

Interactive comment on “Nested-grid simulation of mercury over North America” by Y. Zhang et al.

Anonymous Referee #3

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This manuscript describes a new nested modeling approach to the simulation of atmospheric mercury where a single model is applied first in a low-resolution global mode to provide boundary values for a secondary limited-area high-resolution nested simulation. Having the same chemistry formulations for mercury in both simulations should minimize some of the modeling artifacts encountered in previous works. However, differences in the resolution of the vertical axis could still lead to artifacts especially near the lateral boundaries of the limited-area model. It is not clear from the text whether the 47-layer structure of the global model was also used in the nested model. I would also caution that “high-resolution” is a relative term. There certainly have been simulations of atmospheric mercury performed at finer horizontal resolution than the modeling applied here. The authors might consider identifying their model domains as “parent” and “nested”.

The authors are definitely correct when they state “Considerable uncertainty remains

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on the Hg⁰ oxidation mechanisms and their kinetics.” In this modeling work, bromine atoms are the sole oxidant for Hg⁰. This brings me to question whether it is appropriate to completely discount all other oxidation reactions and whether the emissions, transport, chemistry and deposition processes for bromine are accurately modeled. I see limited supporting evidence to answer these questions. Despite many uncertainties, this new modeling effort has managed to show fairly good agreement with measurements of wet deposition and air concentration of mercury. The improved accuracy achieved with the “in-plume reduction” assumption is especially noteworthy and begs for the identification of the chemical kinetics involved. Unfortunately, our limited understanding of the fundamental processes of atmospheric mercury necessitates the use of such speculative assumptions in simulation modeling. This work makes use of some important assumptions which improve the modeling results and is worthy of publication. However, I do have some questions that require explanation and some suggestions for improvement of the manuscript as detailed below.

Page 2609, last paragraph in section 2.1: I’m a bit confused by “the HgII tracer is assumed to be partitioned between gaseous and particulate phases”. HgII in the particulate phase would be HgP would it not? Later in section 3.3 you bring PBM (particulate-bound mercury) into the discussion. It needs to be made clear throughout the text what mercury species the model actually resolves and how these relate to the observed species. I personally deplore the term “reactive gaseous mercury” or “RGM” as it is an operational term coined back in the days when mist-chambers were used to detect an unexplained fraction of soluble and presumably reactive gaseous mercury. It is unfortunate that HgII is still a necessary generalization in atmospheric mercury modeling. I was surprised at the success of the HgII gas/particle partitioning in Amos et al. (2012) without knowledge of the mercury compounds present. But we should not fault modelers for trying to move forward in the face of these uncertainties as long as they are adequately explained. I am glad to see that Hg in this model is not being scavenged by the Bergeron process of vapor condensation to cloud ice. But what becomes of the now concentrated mercury left in the remaining cloud water? What if

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that cloud water evaporates entirely? Does GEOS-Chem maintain and transport cloud water separately from cloud ice?

Page 2611, lines 24-26: What is the rationale for applying the in-plume reduction of mercury for incinerators? The phenomenon in regard to CFPPs is poorly explained on the basis of chemistry. Expanding its application to incinerators seems unfounded unless the modeling showed it was necessary to achieve the best agreement with observations.

Page 2612, lines 6-11: Since CFPPs are such an important source of atmospheric mercury it would seem advantageous to apply some sort of temporal resolution during the simulation period. Previous modeling efforts have been able to temporally allocate pollutant emissions from power generation sources by time of day and by season based on well documented power demands. The constant-rate assumption applied here could lead to unrealistic vertical mixing and horizontal transport. Was this a necessary simplification? If so, this should be explained.

Page 2613, lines 24-26: Is the higher wet deposition flux over land really attributable to higher updraft velocity? Is it not actually due to the precipitation arising from upward vertical motion on either side of the land/water discontinuity, whether over land in the daytime or over water at night? The presumption of higher wet deposition over land might be based on a lack of measurement over water.

Page 2614, lines 13-15: The correction for snow bias is not well explained. Do you correct all weekly wet deposition amounts during the entire year by the annual (rain+snow)/snow ratio? This would unduly increase the MDN-observed values in the warm seasons and not fully account for the low collection efficiency of the MDN samplers in the cold season.

Figure 4 as discussed in section 3.2: Why did you not show the full 2-year time line? It would be interesting to see how the simulations performed for each month of the 24 month period in addition to each month in 2008 and 2009 averaged together. Viewing

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Figure 4 with the mistaken impression that it shows model performance over a single annual cycle could give a false impression of model accuracy based on averaging. The same holds true for the many line graphs in Figure 8 where it would be informative to see month-by-month comparisons of model to observation rather than multi-year averages of observations compared to 2008-2009 averages of model results.

Page 2617, lines 22-24: The IPR results for TGM in the marine boundary layer show rather low concentrations compared to most of the observations I recall seeing. See Table 11.1 in Sprovieri et al (2009) which shows considerably higher concentrations. I'm not convinced the cycling of mercury between air and water in the MBL is simulated well.

Page 2618, lines 7-10: Vertical mixing seems to be a reasonable explanation for the higher RGM and PBM in the nested model. However, some documentation of the stronger ventilation of the PBL in the nested model would strengthen the argument.

Figure 8 as discussed in section 3.4: The size of the graphs needs to be increased so that the axis labels are legible. The STD simulation results seem to be missing from the CAMNet graphs. As mentioned before, I think it would be preferable to see the full two-year timeline of monthly modeled and observed values for each site where possible. I understand the CAMNet sites did not operate during the 2008-2009 period of the model simulation. I have doubts about comparing model simulations of one time period to observations taken during an entirely different period.

Figure 9 as discussed in section 3.5: The lower ARCTAS TGM concentrations above 7000 meters with their presumed exclusion of some fraction of HgII suggest that HgII concentrations above the tropopause may be significant. The IPR modeling results show very little vertical structure to both the HgII and Hg0 concentrations, which suggests an incomplete treatment of atmospheric mercury processes near and above the tropopause.

Page 2622, lines 10-13: How does having more of the anthropogenic Hg emissions in

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elemental form in the IPR simulation cause more efficient wet deposition of background and natural Hg emissions? Is there a certain "holding capacity" of the atmosphere for elemental Hg such that adding more from anthropogenic sources caused more efficient oxidation and deposition? This needs further explanation.

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