Atmos. Chem. Phys. Discuss., 12, C2294–C2299, 2012
www.atmos-chem-phys-discuss.net/12/C2294/2012/


Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Investigation of effects of varying model inputs on mercury deposition estimates in the Southwest US" *by* T. Myers et al.

**Anonymous Referee #2**

Received and published: 9 May 2012

Overall comments:

This work is an extended effort from previous studies to investigate the significance of boundary conditions in regional mercury model simulations by using CMAQ model. The authors varied some model inputs (e.g. Hg concentrations of the lateral model boundaries, meteorological conditions, etc.) and examine the model results (in terms of dry and wet depositions of Hg). Moreover, the authors tried to demonstrate contributions of Hg from the free troposphere toward Hg depositions and ground-level concentration. The topic is interesting but the reviewer thinks that the manuscript still contains some errors which will be pointed out in the next section. Moreover, the reviewer believes that additional model simulations and further discussion are needed in order to meet the publication quality of this journal.

Major concerns:

Page 10276, lines 3-13: “The inclusion of this reaction mechanism in CMAQ 4.7 was found to overestimate the modeled wet deposition when compared to MDN observations (116% normalized mean bias in January and February 2002 simulations and 11% normalized mean bias (NMB) in July and August 2002 simulations) and found to result in ambient low, sub 1 ng m−3 GEM concentrations, in hemispheric CMAQ simulations. The removal of GEM oxidation by the NO3 radical reduced the January and February wet deposition bias (31% NMB) and introduced a negative bias in the July and August 2002 simulations (−23% NMB but decreased the normalized mean error by from 44% to 39%). CMAQ 4.7.1 with this change to the chemical mechanism was found to simulate wet deposition well when compared to MDN observations and CAMx simulations (Baker and Bash, 2012).”

There are several issues from the statements above. First of all, the reviewer thinks that poor model performance of the CMAQ model is not solely caused by the Hg-NO3 reaction. It can be stemmed from model uncertainties such as natural emissions (re-emissions which are estimated to be about 2/3 of the global Hg emission) and/or inaccurate model assumptions [e.g. deposition of Hg(0) is in balance with re-emission]. In fact, low ambient Hg can be compensated by appropriate natural emission estimation. By the way, lack of hemispheric CMAQ descriptions makes it very difficult for the reviewer to fully evaluate the model results. Secondly, the values of NMB from the CMAQ w/ Hg-NO3 (116% and 11% for Jan-Feb and July-Aug, respectively) are comparable with the recent CMAQ model results w/o Hg-NO3 [NMB 62% to 349% (Jan-Feb-Mar) and -49% to 111% (Jul-Aug-Sep)] from Baker and Bash (2012). Finally, the reviewer does not think that one can claimed “well” model performance for those seasonal reported values. The reviewer believes that Baker and Bash (2012) claimed that their model performance is good for their annual evaluation and did not mean for the seasonal results.

Page 10279, lines 17-23: “Figure 2 demonstrates the substantially different estimates



ACPD

12, C2294–C2299, 2012

of mercury deposition that can result from the different boundary conditions. In particular, dry deposition in some parts of California and Nevada drops from 4 $\mu$g m$-$2 month$-$1 using the GEOS-Chem boundary conditions to about 1.5 $\mu$gm$-$2 month$-$1 using the GRAHM boundary conditions. Simulated wet deposition of mercury in some areas of Arizona is about 1.3 $\mu$g m$-$2 month$-$1 using the GEOS-Chem boundary conditions but increases to 1.5 $\mu$g m$-$2 month$-$1 using the GRAHM boundary conditions."

What is the main message that the authors want to deliver? In general (from Fig. 1), GEOS-Chem seems to provide more Hg concentrations to the lateral CMAQ boundaries than GRAHM does for the most Hg species but HgP. However, it appeared (in Fig. 2) that CMAQ/GEOS-Chem simulated higher wet deposition but lower dry deposition when compared with CMAQ/GRAHM. Are those higher wet depositions from CMAQ/GRAHM caused by higher HgP concentrations coming out from the boundaries? It would be more informative if the authors add more discussions such as mercury species or mechanisms that responsible for differences in dry and wet depositions from using boundary conditions derived from the two global models.

