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Interactive comment on "Application of CMAQ at a hemispheric scale for atmospheric mercury simulations" by P. Pongprueksa et al.

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We thank the reviewer for constructive comments and suggestions. We will rephrase the writ-up as suggested. The reviewer also has some questions and comments regarding the methodology and results that we would like to response as follows:

1. Was there any treatment for the recycling to air of previous mercury deposition?

Yes, the recycling of deposited mercury was pre-estimated using a regression-base model as one of the model emission inputs (offline treatment).

2. Why was the CARIBIC flight trajectories not followed in more detail?

We were trying to capture a seasonal trend of mercury by using the available flight

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data, rather than relying on information from any of individual flight analysis where measurement technology is not yet mature.

3. Is there a reason this particular modeling domain is more desirable than a larger or smaller one?

No, we used this domain in an effort to cover the entire Northern Hemisphere. Without a full access to publication of the US EPA's hemispheric mercury modeling experiments with CMAQ described at the 9th International Conference on Mercury as a Global Pollutant the reviewer mentioned, we learned from the publication which is believed to contain the same study, that our domain's area is almost double the size of their domain (137x137 horizontal grids, presented at the 8th Annual CMAS Conference by Bullock et al., 2009).

4. Did this network (EANET) only measure precipitation and acid deposition? Is it only being used to compare to the WRF simulation of precipitation?

Yes and yes.

5. It seems likely that Case 2 produced greater concentrations of mercury in precipitation than Case 1 because the combined HgII+HgP concentrations at the lateral boundaries defined from the GLEMOS model (Case 2) are so much higher than those from the GRAHM model. These species are most readily wet deposited and it appears that their definitions at the lateral boundaries may have had a significant effect on the hemispheric simulation of wet deposition of mercury. Was this investigated?

Yes. The results of Hg wet deposition have some impact from IC/BC as expected, but we opted not to include them in this manuscript.

6. Why are there 14 sites in one comparison and 5 sites in the other as shown on Figure 5?

There were 14 EMEP sites in 2005 that reported the concentration of TGM, Hg0, or HgT in the air. In the mean while, there were only 5 sites that reported the concentration

of HgP or HgII (4 sites for HgP and one site for HgII to be exact).

7. Do you mean that daily averages of the CMAQ simulated air concentrations along the CARIBIC trajectories are used to compare to the actual measurements?

Yes. Mercury is an inert chemical; therefore, it should not have much temporal distribution at the upper troposphere.

8. I assume that "HgT" represents total mercury concentration in air, but it is not described anywhere in the text.

We had described HgT in Table 2. We will add more in the text.

9. Page 1731, lines 23-24: Figure 6 seems to contradict the statement here that the two modeling cases produced similar outputs for TGM. There are obvious differences both at ground level and at aircraft level. As for Hg0 and HgT, annual average concentration patterns for these species are not shown anywhere in the paper. It would be helpful to show them to demonstrate similarity between the modeling cases. Similar plots of the RPDs across the domain would also be helpful.

The differences of TGM concentration (RPD < 10% at ground-level and RPD < 15% at aircraft-level) are not considered significant for the most part of the domain (ca. latitude > 20 deg., see attached new Figure 6). The RPD of HgT and Hg0 would be similar to the TGM results because Hg0 is the dominant species in HgT. For the other Hg species, similar RPD plots (both ground and aircraft levels) are added into the attached figures. At the aircraft-level the values of RPD for each Hg species (Hg0, HgII, and HgP) are somewhat higher than those at the ground-level.

The reviewer raised a lot of questions for section 3.3. The authors agree that this section is quite difficult to interpret and the figures are not easy on the eyes. We in fact consider shortening the section and emphasizing more on the earlier section (section 3.2) where the results are more straightforward. We would like to respond to the questioned for section 3.3 as follows:

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10. How many monitoring flights were there in total? Were the flights actually monthly as suggested in the caption of Figure 7?

There were 28 monitoring flights with total flight duration of 51 days from mid-2005 to early-2007 being used in Figure 7. The measurements were performed out about once a month in average.

11. How varied were the flight altitudes? Were the CARIBIC data only used while the aircraft were at cruising altitudes?

The static pressure of the aircraft ranged from 186 to 697 millibar representing cruising altitudes.

12. Page 1734, lines 19-21: This sentence indicates that the bubbles in Figure 7c and 7d show the peaks of daily average TGM for the entire year "at each longitude". How were these longitudes selected? Is it every one degree?

Those figures show model results of the two cases. The longitudes were selected by using the simplified flight trajectories and plotted at 1-degree interval. The bubbles are the highest simulated TGM values for the entire year at each longitude.

13. Page 1734, line 25-29: It seems implausible that winds speeds of less than 0.5 m/s would be found at flight altitudes near 10 km over Europe and China, except on rare occasions. Was this indeed the criteria you used to define air stagnation cases?

We use wind speed lesser than 0.5 m/s at the ground level (not the flight altitude) as the criteria to define air stagnation.

14. Page 1735, line 8: By "annual peaks of daily average" do you mean the highest daily average in the entire year?

Yes.

15. Page 1735, lines 17-18: It is hard to believe that the daily TGM peaks at 10km altitude could be caused by emission sources in that area combined with air stagna-

tion. How likely is it to have effective atmospheric mixing from the surface to 10 km regardless of the wind speeds throughout that layer?

Though air stagnation is a rare phenomenon, it's plausible because a vertical height of 10 km is not relatively far when compared to the horizontal grid size of 108 km especially for such an inert chemical as mercury. So it is more than likely that the high concentrations evident at 10 km altitude come from the emission sources in that area.

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Fig. 1. New Figure 6 (RPD plots added)



Fig. 2. RPD for Hg Species at Ground-level

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Fig. 3. RPD for Hg Species at Aircraft-level