Main revisions and response to reviewers' comments

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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 reviewer 1, which help us improve the quality of our manuscript. Following is our responses to the comments and corresponding revisions.

Reviewer #1

1. *Although this is a very good and important paper by Zhao et al: it addresses the important issue of better constraining the uncertainties surrounding anthropogenic mercury emissions from China, the English grammar and spelling are unfortunately of poor quality. The reviewer was, therefore, unable to read the manuscript comprehensively and critically. For example, in the Abstract and Introduction alone at least 50 grammar and spelling errors were identified that require attention: too many to list, and too many to allow for unobstructed reading and reviewing of the manuscript.*

Hence, before the reviewer is able to fully and critically review the scientific content of this important paper it is ugently requested that the manuscript is first grammatically improved by an English tech-writer, after which it should be reconsidered given the importance and critical topic of the paper.

Response and revisions:

We thank the reviewer's comment and positive remark on the importance of the paper. We also fully acknowledge the language problems in the original draft. The manuscript has been carefully edited, and the language has been improved by a native English speaker. We have tried our best to remove grammar errors and to make the text clear.

Reviewer #2

1. *I think this paper provides some new insights on mercury emissions to the atmosphere from China and the authors describe in detail the challenges and solutions to the challenges faced when obtaining accurate emissions estimates. Some of the language of the paper is making it difficult to interpret the findings, such as the author's statement on line 26 of page 26804: "This is attributed mainly to swiftly increased penetration of advanced manufacturing and pollutant control technologies." Are the authors saying that the Hg emission factors are coming down because more industrial facilities are adopting pollution control technologies? The manuscript could benefit from editing for English language correctness.*

Response and revisions:

We thank the reviewer's comment and fully acknowledge the language problems in the original draft. The manuscript has been carefully edited, and the language has been improved by a native English speaker. We have tried our best to remove grammar errors and to make the text clear.

In **lines 43-45 of the revised manuscript**, we have added a sentence "the unclear operational status and relatively small sample sizes of field measurements of those processes have resulted in lower but highly varied emission factors", to explain why the increased use of advanced manufacturing and pollutant control technologies led to bigger uncertainties of emission estimation.

2. *I do however, wonder if the detailed treatment of the topic might be better off in a more suitable journal.*

Response and revisions:

We thank the reviewer's comment. In the revised main text, we have shortened the description on technical details, and have moved original Figures 3, 7 and 9 that stress technical details to Supplement **(Figures S1, S3, and S4 in the revised Supplement, respectively)**. That we must describe technology developments across so many sectors gives rise to technical density, but this is inherent in a study that seeks to examine globally important environmental effects of diverse processes throughout China's enormous economy.

We have also stressed that this work improves understanding of China's atmospheric Hg emissions in following issues:

1) The comprehensive analysis on sector, spatial distribution and inter-annual trends of Hg emissions for China, with detailed information on the application of technology by sector within the country integrated.

As stressed by AMAP/UNEP (2013), research on the application of technology, both industrial processes employed and technology applied to reduce emissions of Hg in different industries and more importantly in different countries, is a future need and priority to improve the estimate of Hg emissions **(lines 69-71 in Section 1 of the revised manuscript)**. That is also part of reason that we conduct such an extremely detailed analysis of China, as noted by the reviewer.

2) Benefits of recent energy conservation and emission control measures on abatement of China's recent and future Hg emissions, even though the measures are not specifically designed for Hg control.

This work tracks the recent and future possible changes in emission control and emission factors for key industrial sources based on newly developed methods of emission estimate, and reveals that Hg emissions in China have and will be constrained through the national policies of emission control. The growth in energy consumption and industrial production should not be assumed to be a proxy for growth in Chinese Hg emissions. Continuing to believe this will lead to overestimates of China's Hg emission growth and its contribution to atmospheric concentrations and deposition at global scale **(lines 523-545 in Section 4.2 of the revised manuscript)**.

3) Quantitative analysis of uncertainty in Hg emissions, with most significant parameters contributing to the uncertainty identified.

As stressed in AMAP/UNEP (2013), there were very few countries quantifying uncertainties in relation to their national emissions reporting, particularly for developing countries. In this work, therefore, a database for Hg emission factors with uncertainties estimated for all the anthropogenic sources is developed, combining the latest results from domestic measurements. Uncertainties in Hg emissions by sector, species and year are then systematically quantified for the country. In particular, we reveal that the uncertainties of China's Hg emissions are enhanced over time and analyze the reasons for that.

