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Interactive comment on "Long range transport of mercury to the Arctic and across Canada" *by* D. Durnford et al.

Anonymous Referee #1

Received and published: 10 March 2010

General Comments:

This article describes a global-scale modeling assessment of atmospheric mercury transport from four separate regions to a variety of monitoring sites in the middle and upper latitudes of the Northern Hemisphere. A large amount of information is provided in this paper, possibly too much. Figures 3 through 8 as printed from the "printer friendly" PDF version of the paper were too small to clearly resolve important details discussed in the text. Figure 3 is already separated into segments based on the latitude of the observation site. I would suggest figures 4 through 8 also be separated in some way to allow the graphics to be larger and provide better resolution of detail.

The paper describes correlation statistics for observed concentrations versus the simulated contributions from each of four source regions. However, the analysis of total

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simulated air concentration of GEM (or is it TGM?) from all sources is entirely qualitative and based on different years of observation depending on the monitoring site. Based on the limited evidence presented in the form of time series plots of GEM or TGM, it is not reasonable to assume that the GRAHM model is simulating atmospheric mercury any more accurately than any other model being used today. There is no test of the model's ability to simulate wet or dry deposition of mercury accurately. Nor is there any demonstration that the simulated reduction-oxidation balance in air or in cloud water is correct. Yet the authors often use words like "determined" instead of "estimated" or "simulated" to describe their assessment results.

This paper does provide valuable information derived from one particular model that can be used to estimate (not determine) primary transport pathways and sourcereceptor relationships for atmospheric mercury on the global scale. None of the findings, in general, run counter to widely accepted notions about global-scale cycling of mercury in the atmosphere. This work is one more piece of evidence in a continuing puzzle, and as such, it should be published after a few technical issues are addressed.

Specific Comments (referenced to the printer-friendly PDF format):

p. 4675, line 3: The statement that mercury is non-toxic should be made specific to elemental mercury and referenced to a published health study. While elemental mercury may have been consumed by humans in hopes of a cure for various ailments before the advent of modern medicine, I don't believe it has ever been proven safe for human consumption.

p. 4675, line 16: I do not understand what is meant by "as a result of resistances". I assume that dry deposition is being discussed here and dry deposition is hindered by resistance term in most numerical modeling. Maybe it should just be stated that deposition also occurs by way of dry processes mediated by resistances to canopy/surface.

p. 4677, line 25: Given that a large fraction of total gaseous mercury (TGM) could in fact be in oxidized form during Atmospheric Mercury Depletion Events (AMDEs), I

have to take issue with the statement that GEM vs. TGM is not an important distinction for this study. Simulation of AMDEs is said to be an important positive aspect of the GRAHM model. The partitioning of TGM between GEM and RGM is certainly important to all atmospheric mercury modeling, not just in the case of AMDEs. To trivialize this distinction here is somewhat disingenuous. The lack of distinction between TGM and GEM in the observations used to evaluate the GRAHM model or any model is certainly unfortunate and needs to be addressed in future work.

Section 2.1.2 Model: The total simulation period for which the GRAHM model was applied is not stated. It appears that a 3-year spin-up period is applied, but it is not clear if the long-range transport (LRT) events described later in the text are accumulated during the 2000 to 2008 period encompassing all of the observation periods at each monitoring site, or if the LRT events are only accumulated during the year of observation for each site. I suggest that the entire simulation period be described here and that section 2.2.2 describe the specific period over which the LRT events are defined.

p. 4680, lines 8-9: I fail to see how smoothing the simulation based on a 12-hr running mean helps to pick out important features. It may help to show trends, but features? Maybe it should just be said that the observations were similarly smoothed using a 24-hr mean (daily average or running mean?). But then, why use 12-hr smoothing for the simulated data. I understand that spatial smoothing by the finite grid of the model was considered here, but why impose any temporal smoothing of the simulation when the spatial smoothing from the finite grid may already be overwhelming. This discrepancy in smoothing between the observations and the simulation needs further explanation.

p. 4682, line 6: To be qualified as an LRT event, does the maximum concentration need to be simply 0.25 of the base run's standard deviation, or does it need to be that quantity over the mean? If the LRT events are defined from the base run, how do you know if it is coming from a particular source region when it is qualified as an intercontinental transport event?

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p. 4686, lines 7-10: How can you conclude that meteorological and chemical processes are similar at "these" sites? Are the sites you refer to only those in Canada? There seem to be differences in mercury behavior between just about all observation sites used in this study. Certainly you are not meaning to say that AMDEs occur the same way at all of the sites, even those at lower latitudes.

p. 4686, lines 12-17: Figure 4 does not show total contributions from all four source regions. It only shows the contributions from each one. It might be helpful to actually show the total contributions if Figures 4 through 8 can be separated and expanded to show details. Nonetheless, I don't think the elevation of the four source region's average combined contribution of 56% over their combined fraction of global total emissions of 46% has much to do with northern hemisphere emissions being greater than southern hemisphere emissions or the efficiency of interhemispheric mixing. It is most likely just an indication of the relative proximity of the source regions to the monitoring sites.

p. 4687, line 18: The word "is" should be "in".

p. 4693, lines 16-17: Earlier, it was stated that local emissions were a large influence on the Reifel Island site, especially in autumn and winter. This is likely another reason for the lower than expected number of Asian LRT events at that site. Heavy local influences could mask those from LRT from any distant location.

Section 3.2.1 Horizontal distributions: Why was 516 sigma (\sim 523 hPa) chosen as the top of the vertical column for the analysis of the horizontal distribution of GEM? Why not go to at least the tropopause height (200 to 300 hPa) to include jet stream transport? The winds shown in Figure 7 are winds from 925 to 400 hPa. Why this different vertical section of the atmosphere? Are these vertically averaged or average winds taken from a variety of levels in this range? Maybe these layer-average winds are not really needed. Long-range transport is a three dimensional phenomenon. Horizontal translation is only a part of what drives long-range transport. Flows can be re-circulating in three dimensions with no indications of such in layer averaged wind analysis.

p. 4697, lines 25-26: Are you saying that the meteorological and chemical processes affecting mercury are the same at all of the 17 observation sites? Maybe you mean to say the actual and simulated processes are similar.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 4673, 2010.

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