Atmos. Chem. Phys. Discuss., 15, C4921–C4929, 2015
www.atmos-chem-phys-discuss.net/15/C4921/2015/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source attribution and process analysis for atmospheric mercury in East China simulated by CMAQ-Hg" *by* J. Zhu et al.

J. Zhu et al.

njuzjl@126.com

Received and published: 15 July 2015

We thank referees for the positive comments and suggestions. In our response, we have addressed all of the concerns of the reviewers and revised the paper accordingly.

Reply to Referee #1: The manuscript presents findings from a modeling study of atmospheric mercury deposition in eastern China. This study offers a valuable addition to the scientific literature by providing detailed information on mercury source attribution in a highly polluted region using a fine resolution regional photochemical model. The results will be potentially useful in the prioritization of mercury control measures for different industries in the region. Technical and editorial comments are provided below. I recommend acceptance subject to these minor revisions.

Q: The modeling results are strongly dependent on the mercury chemistry applied in CMAQ. Also, there is considerable uncertainty regarding which mercury transformations actually happen in the atmosphere. Therefore, it would be helpful to provide the details of the chemical mechanism applied in this study.

Re: We quite agree with the referee that atmospheric mercury chemistry is an important uncertain factor in mercury model now since mercury transformations actually happen in the atmosphere and their reaction rate are not known very clearly. As seen in Section 2.1, the chemical mechanism of mercury used in our simulation is CB05 mechanism including mercury gaseous reactions with ozone, OH, H2O2 and Cl2 as described by Lin and Tao (2003). In the future work, we will try to add the mercury reactions with Cl, Br and BrO in CMAQ model to improve the chemical mechanism of mercury.

Q: The authors appear to have not applied the algorithm for bidirectional mercury in CMAQ but instead included "re-emissions" in the natural mercury emissions. This should be qualitatively discussed.

Re: Yes, we did not apply the algorithm for bidirectional mercury as the CMAQ of version 4.6 we used does not involve this subroutine. Secondary emissions that resulted from deposited mercury transformed to GEM and re-emitted to the atmosphere from soil and water were also considered as described in Shetty et al. (2008).

Q: The basis for the mercury speciation in the anthropogenic source categories should be discussed as this has a strong influence on the predicted impacts. For example, CEM is mostly GEM and thus affects GEM concentrations but has little effect on deposition, whereas IND has a strong effect on deposition.

Re: The mercury speciation in the anthropogenic source categories is given in Table 1. Additionally, the speciation characteristics of each source category were discussed briefly in the beginning of each section for source apportionment (Section 3.2.*).

Q: The work of Zhang et al. regarding mercury in the Beijing area is relevant and should be cited (Atmos. Chem. Phys., 13, 10505–10516, 2013).

Re: OK. We have cited this paper in our manuscript.

Q: "Domestic life" is not a common source category; the basis for the sources including in this category needs to be explained.

Re: As we described in Section 2.2, emission from domestic life (DOM) includes waste incineration, domestic coal burning and application of battery and fluorescent lighting.

Q: In Table 1, Figure 1 and elsewhere in the manuscript: yrËĘ-1 is not scientific notation, suggest using either aËĘ-1 or spell out "year".

Re: Thank you for the comment. We have changed all “yr” in the manuscript to “year”.

Q: How are wildfire emissions of mercury treated? If they are in the natural source category, there should be a non-zero TPM fraction in Table 1. Also, see line 16 on p.10399.

Re: In our simulation, we treat wildfire and biomass burning emission as part of anthropogenic sources with 13.97 Mg/year GEM and 0.42 Mg/year TPM in China area. However, we did not analyze the modeling result about wildfire and biomass burning emission in detail in out manuscript because of the small emission quantity of this category and the uncertainty of spatial distribution from year to year.

Q: Caption for Figure 2 is inaccurate and needs to be re-worded. Clarify the averaging time period used for evaluation of mercury air concentrations

Re: The time period used for evaluation is 2011 and we have added the time in the caption of Figure 2.

Q: p.10393: Line 6: Provide resolution of the parent grid in the nested grid configuration.

Re: We have added the parent grid resolution of 81x81 km2 in this sentence.

Q: p. 10395: Line 20: Provide citation for GEOS-Chem results.

Re: We are sorry that the GEOS-Chem results we used have not been published anywhere for citation.

Q: The grammar and style of writing needs to be checked throughout the manuscript. For example, I recommend the following changes. p. 10391: Lines 9-10: Atmospheric mercury is divided into three species according to various physical and chemical properties Line 12: GEM is the predominant form (> 95 %) in atmosphere; it is very stable

Re: We have revised as referee recommended.

Q: p. 10395-10396: Last line: A comparison was made of characteristics of processes influencing atmospheric mercury species in urban and non-urban areas

Re: We have revised as referee recommended.

Q: p. 10399: Line 1: "NAT was still an important contributor to ..." p. 10396: Line 14: "are shown in Fig. 2"

Re: We have revised as referee recommended.

Q: p. 10399: Line 9: "The effect from NAT was decreasing ..." Abstract: Re-phrase "made gain of mercury"

Re: We have revised as referee recommended.

Q: The notes presented above are just some examples. It would be helpful to proofread the entire manuscript.

Re: Thank you for comments before. We have proofread the entire manuscript again and check the grammar and writing style through the manuscript.