Reviewer #3

1. *This is an extremely detailed study that provides important information about mercury emissions in China and recent time trends. It goes beyond previously-published studies by explicitly treating uncertainty in emissions with a Monte Carlo approach. In addition, the study extrapolates trends to 2030, putting together future emissions scenarios. However, it does not add to the understanding of the Hg problem in China, and gives no indication of whether this estimate is objectively better given our understanding of the environment and atmospheric chemistry. Thus, this paper may be more suitable as an emissions inventory development paper for a journal such as GMD?*

Response and revisions:

We thank the reviewer for his/her important comment. In the revised manuscript, we have stressed that this work improves understanding of China's atmospheric Hg emissions with regards to the following issues:

1) Comprehensive analysis of sector, spatial, and inter-annual trends of Hg emissions in China, integrating detailed information on the application of technology by sector.

There are several global inventories that include emission data for China. (As proposed by the reviewer, those inventories have been reviewed and carefully compared with the current work in the revised manuscript; please refer to our response to Question 3 for details.) Given the high data requirements, however, those inventories have made many rough assumptions about the status of technologies across countries and generally applied global emission factors for most sources, ignoring for simplicity the influence of country- or region-dependent parameters on the emissions. However, as indicated by AMAP/UNEP (2013), research on the application of technology affecting emissions of Hg (both industrial processes and technologies applied in different industries, and, more importantly, in different countries) is a strong need and priority to strengthen estimation of Hg emissions (as reflected in **lines 69-71 in Section 1 of the revised manuscript**). That is also part of reason that we conduct such an extremely detailed analysis of China, as noted by the reviewer. With information on industrial processes and emission control technologies at the provincial-level (or even unit-level for power plants), a database of domestic Hg emission factors by sector is developed, and detailed emissions by region, sector, and year are provided for the country. These are very broad improvements starting with the primary data underlying the inventory, and we believe that the work improves the understanding of the Hg problem in China. In general, global default values (such as those used in the global emission inventories) are what the community employs to get a sense of emissions precisely until more refined input data and supporting methods—i.e., like those we develop and apply in the current paper—can become available. We further examine the emissions estimated with our domestic emission factors compared to those using global ones for particular sources (Figures 2 and 6 in the revised manuscript), and the clear differences suggest the value of incorporating up-to-date domestic information over global default values in emission estimation.

2) Benefits of recent energy conservation and emission control measures on

abatement of China's recent and future Hg emissions, even though the measures are not specifically designed for Hg control.

Understanding the trends in Hg emissions in Asia (mainly in China) is crucial for understanding the global mercury cycle (Jaffe and Strode, 2008). As indicated in **lines 91-94 in Section 1 of the revised manuscript**, historical and future emissions of global primary Hg were estimated to increase under most GHG scenarios, driven mainly by expansion of industrial sources in Asia (Driscoll et al., 2013; AMAP /UNEP, 2013). This is inconsistent with the decreased worldwide trends in background atmospheric Hg concentrations (Ci et al., 2012), and thereby poses a scientific question: are current inventories overestimating growth in emissions? This paper makes an affirmative case for it, and proposes an explanation. Since 2005, China has been implementing a series of control measures on coal-fired power plants and certain other important industrial sources to mitigate serious air pollution across the country. The ancillary benefits of those measures (particularly for sources other than power plants) on Hg emission control have been scarcely considered in previous inventories, potentially driving mistaken conclusions about China's Hg emission trends and their role in the global mercury cycle. This work, therefore, tracks the recent and future possible changes in emission control and emission factors for key industrial sources based on newly developed methods of emission estimation, and reveals that Hg emissions in China have been and will continue to be constrained by national policies of emission control. In particular, as shown in Figure 5(b) of the revised manuscript, the growth in energy consumption and industrial production should not be assumed to be a proxy for growth in Chinese Hg emissions. Continuing to believe this will lead to overestimates of China's Hg emission growth and its contribution to atmospheric concentrations and deposition at global scale **(lines 523-545 in Section 4.2 of the revised manuscript)**.

3) Quantitative analysis of uncertainty in Hg emissions, and identification of the most significant parameters contributing to the uncertainty.