Section 3.2: It is not clear why the boundary conditions of GEOS-Chem needed to be adjusted and why the authors chose a hemispheric CMAQ to downscale the GEOS-Chem data. Since the hemispheric model configurations and descriptions are not available to the public, the reviewer is skeptical about validity of the hemispheric model results.

Fig 4: The figures are poorly displayed. The sizes of the figures are different. The unit of the Hg(II) concentration is missing and the x-y axes are not labeled.

Section 4: Meteorological condition seems to pose a strong impact to Hg depositions (especially spatial distribution) which is not surprising. An interesting question would be "How does the model perform under different scenarios (which can be accessed by using various performance metrics)?" instead of "Will model results differ when use different meteorological conditions?" However, model evaluation is missing in this



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12, C2294–C2299, 2012



manuscript. The reviewer believes that adding some evaluations of measurement and simulation data can improve soundness of the manuscript.

Section 5: The zero-out technique used in this section produced some negative values (as high as 17.9%). These negative values cast a doubt over the robustness of the technique used and hence further discussion is required. The authors should also zero-out layers that below 5400 meters for a more complete analysis. It is quite strange that Hg concentrations at high-altitude (above cloud level) would have such a strong impact on wet deposition. In addition, a recent study by Lyman and Jaffe (2011) has estimated contribution of Hg(II) from the upper atmosphere to be only 4% of the Hg(II) deposition globally. Is it possible that Hg concentrations at lower altitude were diluted by the zero-out method? Did the Hg concentrations at the top layers remain zero at the end of the simulation period? Moreover, Hg concentration from ground-level should be shown in order to compare the results of zero-out method and the results of the inert tracer method shown in the latter sections (6&7).

Section 6&7: It is unclear why the authors used as many as 10 tracers for each Western and Northern boundaries and only 2 tracers for each Eastern and Southern as shown in Table 2. It would be more useful if the authors could tag individual Hg species just for two layers (1-12 & 13-14) instead of tagging total Hg.

Page 10285, lines 18-20: “The contribution from upper layers to wet and dry deposition of Hg is large regardless of global model used for boundary conditions.”

The above statement for wet deposition may be true but the authors presented the results from one global model (GEOS-chem). Experiment with other global model can be very different. Besides, the contribution of upper layers Hg to dry deposition is not as large as wet deposition. The reviewer thinks that the statement should be modified.

Page 10286, lines 13-17: “The influence of Hg concentrations on the dry deposition from the free troposphere in CMAQ is in agreement with the model and measurement comparisons of Amos et al. (2012). These results may partially explain the recently



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12, C2294–C2299, 2012



documented discrepancies between modeled and observed speciated mercury concentrations (Baker and Bash, 2012)."

Amos et al. (2012) compared between modeled and observed data in terms of wet deposition and speciated Hg concentrations in the air. They did not compare dry deposition. The reviewer cannot make a connection between this work and the recent work done by Amos et al. (2012). Moreover, the reviewer do not understand what specific results may explain overestimated Hg species concentrations by Baker and Bash (2012). Do the authors suggest that Hg species concentrations at the upper part of the boundaries used in the work done by Baker and Bash (2012) were inaccurate (maybe too high)?

Minor concerns:

Page 10275, line 3: Pongprueska is misspelled. The correct name is Pongprueksa.

Page 10275, line 27: Wrong reference is cited (Subir et al. 2012), the correct one is "Subir et al. 2011".

Page 10276, line 10: Change "by from" to "from".

Page 10277, line 16: GEOS-Chem data were generated from Harvard not MIT.

Figures 2, 5, 6, 7: The scale of the color-bar is not linear and it is very difficult for the reviewer to interpret the data. Is there a particular reason to use such scale instead of using a simple linear scale? Moreover, the bubbles representing observed data embedded in those figures are not easy on the eyes. Those data would be more appropriate if put in a separate table which can also be served as model performance evaluation.

Figures 8, 9, 12: The color-bars are inconsistent. The reviewer suggests using a consistent color scheme for a better comparison.

References



ACPD

12, C2294–C2299, 2012



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