

Since the types of probability distributions appeared in Table 1 for the first time, we have described them **in the footnote of Table 1 in the revised manuscript**.

Reviewer #2

1. *Typo on the word "grown" on line 493*

Response and revisions:

We thank reviewer for his/her reminding and the error has been corrected.

2. *Conclusion should mention that the emissions estimated in this work are generally larger than those of global inventories, and summarize the reasons for this discrepancy.*

Response and revisions:

We thank the reviewer for his/her comment. We have revised the conclusion section, adding the texts: “The values are generally larger than those of global inventories, as higher activity levels (energy consumption and industrial production) and lower penetrations of emission control devices for certain industrial sources are applied in this study” **(lines 725-728, page 33 of the revised manuscript)**.

3. *The phrase in the abstract, lines 21-22, "based on available field measurements" is vague and should be reworded.*

Response and revisions:

We thank the reviewer for his/her comment, and have revised the text as “based on available data from domestic field measurements”, in **lines 21-22, page 2 of the**

revised manuscript.

4. *In the abstract, it should be explained how the emissions from the 4 Category 3 sources have come down by 50%, yet the total emissions for China have increased from 679 to 750 t Hg.*

Response and revisions:

We thank the reviewer for his/her comment. A sentence has been added to the abstract: "Emissions from other anthropogenic sources are estimated to increase by 22% during the period" **(lines 32-33, page 2 of the revised manuscript)** .

5. *Based on Figure 5, how do the authors find that there has been a 50% reduction in Category 3 sources?*

Response and revisions:

We appreciate the reviewer's important comment and acknowledge the confusing texts. A 50% reduction means that if no progress of emission controls had been made from 2005 to 2012, the Hg emissions for category 3 sources in 2012 would be almost twice of current estimate, exceeding 600 t. This is described in **lines 534-538, page 25 of the revised manuscript**, and illustrated in Figure S3 in the Supplement. Comparing the best guess of emissions for 2005 and 2012, however, the emissions for category 3 sources are estimated to decline by 3%. To avoid the confusion, we have corrected the values both in abstract **(lines 29-32, page 2 of the revised manuscript)** and conclusion section **(lines 729-732, Page 33 of the revised manuscript)**.

Reviewer #3

1. *I think the authors have done a good job responding to my suggested revisions. One issue that I think could be more specifically described is the speciation of*

emissions -- I was surprised specifically (line 454) that speciation hasn't changed, given the preferential capture of different forms by different APCT. A comment/explanation would be useful on that point.

Response and revisions:

We thank the reviewer for his/her important comment. Although the speciation of Hg emissions of anthropogenic origin as a whole has not changed significantly, clear changes in Hg speciation are found for different sectors. For example, the fraction of Hg^0 to total Hg for power generation is estimated to increase from 59% to 75% during 2005-2012, due to increased use of FGD systems; while that for cement and iron & steel production decrease from 44% to 33% and 43% to 30%, respectively, due to increased use of fabric filters. The varied speciation of emissions from different sectors compensates each other, leading to relatively stable speciation of total anthropogenic Hg emissions. We've added this explanation in **lines 447-453, page 21 of the revised manuscript**.

2. *Also, the authors could be a little more careful in the introduction -- I think they are overstating a bit the difference between their estimates and previous estimates, which are associated with a high degree of variation. Other than these minor concerns, I think the paper could be published.*

Response and revisions:

We thank the reviewer for his/her comment. We have revised the introduction section as "Most current inventories, however, did not sufficiently consider the differences in application of technologies, or made full use of country- or region-dependent information related to emissions. Global emission factors were applied instead to many sectors" in **lines 67-69, page 4 of the revised manuscript**, to avoid the overstating.