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***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.**

Anonymous Referee #2

Received and published: 27 March 2012

Overall, I thought that this was a very good and well-organized manuscript. I believe that the authors provided a reasonable description of the previous work performed in this research field and they included references to a wide range of authors who have contributed to this field. The methodology section was well-written, though some improvements could be made as detailed below. While the presentation of results was a bit cumbersome at times given the significant quantitative results presented in the text, I was pleased to see that the authors provided information on the ranges of values obtained in the study, as this helped to put into perspective the large range of mercury dry deposition flux rates that are likely present in nature. Additionally, I was pleased to see a significant discussion of the uncertainties associated with the

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inferential modeling approach. This was underscored by the authors' discussion on how their estimates might vary due to the uncertainties in the input parameterizations.

I believe that this manuscript provides an important contribution to the understanding of science of mercury dry deposition in part because it addresses the sometimes ignored importance of gaseous elemental mercury (GEM) dry-deposition deposition to natural ecosystems. The assumption is often made that the dry deposition of gaseous elemental mercury is of lesser importance than that of gaseous oxidized mercury (GOM) and/or particulate bound mercury (PBM). The results of this modeling effort, and the information provided by litterfall measurements, suggest that modelers must seriously consider this process and not simply assume that GEM dry deposition is negligible due to its volatility and relative insolubility. Even small deposition rates could result in significant deposition due to the size of the GEM atmospheric pool. That said, the authors do a nice job of emphasizing differences in deposition and NET deposition of GEM. One issue that the authors might consider commenting upon (see below) is the bioavailability of GEM once taken up by vegetation and then deposited via litterfall.

While not the focus of this paper, I felt that the discussion on the relative importance of wet and dry deposition was too short. This is a very important issue in the attempt to obtain a better understanding of the environmental cycling of mercury. If the authors are going to include this section, they should expand discussion and provide a more complete and detailed comparison. I believe that this could be done relatively easily, without greatly expanding the length of the manuscript.

Finally, "the Conclusions and Recommendations" section was a bit short, but was to the point and provided an adequate review of the manuscript's main findings.

My main concerns with the manuscript are:

a) While the authors are correct in noting the importance of GEM to the total mercury dry deposition loading from the atmosphere, they neglect to discuss a second and related key issue: How bioavailable is the mercury taken up by the vegetation (presumed

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to be mostly GEM) and delivered to the surface as litterfall? While from a numerical perspective GEM dry deposition is important (typically greater than GOM or PBM), if it is bound to organic matter and not released, are not GOM and PBM then more important in the overall cycling. Based upon recent work that shows that biomass burning is capable of releasing significant amounts of GEM to the atmosphere, it is quite possible that over longer time scales, the organically bound GEM may eventually be released back into the atmosphere. I think some very brief (one paragraph) discussion of this issue would make the manuscript be more complete.

b) Throughout the paper, the authors refer to the results from the model as "estimates". The truth is, surrogate surface measurements and litterfall measurements are also estimates. By not acknowledging this fact, the authors assign greater level of certainty to the measurement results than might be appropriate.

While there are a few sections within the manuscript which should be strengthened, I feel that this manuscript is worthy of publication following minor revisions.

Specific Comments:

Page 2788, Line 14: I believe that it is more appropriate to spell out numbers less than ten when not indicating a measured value. In other words, "two-hourly" is probably more appropriate than "2-hourly". Such corrections would be needed in other places within the manuscript.

Page 2788, Line 14: The authors note that individual hourly GEM and two-hourly GOM and PBM values were used. Were two Tekran speciation systems used at each site, with one system for GEM and a separate system for GOM and PBM? When a single system is employed, the typical operational procedure with these systems is to have a one-hour sampling cycle (during which time GEM is quantified using five minute averages) and then a one-hour desorption cycle (during which time GOM and PBM are quantified). The result is to have what amounts to hourly average GEM, GOM and PBM data reported every other hour.

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Page 2789, Line 12: The authors note that net GEM deposition was obtained using inferentially modeled GEM deposition, minus GRAHM estimated re-emission plus natural emissions. Were the GRAHM modeled re-emission values consistent with those reported by other regional chemical models?

Page 2790, Line 3: What is the justification for the mesophyll resistance value assigned to GEM?

Page 2790, Line 10: The authors should provide a more detailed description of their selection of scaling factors (alpha and beta) used for GEM. The explanation that "adjustments were based upon GRAHM-simulated GEM concentrations" does not provide much information for the reader to determine if such adjustments are appropriate.

Page 2790, Line 25: Feddersen reference is listed as "to be submitted", but listing as in Line 25 would suggest that it is an accepted, peer reviewed publication. Perhaps it would be more appropriate to list this as "personal communication" or "unpublished data". I am not questioning the quality of the Feddersen work or the validity of its conclusions, but I am not sure that this a valid reference as listed.

Page 2791, Line 17: The authors should specify the Canadian meteorological model used, which I assume to be the Global Environmental Multiscale (GEM) model.

Page 2791, Line 21: The meteorological data used for the inferential model was obtained from a 15km x 15km resolution meteorological model. One might assume that the turbulence characteristics of the meteorological data were influenced by the description of the land use at this resolution. How might this have influenced results, when the inferential model only considered land use of the surrounding one kilometer?

Page 2793, Line 1: The authors note that: "Dry deposition of GOM+PBM, net dry deposition of GEM, and litterfall measurements were also marked on a wet deposition map for easy comparison." Which map are they referring to?

Page 2793, Line 6: Ambient concentrations are reported here and in Figure 2. What

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are the instrument detection limits for the GOM and PBM species and how might this impact the interpretation of overall results?

Page 2793, Line 8: It is not clear what the authors mean by geographical ratios?

Page 2796, Line 11: Lombard et al. (2011) reference is missing in the "References" section at end of manuscript.

Page 2796, Line 24: What do the authors mean by "concentration adjustment"?

Page 2799, Line 1: Also, the authors later note that Huang et al (2012) used the same inferential model with observed meteorology. For the present study, were comparisons made between dry deposition velocities derived from modeled meteorology and observed meteorology at any of the sites? Just curious?

Page 2799, Line 11: If the coarse fraction of PBM is assumed to be 30% of the total, why would the dry deposition of PBM need to be adjusted by a factor of two or more? Is this due to a the larger dry deposition velocity for coarse PM? The magnitude of adjustment seems extreme. Perhaps not, but a more detailed explanation would be helpful here.

Page 2802, Line 22: The wording used in this line is awkward. I would suggest: "dry and wet depositional loadings".

Page 2803, Line 6: An example of what was noted earlier. Namely, the authors only refer to inferentially modeled values as estimates. In reality, surrogate surface measurements are only estimates, as well.

Page 2805, Line 15: Engle reference out of alphabetical order.

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

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