

Comments on “GNAQPMS-Hg v1.0, a global nested atmospheric mercury transport model: model description, evaluation and application to trans-boundary transport of Chinese anthropogenic emissions” by H. S. Chen et al.

This manuscript developed a Hg module in a nested atmospheric model, by considering the emissions, chemistry and deposition. The authors have evaluated the modeling of total gaseous mercury (TGM), oxidized mercury, wet and dry deposition of Hg. At last, the nested model was used to study the outflow of Hg from China (mainland?). This work may be potentially important, however, I have several serious concerns with the novelty and methodology of this study. There are many mercury models and it's not clear whether the model developed in the paper is more advanced than other models. For example, Br chemistry has been considered in other models (Amos et al., 2012), but not in the present model. Treatment of the re-emissions from land and ocean is a very weak aspect. In addition, there is a lack of detailed methodology in the model, in particular for some key chemical and deposition processes, making it hard to judge if the model is advanced or not. At last, as a major weakness, the diurnal variations and vertical trends are not evaluated, leaving it questionable whether the model captures the key chemical processes of Hg. In general, the present paper doesn't provide enough novelty to get published by GMD.

Please find some specific comments below:

Introduction:

The major Hg chemistry and mechanisms are not well described (e.g. gas-particle partitioning). The authors need to explain what they have improved in the modeling of Hg in the present work. Otherwise, there is no novelty.

2.2 Mercury chemistry:

1/ the effects of temperature and relative humidity on Hg chemistry are not well explained. 2/ the treatment of the gas-particle partitioning of Hg (II) is not clear.

Mercury deposition:

1/ the method description is not clear. Detailed equations and parameterizations for dry and wet deposition are needed, otherwise it is hard to judge if the model is rigorous or not. 2/ it seems all precipitations are treated in the same manner, without distinguishing the large-scale and convective precipitation. 3/ for wet deposition, the release of Hg (P) when water freezes to ice is not considered.

Mercury emissions:

1/ how is the emissions from biomass burning, geogenic emissions, land and ocean specified for Hg(0), Hg(II), and Hg(P)? 2/ neglecting the seasonality of Hg emissions from anthropogenic sources is a weakness of the present work. 3/

a major weakness in this section is the treatment of Hg reemissions from land and ocean. The total emissions from land and ocean are not justified by any observations, and the method used in spatial allocation is not convincing. I don't see any relationship between the biogenic CO emission and the Hg reemission.

Model setup

1/ some information are missed in this section (e.g. what is the time step in the model calculation? what is the vertical coordinate used in the model?) 2/ a coarse-resolution inventory (0.5 degree for AMAP, and 0.5 degree when using GEIA inventory for an interpolation) does not match the resolution in the model (0.3 degree).

Model evaluation

Line 17, Page 6960: the time periods of the measurements do not match with those of the simulations. Dismatch of the time periods when comparing the model with the observations is a major weakness. In particular, there is a large bias when comparing modelled annual mean Hg concentrations with daily measurements by cruise. Unfortunately, the authors only attribute model-observation discrepancies to this mismatch, without making any efforts to assess this influence.

Total gaseous mercury (TGM)

1/ Fig. 3: scatter plots by region are needed to evaluate the model performance when comparing model with observations. 2/ a major weakness in this section is that the reasons for the discrepancies are not well explained. There are uncertainties in emissions, chemistry, and deposition. Without discussion on these sources of errors using enough sensitivity tests, it is hard to judge if the treatments of these processes in the model are rigorous or not. 3/ the modeled TGM over the Pacific is 1.4-1.6 ng/m³, compared to the observed 2.6-3.0 ng/m³. However, this large discrepancies have not been explained. 4/ Fig. 5: in East Asia, as a most important source region, the model doesn't capture the low concentrations in summer and overestimates the TGM concentrations in autumn, and these discrepancies are not explained. As a result, it seems that the model doesn't capture the key processes governing the chemistry and deposition of Hg.

Oxidized mercury

Line 5, Page 6963: the authors don't provide convincing explanation for the overestimation of the oxidized mercury concentrations. As a result, it seems that the model fails to simulate the key processes governing the chemistry and deposition of Hg.

Dry deposition

Line 13, Page 8: the authors attribute the model overestimation to Hg(II) and

Hg(P) emissions. However, a discrepancy of 98 v.s. 648 pg m⁻³ is out of the uncertainty range of emissions. It seems that the model fails to simulate the key processes governing the chemistry and deposition of Hg.

3.7.1 East Asia vs. North America and Europe

1/ Line 5, Page 6966: I suggest that the authors give some estimates of the Hg emissions from 2000-2010 to support their first explanation. 2/ Line 6, Page 6966: I suggest that the authors compare the model-observations discrepancy over East Asia between the global and nested model to support their second explanation. 3/ Line 6, Page 6966: there is no evidences showing that the emission uncertainty in East Asia is larger than that in North America and Europe. 4/ In general, it should be careful when comparing the different model performance among different regions. The miss of some chemical processes in the model should also explain the poor model performance over East Asia. Insufficient explanation of the discrepancy causes the model to be very uncertain.

3.7.2 Global vs. nested simulations

The authors state that the emission, chemistry and deposition are self-consistent between the global and nested simulation. However, from Figure 7 and Figure 9, the regional TOTAL wet and dry deposition seem to be very different between the two simulations. I am not sure if this is only due to the smooth effect of mapping. I suggest that: 1/ the authors remove the smooth effect in these maps by showing the original model resolution; 2/ there should be maps showing the absolute and relative differences between the two simulations; 3/ there should be a detailed comparison of Hg budgets between the two simulations. Then, the author should provide enough evidences to substantiate that the two simulations are really self-consistent.

Impacts of Chinese primary anthropogenic sources

1/ the authors state that 30% of surface Hg concentrations was contributed by China's primary anthropogenic sources. Then, what sources and which regions contributes to the remaining 70%? If the contribution of Hg reemissions is large, only accounting for the impact of China's primary anthropogenic sources would have very limited significance.

2/ change "Hg concentrations" to "surface Hg concentrations" at any place if necessary.

3/ the trans-Pacific transport of Hg is not validated by any observations (e.g. the time series at Okinawa).

Reference:

Amos H M, et al. Gas-particle partitioning of atmospheric Hg (II) and its effect on global mercury deposition. *Atmospheric Chemistry and Physics*, 2012, 12(1):

591-603.