

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

**P. Pongprueksa et al.**

pruek.pongprueksa@lamar.edu

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We thank referee #2 for the constructive comments and suggestions. I would like to response to those comments and suggestions below:

1) .....an application of CMAQ-Hg on the hemispheric scale was already reported two years ago. No new model development is shown here.

By details, this study is absolutely different from the early works by Bullock et al. (presented at CMAS conferences in 2009) in many aspects. Firstly, the number of horizontal model grid-cell in our work is almost 2 times larger than the number from Bullock's domain (1.86 times, to be exact). For the second aspect, we performed a much longer model simulation period (one year plus 10-day spin-up) than what they did (15 days).

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In total, our simulation period is 25 times longer than their simulation period. To the best of our knowledge, there were no comparisons between measurement data and simulation results of CMAQ on a hemispheric scale being reported before. We are confident that our model configurations are appropriate in this study.

2) .....no comparisons with CMAQ runs on the regional scale are shown in this paper.

We agree that a comparison between CMAQ on a regional scale and a hemispheric scale would provide some useful information. However, mercury model simulations on a regional scale such as a CONUS domain are significantly affected by BC as demonstrated in a previous study by Pongprueksa et al. (2008). Also the current study suggested that results from the two different global models were convergent. Therefore, the outputs from simulations of both CMAQ on hemispheric and regional scales that uses IC/BC derived from the same hemispheric scale are expected to yield very similar results regardless of versions of the model where model physics do not vary significantly.

Bullock et al. (2008;2009) also performed an inter-comparison of mercury models using several downscaling combinations of three regional models (i.e. CMAQ, REMSAD, and TEAM) and three global models (i.e. CTM, GEOS\_Chem, and GRAHM) in a North America domain (NAMMIS). The study demonstrated the strong impact of the BC obtained from global models. For example, as shown in the attached Fig. 1, the model performance statistics for all three regional models have the same trend as influenced by the global model data, indicating the importance of the global model data towards model downscaling.

3) The comparisons to observations presented in this paper to demonstrate the quality of the model results are worthless because they rely on model runs which are not performed in a proper way.

We understand that the comparisons of model results to the observation data after the proper spin up period would yield a true performance assessment of the CMAQ model.

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However, in this work, we do not intend to evaluate the model, instead we aim to examine a promising simulation technique in reducing its dependency on global models. Between the two cases, comparisons were done in order to show the differences in results caused by using two different global models. The comparisons between simulated precipitation and measurement data are beneficial in demonstrating validity of the meteorological input.

We think that our simulation results shown in this study are sufficient to address what we focus (i.e. influences from IC/BC and demonstrate model configurations as well as model spin-up requirements). We intend not to perform further simulations because we recognize that the chemistry of atmospheric mercury is still under development and our key conclusions would not be altered by performing those extra simulations.

4) When investigating the influence of the ICs on the model results, this should not be shown along the flight path alone. The authors should also distinguish clearly between ICs and BCs and treat them in different sections.

We will add the figures showing the Hg concentrations changes for the entire domain. We can evaluate both IC/BC altogether as it has been generally recognized that the effects of IC would be reducing while the simulation time is increasing (not exactly for how long just yet), likewise, the effects of BC have been known to occur near the model lateral boundaries (proximity was not known accurately) and do not change with time.

5.1) It is not clear what the bullets in Fig. 7a really represent and how the underlying color map is constructed. In Figure 7a it is unclear how the high TGM values at 20 W - 20 E between day 100 and 170 were derived.

The bullets or bubbles represent the data recorded during the flights from 2005 to 2007 with days of those years represented in y-axis. A similar plot has been used by Slemr et al. (2009) to show the same CARIBIC measurement data. The main purpose of making such plot is to show seasonal variation and unexpected high TGM peaks. The plot is also useful in supporting the assumption of the concentration peaks to be

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caused by air stagnation. The plot was generated by using a Delaunay triangulation of the data with the nearest neighbor interpolation (please see the link for more details: <http://www.mathworks.com/help/techdoc/ref/griddata.html>).

5.2) The flights comprise a period from of almost two years (2005 to 2007), as stated in line 26 on page 1733, but the figure is only for one year.

We combined the data between 2005 and 2007 in one single plot in order to show a full-year dataset (apparently some data are still missing). We assumed that TGM concentrations were not varying across those years and it would show a seasonal pattern.

5.3) Fig 7c and 7d clearly show the influence of the ICs, which is probably the reason why the highest TGM levels are detected at the end of your simulation period.

The highest concentrations of TGM at the aircraft level shown around the end of the simulation period could be an indicator of Hg transported from the lower model levels where concentrations are usually higher.

5.4 In section 3.3.2 you try to explain these "peaks" with vertical mixing into the upper troposphere (7-10 km altitude). From a meteorological point of view this vertical mixing under stagnant conditions in winter is not plausible.

The peaks we focused are the peaks in Asia that occurred during summer and early fall. Besides, frequent observations of strong updrafts of adiabatically cooled and washed-out tropospheric air rich in chemical and greenhouse gases by convective overshooting demonstrate the high frequency of occurrence of such events in contrast to their generally assumed scarcity (Jean-Pierre, 2010).

5.5 I would expect that the model is not able to properly represent the tropopause, the vertical resolution in higher altitudes is too low.

We agree that our vertical resolution is too coarse and this model is not capable of performing simulation of chemicals in the upper troposphere and lower stratosphere

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with high accuracy. However, this would not change our conclusions since Hg pollution usually occurs within the lower troposphere.

5.6 Large differences between the model results and the observations can be expected for these cases. This is not discussed at all in the paper.

Apparently, the plots of measurement data and model data were quite different. We did not discuss these differences because the measurement data seem to be insufficient and there are still lacks of understanding in Hg chemistry especially in the upper atmosphere.

Note: The recommendations are similar to the comments that are already answered in our response above.

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**Table 3. Various Model Performance Metrics Based on Weekly MDN Sample Collections in 2001<sup>a</sup>**

	CMAQ			REMSAD			TEAM			MM5 Precip.
	CTM	GEOS-Chem	GRAHM	CTM	GEOS-Chem	GRAHM	CTM	GEOS-Chem	GRAHM	
$r^2$	0.15	> 0.12	< 0.14	0.16	> 0.15	< 0.16	0.14	> 0.12	< 0.18	0.35
Mean bias (ng m <sup>-2</sup> )	-12.2	< 46.9	> 40.2	67.8	< 100.2	> 41.3	164.2	< 220.3	> 155.2	1.9 (mm)
Mean error (ng m <sup>-2</sup> )	178.1	< 213.0	> 207.0	226.0	< 248.7	> 213.8	278.8	< 326.3	> 264.8	15.3 (mm)
Normalized mean bias	<b>0.049</b>	< 0.187	> 0.160	0.270	< 0.399	> 0.164	0.653	< 0.876	> 0.617	0.078
Normalized mean error	0.708	< 0.847	> 0.823	0.899	< 0.989	> 0.850	1.109	< 1.298	> 1.053	0.620
Mean fractional error	0.725	< 0.771	> 0.759	0.839	< 0.861	> 0.835	0.885	< 0.928	> 0.867	0.641

<sup>a</sup>The most desirable outcomes for Hg wet deposition statistics are shown in bold.

**Fig. 1.** An illustration of strong global models' impact to regional models by using data from Bullock et al. (2009)