

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

Anonymous Referee #1

Received and published: 30 August 2011

This manuscript describes adaptation of a model that formerly has been used only for regional air quality studies. Results and conclusions from these new hemispheric simulations are generally reasonable, but some of the analysis performed is not well explained and some of the conclusions drawn do not appear to be supported by the evidence presented. The comparison of 3-dimensional model simulations with measurements taken from commercial aircraft is especially confusing. The authors may want to reconsider the type of graphics used to describe model-to-measurement comparisons. Details in many of the figures were impossible to see clearly in the documents available for review. The text is rather difficult to interpret in some places because of unusual phrases and incorrect English grammar. The paper does demonstrate that expansion of the CMAQ model to a hemispheric scale can reduce problems that are associated with lateral boundary effects and that such a scale of simulation is appropriate for long-

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lived air pollutants like mercury. The work is generally worthy of publication, but more or better-described evidence is needed for some of the other conclusions. Specific comments supporting this appraisal are given below.

Page 1724, lines 14-17: Instead of saying that the hemispheric models give a better circulation of the pollutant, you might want to say regional models are not capable of simulating zonal circulations around the globe.

Page 1725, lines 8-9: Instead of “understanding”, I would say “lack of understanding”.

Page 1725, line 10: Instead of “distributing”, I would say “distribution”.

Page 1725, line 13-14: I am not sure what is meant by “These proceeding studies would lead to a better understanding of Hg cycling in the atmosphere.” I am pretty sure that these studies did add to our understanding of mercury cycling.

Page 1726, lines 23-25: I think what you are trying to say here is that grid nesting within a single model going from hemispheric to regional scale is a better option than developing boundary conditions for a regional model from a separate global or hemispheric model having a different definition of physics and chemistry.

Page 1728, line 13: More information is needed on how this modeling study defined emissions of mercury from natural processes. Was there any treatment for the recycling to air of previous mercury deposition?

Page 1729, line 5: Why were the CARIBIC flight trajectories not followed in more detail? Intercontinental flights do not always follow the same path, vertically or horizontally. If these CARIBIC measurements were only taken once a month or so, it seems they could each be analyzed separately.

Page 1729, line 26-27: The polar stereographic projection used in this work (187 x 187 with 108 km horizontal resolution) is exactly the same grid definition used by the US EPA in hemispheric mercury modeling experiments with CMAQ described at the 9th International Conference on Mercury as a Global Pollutant. Is there a reason this

particular modeling domain is more desirable than a larger or smaller one?

Page 1730, lines 11-12: Figure 4 referred to here does not show any Hg concentrations in precipitation from the EANET. Did this network only measure precipitation and acid deposition? Is it only being used to compare to the WRF simulation of precipitation?

Page 1730, lines 18-19: 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?

Page 1730, lines 22-25: It would be helpful to show some of the Lin et al. (2007) results to support the statement that the results of this study were more favorable. At the very least you should be more specific about what you mean by "more favorable".

Page 1731, lines 1-5: I am confused by the grouping of the HgP and HgII concentrations at the EMEP sites. How could you group HgP with HgII? They are two separate mercury species that I would assume are easily measured separately. Why are there 14 sites in one comparison and 5 sites in the other as shown on Figure 5? I also do not understand how the aircraft measurements could be daily averages. Do you mean that daily averages of the CMAQ simulated air concentrations along the CARIBIC trajectories are used to compare to the actual measurements?

Page 1731, lines 20-22: I don't understand the last sentence in this paragraph at all. I assume that "HgT" represents total mercury concentration in air, but it is not described anywhere in the text.

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

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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.

Page 1732, line 8: Are you saying that Hg0 is the dominant species of mercury in TGM and HgT? If so, I would replace "(TGM and HgT)" with "in TGM and HgT".