As stated in AMAP/UNEP (2013), there are very few countries quantifying uncertainties in their national emissions reporting, particularly developing countries with limited data of poorer quality, such as China. In this work, therefore, a database for Hg emission factors with uncertainties estimated for all the anthropogenic sources is developed, combining the latest results from domestic measurements. Uncertainty in Hg emissions by sector, species and year are then systematically quantified for the country. In particular, we reveal that the uncertainties of China's Hg emissions are enhanced over time, resulting mainly from swiftly increased penetration of advanced manufacturing and pollutant control technologies that have highly varied emission factors due to the unclear operation status or relatively small sample size of field measurements on those technologies.

We agree with the reviewer that it is important to evaluate the accuracy of the current estimates compared to previous studies; conducting a comprehensive quantitative assessment of the Hg emission uncertainties in China aims to create new metrics for this task. In this work, results of available domestic field measurements are compiled to establish an Hg emission factor database for China, and emission factors are well linked to detailed information on current industrial processes and application of emission control technologies. Thus we believe that this is an improvement of the bottom-up estimates for China's Hg emissions.

The observed concentrations can be the evidence of the estimated trends in emissions. As we stated above, our estimate indicates that China's anthropogenic Hg emissions have been significantly constrained despite fast growth rates in the economy and energy consumption, and if we are correct, it helps explain the decreased Hg concentrations observed worldwide. We don't want to overstate this conclusion, however, and need to acknowledge a limitation that very few long-term Hg observation studies have been conducted in China, in either polluted cities or background areas, as stated in **lines 448-454 in Section 4.1 of the revised manuscript**. We hope that better evaluation of bottom-up emission estimates can be conducted once observations of the inter-annual trends in Hg concentrations become available in China.

Despite slower estimated growth rates, our estimates are larger than those of most previous emission inventories, and this can be supported limited top-down research (e.g., Pan et al., 2007), which implied that China's Hg emissions quantified bottom-up might be underestimated **(lines 569-572 in Section 4.3 of the revised manuscript)**. We also include most recent studies using a chemical transport model to compare different inventories in Section 4.3 of the revised manuscript. With smaller emissions estimated by Wang et al. (2014) and Muntean et al. (2014) than this work, generally lower concentrations were simulated using the GEOS-Chem model compared to observation **(lines 572-577 in Section 4.3 of the revised manuscript)**. Although model evaluation is not the main focus of current manuscript, we prepare the gridded emissions by species and provide them for modeling studies, as shown in **Figure S2 in the revised Supplement**. Please also refer to response to Question 5.

2. *The paper is lengthy, detailed, and extremely dense. In general, lack of English language editing makes the text confusing, introduces error, and limits the ability of the reader to evaluate the science.*

Response and revisions:

We appreciate the comment and fully acknowledge the language problems in the original draft. The manuscript has been carefully edited, and the language has been improved. We have tried our best to remove grammar errors and to make the text clear. That we must describe technology developments across so many sectors gives rise to technical "density," but this is inherent in a study that seeks to examine globally important environmental effects of diverse processes throughout China's enormous economy. We could simplify by focusing on only some segments, but the global implications would be lost and the paper would provide only a narrow, less compelling, advance. The comprehensiveness of the study is a fundamental objective and research strength.

3. *A major limitation to the paper which should be addressed in any revision would be the comparison to previous studies. The paper states (p. 26825): "the estimated China's total Hg emissions in this work can hardly be compared directly with other studies" and gives reasons for this, but regardless of the methodological differences, this is critical for the reader to put this work into context. There are numerous global inventories which contain data for China for different years (2000, 2005, 2010). There are also several top-down estimates based on atmospheric observations. A more thorough discussion of how these results differ would be useful to the reader.*

One substantial oversight is that the authors do not compare to or cite the recently published 1970-2008 inventory of Muntean et al (Science of the Total Environment), of which three years overlap with the present work, and which takes a novel approach to mercury inventory development. In addition, Muntean et al. also introduce a methodology for projecting emissions from ASGM which the authors might find useful.

Response and revisions:

We thank the reviewer for an important comment. We have expanded Section 4.3 and made a more thorough comparison to previous studies, particularly to available global inventories including those by United Nations Environment Programme (AMAP/UNEP, 2008; 2013; Pacyna et al., 2010), International Institute for Applied System Analysis (IIASA, Rafaj et al., 2013), Emission Database for Global Atmospheric Research (EDGAR, Muntean et al., 2014), and Peking University (Y Chen et al., 2013) **(lines 547-550 in Section 4.3 of the revised manuscript)**. Figure 3 in the revised manuscript has been modified, with the estimates for various years between 2005 and 2012 from those inventories illustrated. As indicated in **lines 569-577**, the limited available top-down research (Pan et al., 2007) and GEOS-Chem modeling work (Wang et al., 2014; Muntean et al., 2014) are also included in the comparisons, to support the improvement of emission estimates by this work.