Reply to Referee #3: This is a nice study investigating source contribution and process analysis for atmospheric mercury in a mercury-polluted region using the CMAQ-Hg

model with a nested technique. The findings of this study are useful for better understanding and identifying the key factors that significantly affect atmospheric mercury level and behavior in East China. After the following comments are addressed, I recommend that the manuscript be accepted for publication.

General comments

Q: 1) Anthropogenic Hg emissions from countries around China are generally considerably large and apparently were not considered in this study (Fig. 1a). Those emissions could significantly affect atmospheric Hg level and behavior in China. Thus they could significantly alter modeled results presented in this paper. The potential impact of neglecting those emissions should be discussed in the manuscript.

Re: We have added some uncertainty analysis for underestimate the anthropogenic sources from other countries in Section 3.2.7. It would lead to the underestimate the contribution from out of China. We will improve the accuracy of anthropogenic sources out of China in the future work.

Q: 2) Elemental Hg demonstrates bi-directional mass exchange between air and surface. A bi-directional exchange model is ideal for accurately simulating dry deposition of element Hg. What kind of scheme/model was used to simulate dry deposition in this study? bi-directional or uni-directional? Are obviously different results expected by using a bi-directional exchange model? The manuscript should give a brief description about the dry deposition model used in the study.

Re: As the CMAQ of version 4.6 of we used does not involve the algorithm for bi-directional mercury. Dry deposition velocity of mercury is calculated by MCIP combined with meteorological condition and the category of land use. Secondary emissions that resulted from deposited mercury transformed to GEM and re-emitted to the atmosphere from soil and water were considered in natural sources as described in Shetty et al. (2008).

Q: 3) GEOS-Chem model generally has much coarser spatial and temporal resolutions than CMAQ-Hg. What are the spatial and temporal resolutions of GEOS-Chem output that were used in this study? If they are coarser than those in CMAQ, how were the GEOS-Chem output processed and adapted to the resolutions employed in CMAQ-Hg simulations?

Re: GEOS-CHEM output we used with spatial resolution of 4°*4.5°and temporal resolution of one hour. The output from GEOS-CHEM can be used as the boundary condition for CMAQ with interpolation. However, the coarser resolution will lead to uncertainty for CMAQ modeling results.

Q: 4) In section 3.2, the manuscript presents many numbers and points, but doesn't give sufficient and specific figure citations to support them. It is really difficult for readers to figure them out.

Re: Figure 4 and Figure 5 are used to support section 3.2. Because there are too many categories of emission sources to be compared, it is difficult to express all information in one or two figures and also we cannot show too many figures due to the limited space available for publication.

Technical comments

Q: P10390 L5: "run with nested grid resolution of 27km" to "run with a nested domains"

Re: We have revised as referee recommended.

Q: L10: "regard" to "regarded"

Re: We have revised as referee recommended.

Q: L13: "86.7" to "86.7%"

Re: We have revised as referee recommended.

Q: L24: "7.3" to "7.3 ngm-3"

Re: We have revised as referee recommended.

Q: P10391 L3: Majority of natural emission is element Hg which is chemically inactive. Why is it more significantly than anthropogenic emission to influence CHEM and AERO?

Re: CHEM and AERO indicate the transform from GEM to GOM and PBM. As GEM from natural emission is larger than that from anthropogenic emission, natural emission influence CHEM and AERO more.

Q: P10392 L27: "to simulate of regional" to "to simulate regional".

Re: We have revised as referee recommended.

Q: P10393 L8: "with grid resolution of 27x27 km2" to "with a grid resolution of 27 km".

Re: We think “grid resolution of 27x27 km2” is more accurate.

Q: P10395 L16: "comprises" to "covers"

Re: We have revised as referee recommended.

Q: P10396 L7: "natural emissions (NAT) excluded", reword this sentence.

Re: We have reworded this sentence.

Q: P10397 L15: What are time frames and monitoring frequencies of observations?

Re: The time frames and other detail information of observations were summarized in Zhu et al. (2012) and Zhu et al. (2014).

Q: L24: What are specific unexpected complexity for emission and meteorological condition? why?

Re: As the measurement in Pudong last for one month, some sudden emission (without consideration in our emission inventory) during this time frame will affect the averaged condition much. This will lead to the difference between our simulation result and

observation.

Q: P10398 L13: PBM concentrations was significantly underestimated as well (by 60%).Why is less precipitation predicted the only reason that caused the underestimation?

Re: We agreed with referee. Less PBM concentration predicted is another reason that caused the underestimation of wet deposition in Nanjing. We have added this reason in that sentence.

Q: P10403 L17: This sentence is confusing. Reword it.

Re: We have reworded it.

Q: L19: "a factor of >5 larger than" is confusing. Change it.

Re: We have changed it.

Q: P10404 L8: It looks like EMIS and DDEP, instead of EMIS and VDIF, were also the dominant processes. Please check it.

Re: We have checked it again. This sentence talked about EMIS and VDIF and next sentence focused on DDEP.

Q: L18: How are diurnal profiles calculated? Averaged all over non-urban grid cells for a year? A brief description should be added.

Re: Yes, we calculated diurnal profiles by averaged all over urban or non-urban grid cells for a year. We have revised that sentence.

Q: P10416 : label error in figure caption.

Re: We have revised it.

Q: P10421 : add x axis title

Re: We have added the title.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C4921/2015/acpd-15-C4921-2015-supplement.pdf

---

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 10389, 2015.