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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimation of speciated and total mercury dry deposition at monitoring locations in Eastern and Central North America" *by* L. Zhang et al.

L. Zhang et al.

leiming.zhang@ec.gc.ca

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We greatly appreciate all of the comments, which have improved the paper. We have addressed all the comments in the revised the paper as detailed below.

RC- Reviewer's Comments; AC – Authors' Comments

RC: General Comments

This manuscript details model estimates of annual GEM, GOM, and PBM dry deposition to several sites in Eastern and Central North America. Although this model is currently regionally specific I believe this to be a very important progression in mod-

eling mercury deposition. By using measured concentrations of speciated mercury, I believe this is an important use of an established network of atmospheric measurements. I do believe, however, that the authors should be careful calling this model more “realistic” than other models. The Tekrans used to measure speciated mercury measure an “operationally defined” GOM and PBM quantity, therefore it is not actually known what “realistic” is. As other more global mercury monitoring networks come online, tools like this model will be extremely important. After a few minor revisions, I support publication of this manuscript.

AC: See our detailed answers below.

Specific Comments

RC: 1) There are several typos. I will mention the ones I found in the technical suggestions, but this manuscript could benefit from an outside proofread. There are also severalareas where the qualitative discussion could benefit from some quantitative discussion/details. More specifics follow.

AC: Typos have been corrected. Careful proofreading has been done of the revised paper. Wherever possible, qualitative discussions were replaced with quantitative discussions.

RC: 2) The entire manuscript could benefit from some greater explanation of variation and trends. More in-depth discussion of the model predictions at individual sites, or geographically similar sites would be very helpful. For instance, P. 2794 lines 1 – 13 provide only a cursory look at the GEM concentrations, mentioning “two other urban sites”. I feel more discussion is needed here. Other areas that need more in depth discussion of regional or site by site variation, and what may be driving that variation are page 2795 lines 25 – 30. Terms like “generally” should also be avoided (i.e. page 2796 line 1) and more specifics should be given.

AC: We have improved the discussion in Section 3.1 based on this and the other two

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reviewers' comments. We feel that a too-detailed discussion on concentration fields should be avoided here because: (1) the focus of the present study is on dry deposition; (2) detailed analyses of the concentration data have been published for a few sites by individual groups who collected the data; and (3) from the presentation in the 10th mercury conference in Halifax (July, 2011), another group has been working on these concentration data and solely focusing on the geographical pattern of the concentration. We do not want to overlap their work.

The geographical variations of GOM and PBM dry deposition were mostly decided by the emission sources (and thus the concentrations) while the deposition of GEM strongly depends on the surface types and meteorological conditions. These factors have all been discussed in the paper.

RC: 3) P. 2793 line 24 to 2794 line 13 – This entire paragraph needs to be more quantitative. The qualitative descriptions are good, but should be backed up with some data. "Seasonal variation of GOM and PBM were highly variable. . ." What is meant here? What does highly variable mean? What were the ranges? Was there also some diurnal variation? P. 2794 – line 4 – What does "GOM concentrations in spring were much higher than in any other season" mean? 10 times higher? 50 times? And so on for that paragraph. . ..

AC: Section 3.1 has been revised substantially to make the discussions logical and quantitative. As mentioned above, we tried to avoid discussions that were too detailed so the diurnal variations were not included here.

RC: 4) P. 2794 line 15 to 18. A brief description of why the Vd values vary based on these conditions would be helpful.

AC: The first sentence has been rewritten. We have also added a statement in the revised paper (the second paragraph of this section) that all of the resistance terms are smaller under strong wind conditions.

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RC: 5) P. 2795 line 3 – GEM, Vd was higher during seasons with larger LAI. This is redundant with the earlier paragraph and still does not explain why.

AC: The sentence has been revised to: 'Vd was higher over forests and during full growing seasons than over other surfaces or during other seasons due to the dominant effect of LAI on Vd."

RC: 6) P. 2795 line 12 – 28. The annual total GEM deposition numbers are high. How do these estimates compare to other model/measurement estimates. Others have done estimates without doing re-emission estimates, so how do these compare and why are they so high?

AC: Most existing studies have simply excluded GEM in the dry deposition budget. A few studies or Hg transport models have included GEM dry deposition but with Vd values one to two orders smaller than the one used here (see Baker and Bash, 2012 for example). We have added this explanation in the revised paper.

RC: 7) P. 2797 Line 11 (and line 22). As mentioned earlier, I believe that this statement may be a little misleading. Large scale atmospheric transport models are typically based on known chemical kinetics and known chemical constituents. However, measurements made by Tekran analyzers are operationally defined and it is not known exactly what they are measuring therefore, the statement that estimates made from AMNet data are more "realistic" is entirely misleading.

AC: Although the Hg transport models are based on known chemical kinetics and known chemical constituents, the concentration of GOM+PBM predicted by these models were much higher (e.g., a factor of 2 to 10) than available data (Zhang et al., 2012; Baker and Bash, 2012). These large discrepancies were probably caused by a combination of errors in the emission sources, improper partitioning between the speciated Hg in the emission sources and in the oxidized products (GOM and PBM), and improper treatment of other physical and chemical processes. Until these models can predict speciated Hg concentrations comparable to the monitored data, we have more

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faith in the dry deposition estimates using monitored concentration data than those from the large scale model outputs at the monitoring locations.

RC: 8) Section 3.4 “uncertainties” should be placed at the end of section 3. It seems to really break up the progression of the discussion.

AC: We originally did that. However, the uncertainties discussions (Section 3.4) were only for the dry deposition estimates (results from Section 3.3). Section 3.5 focuses on the dry deposition-litterfall comparison and Section 3.6 focuses on dry and wet deposition comparisons. More importantly, some information presented in Section 3.4 is needed to facilitate the discussion in Section 3.5. Thus, it is more logical to place the uncertainties discussion after the dry deposition estimation. We have changed the title of Section 3.4 to ‘Potential uncertainties in the estimated dry deposition’ in the revised paper to make it look more logical.

RC: 9) P. 2802 line 24 – 25. A summary or suggestion of what specifically drives the “relative contribution” differences would be nice here.

AC: A new paragraph has been added in the revised paper to discuss the relative magnitude of dry and wet deposition at AMNeT/MDN collocated sites. Discussions on the driver of the relative contribution have also been added here.

RC: Technical Suggestions:

AC: Technical corrections have all been addressed. The second paragraph of the Introduction has been rewritten. A thorough editing has also been done.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 2783, 2012.