We have further compiled a new table in the Supplement **(Table S8)**, summarizing the emissions from certain sectors in global inventories and in this work. The reasons for the discrepancies between various studies are thus carefully analyzed based on the detailed information of the table and relevant studies **(lines 563-595 in Section 4.3 of the revised manuscript)**.

Regarding ASGM, we have cited the work by Muntean et al. (2014), and have improved the section describing the method and uncertainty analysis **(lines 152-156 in Section 2.1 and lines 221-223 in Section 2.3 of the revised manuscript)**.

4. *While there is substantial uncertainty analysis conducted in the paper, the authors do not well-integrate this with their main findings of trends and projections of future emissions. Given the large uncertainties, why should the reader believe these (relatively small in comparison) changes and projected trends? I think the authors have a method that can account for these effects (with the correlation analysis in the Monte Carlo analysis) but it is not stated clearly.*

Response and revisions:

We thank the reviewer for the valuable comment. As shown in Table 5, parameters related with emission factors contribute most to the uncertainties of Hg emissions, including Hg contents of coal for provinces with large consumption, removal efficiencies of dominant APCD, and emission factors of biomass burning and certain technologies for nonferrous metal smelting. We assume that there is no inter-annual variation for those parameters/emission factors during the research period, even though high uncertainties exist for them in any given year. The changes in emission factors over time at the sector level are thus driven mainly by the penetration of industrial processes and emission control technologies. Under this assumption, each individual parameter/emission factor applied for estimation of emission uncertainties for a given year is statistically correlated with it for another year in the Monte-Carlo simulation framework. The uncertainties in emissions for individual year, whether big or small, are thus not associated with the inter-annual trends in emissions.

Even small increases in large uncertainties over time, given that they are generated using the same methods, is worth noting. We state this assumption in **lines 673-678 in Section 4.4 of the revised manuscript**.

It should be acknowledged that such an assumption is an ideal one, and reevaluation is suggested once more results from field measurements of those parameters/emission factors become available over the long run.

5. *Also, the authors do not state whether they plan to make this emissions inventory available for modeling, which would enable quantitative evaluation of it vs. measurements.*

Response and revisions:

Yes we would like to provide the data for modeling studies and better evaluation of emissions upon request, as we indicate in **lines 486-491 in Section 4.1 of the revised manuscript**. We prepare the gridded emissions at resolution of $0.25^{o} \times 0.25^{o}$ and provide the detailed information in **Figure S2 in the revised Supplement**. In particular, the emissions from coal-fired power plants, cement, and iron & steel production are allocated at unit/plant level using the updated database of Chinese emission sources (Zhao et al., 2008; Lei et al., 2011). We've added the information in **lines 178-186 in Section 2.2 of the revised manuscript**.

6. *A few more minor comments follow.*

The "ASGM" acronym is not as commonly used (M=mining, usually)

p 26826 use of term "actual facts" is confusing

Response and revisions:

We appreciate the correction. The full name of ASGM has been revised as "artisanal and small-scale gold mining" **(line 126 in Section 2.1 of the revised**

manuscript). The term "actual facts" has been replaced with "official statistics" **(line 611 in Section 4.3 of the revised manuscript)**.

7. *There are a lot of figures/tables (6 tables, 10 figures) the authors should consider moving some of them to the supplementary information. In addition, the authors might consider shifting some of the technical details to SI and incorporating more discussion/literature comparisons in the main text.*

Response and revisions:

We thank the reviewer for his/her comment. We have followed the reviewer's suggestion and moved original Figures 3, 7, and 9, which stress technical details, to the Supplement (**Figures S1, S3, and S4 in the revised Supplement**, respectively). In the revised main text, we have shortened the description of technical details, and have expanded the discussion of literature comparisons with the reasons for differences in emission estimates between studies (Section 4.3). Please refer to our response to Question 3 for details.

8. *Given the amount of primary data used, it is unclear in places what is new in this work vs. what has been previously published or collected together. A revision to the writing could help make the use of information from citations more clear.*

Response and revisions:

We thank the reviewer for the comment. We have revised the manuscript, and have stressed the new advances in methods and data compared to previous studies of Hg emission estimates for China.

