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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Model analyses of atmospheric mercury: present air quality and effects of transpacific transport on the United States" *by* H. Lei et al.

Anonymous Referee #2

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General comment

The manuscript presents description of a newly developed model CAM-Chem/Hg of mercury dispersion and cycling in the atmosphere. The model was thoroughly tested and evaluated against observations. It was also applied for evaluation of chemical mechanisms and for assessment of contribution of the transpacific transport to mercury pollution in the U.S. The subject of the paper is relevant to the scope of the journal and the work makes up a new and original contribution. However, the manuscript contains numerous inconsistencies and requires considerable revision. It can be suitable for publication after addressing the specific comments mentioned below.

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Specific comments

1. Page 2, line 49: The term “reactive mercury”, probably, does not reflect the nature of one of the mercury forms mentioned in the paper (HgII). Divalent mercury (HgII) occurs in the atmosphere both in gaseous and particulate forms. So it should be specified as reactive gaseous mercury (RGM) or gaseous oxidized mercury (GOM).

2. Page 3, lines 53-54: “. . . Several atmospheric models such as GEOS-Chem, CMAQ, and HYPLIT have been developed and achieved successful simulations of tropospheric mercury against available observations . . .” The list of contemporary mercury models is much wider. Some information can be found, for example, in (AMAP/UNEP, 2008; Pirrone and Keating, 2010; AMAP/UNEP, 2013)

3. Page 3, lines 72-74: “. . . aircraft-based measurements in the upper troposphere and lower stratosphere show that ozone concentrations rapidly respond to the variation in concentrations of total mercury and reactive mercury, indicating the importance of the ozone-OH oxidation mechanism in the atmosphere . . .” Co-variation of ozone and reactive mercury does not necessarily mean interation between them. It can result from similarity of depletion mechanisms of Hg and ozone in the lower stratosphere (e.g. in reaction with halogens) as it is mentioned by Lyman and Jaffe (2011).

4. Page 4, lines 88: “. . . Anthropogenic sources take up a quarter of the current mercury emissions . . .” There are different estimates of global mercury emissions (e.g. Mason and Sheu, 2002; Selin et al., 2007; Soerensen et al., 2010; Holmes et al., 2010; Mason et al., 2012). Given uncertainty of these estimates, I would say that anthropogenic sources take up from a quarter to one third of the current mercury emissions.

5. Page 8, lines 185-186: It has been shown by Gardfeldt and Jonsson (2003) that the reaction of aqueous Hg(II) reduction by HO2 should not occure in the atmosphere.

6. Page 9, lines 199-201: It is not clear what is the mechanism of the “uptake by the

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marine boundary layer" mentioned in the paper. Some more extended description is needed.

7. Page 10, lines 220: I wonder why the authors refer the global mercury emissions inventory for 2000 to the Global Emissions Inventory Activity (GEIA). To my knowledge GEIA participated neither in preparation nor in distribution of this dataset. There is no mentioning of GEIA in the paper by Pacyna et al. (2006) cited in the text.

8. Page 13-14, lines 302, 308, 309: ". . . The most-polluted area . . ." Here and after the authors use term "pollution" when talking about TGM. Mercury is not an air pollutant. Background concentrations of TGM are to low to have any adverse impact on human health or ecosystems. The main exposure pathways are through deposition to the surface and accumulation in biota. So it is more correct to refer pollution levels to atmospheric deposition.

9. Pages 15-16, Figures 2, 3: It is evident that the model significantly overestimates the observed TGM concentrations in the Southern Hemisphere and does not reproduce the inter-hemispheric gradient. Some discussion and explanation of this fact is required.

10. Page 19, line 424: ". . .The additional bromine reactions will accelerate the transformation of Hg(0) to RGM, the major part of which will stay in TGM . . ." The major part of RGM will not stay in TGM but will quickly (within hours) removed by precipitation or taken up by the surface.

11. Page 19, lines 428-429: ". . .Bromine may also increase the TGM in some places where the reduction reactions become more significant . . ." It is not clear how bromine as a strong oxidant can increase TGM concentration anywhere.

12. Page 19, lines 431-432: Including the bromine chemistry should significantly affect wet deposition. Therefore, it would be reasonable to study this effect in comparison with the wet deposition measurements.

13. Page 20, line 443: There are up to 35 MDN sites measuring wet deposition over

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the period 1999-2001. Selection of 26 sites is not evident.

14. Page 22, Figure 9: As seen from the figure the model predicts strong vertical gradient of TGM at 60N latitude. TGM concentration is below 0.4 ng/m3 at the heights upper than 700 hPa (ca. 3 km). On the other hand, many aircraft measurements showed relatively even distribution of TGM within the lowest 5-7 km (see Swartzendruber et al., 2009 and references herein). It needs some discussion/explanation in the paper.

15. Pages 28-42: A lot of references are missed in the reference list

References:

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