Page 1732, lines 19-20: Here again, I think the differences in annual average TGM shown in Figure 6 are rather significant. A polar stereographic projection extending across the equator gives the false impression that areas near the lateral boundaries are much larger than they really are. I would say that there are obvious differences in TGM concentration over most of the true area of the model domain. Apparent similarities between Figure 6a and 6b could be due largely to the color map employed. Figures 6c and 6d show fairly large differences in aircraft-level TGM near the lateral boundaries. Plots showing the RPD in TGM at both levels would be very helpful to support statements made throughout section 3.2.

Page 1733, lines 25-26: Again, I don't understand how the CARIBIC data are daily averages. Figure 7a does not appear to show all of the measured data from May 2005 to March 2007, as the vertical axis only goes from day 1 to day 360 (or maybe 365). The bubbles in Figure 7a and 7b are so close together that it is impossible to interpret their colors. I suggest you use separate line graphs of the modeled and measured data for each of the CARIBIC flights (or for each line of bubbles currently shown). I'm not really sure what each line of bubbles is supposed to represent.

Section 3.3 CARIBIC aircraft data and CMAQ model results: This section is the most confusing part of the manuscript. I have a lot of questions about the CARIBIC data and how they are compared to the CMAQ simulation. How many monitoring flights were there in total? Were the flights actually monthly as suggested in the caption of Figure 7? How varied were the flight altitudes. Obviously the flight altitudes would be

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lower near the departure and arrival cities. Were the CARIBIC data only used while the aircraft were at cruising altitudes? In general, the methods used to compare the CMAQ simulation to the CARIBIC data are not clearly described and Figure 7 attempts to encapsulate too much information.

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? The values indicated by the color of the bubbles in Figs 7c and 7d do not match the color of the background pattern where they are located, so I am left to assume these bubbles represent the measured peak values and the background pattern is derived somehow from the model. Figure 7 may be an innovative way to describe a lot of data in a small space, but it needs a much better explanation.

Page 1734, line 25-29: It seems implausible that wind 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?

Page 1735, lines 1-6: The grammar in this paragraph is poor, making the intended message difficult to understand.

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

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

Page 1736, lines 13-15: I do not think it is accurate to say “boundary effect is not obvious” when Figure 9c shows non-zero RPD for HgII maintained near the western extent of the trajectories (30W to 20W) for the entire year of the simulation. The effects

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of initial conditions for HgII do diminish to near zero after a month or two. Maybe you should say the temporal boundary effect is not obvious or that it diminishes to near zero.

Section 3.4 Future model improvement: Some of the statements in this section are quite obvious and could be omitted. The first sentence seems to say the obvious, that simulations can be improved by using better input data and by using more realistic models. Unless you can specify the data or the model processes in question, this sentence is superfluous. The remainder of the first paragraph of this section was difficult to understand, but it seems that the intended message was quite obvious. Better meteorological and emissions input data will improve model simulations. The idea proposed at the end of the section, using hemispheric simulations from a model to inform the boundary conditions for regional simulations of the same model certainly has merit. Inconsistencies in model formulations between a global/hemispheric model providing IC/BCs and a regional model using those IC/BCs can certainly cause problems in the regional model. In fact, direct model grid nesting could eliminate all spatial and temporal dispersions between the two models.

Section 4 Conclusions: The comparison of the CMAQ simulation to surface-level data is not very strong. The only evidence given is in Figures 4 and 5 in the form of box plots where the number of actual observations is not clear. It seems the observational evidence for mercury concentration and deposition is severely limited and confined to only a few specific regions of the globe. The conclusion that the GRAHM and GLEMOS modeling cases yielded comparable results seems to be based on the fact that one case was superior in terms of medians and the other was superior in terms of means. There are a lot of differences between these cases that are evident throughout the paper, but not fully discussed. To say they are comparable is puzzling. Although it may very well be true, there is no evidence presented in this paper to support the conclusion that modeling at large scales also benefits simulations of short-lived pollutants. Finally, the conclusion that elevated TGM concentrations in East Asia are caused by

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air stagnation is only very weakly supported for reasons outlined above.

Interactive comment on Geosci. Model Dev. Discuss., 4, 1723, 2011.