1) The methods of emission estimation are improved for key sectors including coal-fired power plants, cement, iron & steel production, and non-ferrous metal smelting **(Section 2.2 of the revised manuscript)**. With detailed unit- or technology-based information and the inter-annual changes of those sources

incorporated, the changes in Hg emission factors over time are determined, and the effectiveness of ongoing pollution control measures on constraining Hg emissions is revealed, as indicated in Figure 1 of the revised manuscript.

2) A database for Hg emission factors (and related parameters) by sector is established, with the uncertainty for each emission factor/parameter analyzed and presented as a probability distribution function, based on available results from measurements. We have stressed this in **lines 251-258 in Section 3.1 of the revised manuscript**. As summarized in Tables 1 and 2, the database provides important new support for quantitative uncertainty analysis of anthropogenic Hg emissions in China.

References:

AMAP/UNEP, 2013. Technical Background Report for the Global Mercury Assessment 2013. Arctic Monitoring and Assessment Programme, Oslo, Norway/UNEP Chemicals Branch, Geneva, Switzerland, 263 pp.

AMAP/UNEP, 2008. Technical Background Report to the Global Atmospheric Mercury Assessment. Arctic Monitoring and Assessment Programme, Oslo, Norway/UNEP Chemicals Branch, Geneva, Switzerland, 159 pp.

Chen, Y., Wang, R., Shen, H., Li, W., Chen, H., Huang, Y., Zhang, Y., Chen, Y., Su, S., Lin, N., Liu, J., Li, B., Wang, X., Liu, W., Coveney, R. M., and Tao, S.: Global mercury emissions from combustion in light of international fuel trading, Environ. Sci. Technol., 48, 1727-1735, 2013.

Ci, Z., Zhang, X., Wang, Z.: Enhancing atmospheric mercury research in China to improve the current understanding of the global mercury cycle: the need for urgent and closely coordinated efforts, Environ. Sci. Technol., 46, 5636-5642, 2012.

Driscoll, C. T., Mason, R. P., Chan, H. M., Jacob, D. J., and Pirrone, N.: Mercury as a global pollutant: sources, pathways, and effects. Environ. Sci. Technol., 47, 4967-4983, 2013.

Jaffe, D. and Strode, S.: Sources, fate and transport of atmospheric mercury from Asia, Environ. Chem., 5, 121-126, 2008.

Lei, Y., Zhang, Q., Nielsen, C. P., He, K. B.: An inventory of primary air pollutants and CO_2 emissions from cement industry in China, 1990–2020, Atmos. Environ., 45, 147-154, 2011.

Muntean, M., Jassens-Maenhout, G., Song, S., Selin, N. E., Olivier, J. G. J., Guizzardi, D., Maas, R., and Dentener, F.: Trend analysis from 1970 to 2008 and model evaluation of EDGARv4 global gridded anthropogenic mercury emissions, Sci. Total Environ., 494-495, 337-350, 2014.

Pacyna, E. G., Pacyna, J. M., Sundseth, K., Munthe, J., Kindbom, K., Wilson, S., Steenhuisen, F., Maxson, P.: Global emission of mercury to the atmosphere from anthropogenic sources in 2005 and projections to 2020, Atmos. Environ., 44, 2487-2499, 2010.

Pan, L., Chai, T., Carmihael, G. R., Tang, Y., Streets, D., Woo, J. -H., Friedli, H. R., Radke, L. F.: Top-down estimate of mercury emissions in China using four-dimensional variational data assimilation, Atmos. Environ., 41, 2804-2819, 2007.

Rafaj, P., Bertok, I., Cofala, J., and Schopp, W.: Schopp, W.: Scenarios of global mercury emissions from anthropogenic sources, Atmos. Environ., 79, 472-479, 2013.

Wang, L., Wang, S., Zhang, L., Wang, Y., Zhang, Y., Nielsen, C., McElroy, M. B., Hao, J.: Source apportionment of atmospheric mercury pollution in China using the GEOS-Chem model, Environ. Pollut., 190, 166-175, 2014.

Zhao, Y., Wang, S. X., Duan, L., Lei, Y., Cao, P. F., Hao, J. M.: Primary air pollutant emissions of coal-fired power plants in China: current status and future prediction. Atmos. Environ., 42, 8442-8452, 2008.