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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Identification of mercury emissions from forest fires, lakes, regional and local sources using measurements in Milwaukee and an inverse method" *by* B. de Foy et al.

Anonymous Referee #1

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The manuscript by de Foy et al. uses a hybrid inverse modeling technique to assess the contribution of various mercury sources to observations in Milwaukee. The topic and tools the authors use to address this question are timely and novel. The inversion technique is first characterized using synthetic observations. When using actual measurements, the broad spatial pattern of emissions from coal sources is consistent with existing inventories. The impacts of forest fires are found to be highly variable, and the nearby lake sources seems to contribute more than initially expected. With only one measurement site and no a priori emissions included in the inversion, it is difficult to assess what these constraints mean on absolute emissions inventories, an issue which needs to be clarified. Further, it remains to be discussed what their approach

and findings provide in terms of constraints on previously recognized sources of uncertainty (i.e., local point sources). These issues and a few other questions are outlined below. Mostly I feel like additional explanation or more precise interpretation of results would address most of my concerns. The article is grammatically clear and otherwise well presented. I feel it will be suitable for publication in ACP after modest revision.

1 Major comments

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2 Major comment

- The results of this manuscript show very little contribution from local sources, likely as a consequence of excluding high measurements.
  - Their approach (47.27) to excluding outliers seems arbitrary. How is it justified? It would appear to be an important assumption, as it causes the inversion to neglect high-frequency peaks in the measurements and thus estimate low values for local sources.
  - Previous studies of Wisconsin discussed in the introduction indicated significant missing local point sources, which would not be represented in coarse Eulerian grid models. The relevance of the method and approach in the present work is thus not clear. What can the authors conclude about such missing sources and the ability of their modeling framework to address these? At the very least, somewhere in the discussion and conclusion their findings should be compared to those discussed earlier from the literature.

- 51.8-9: The authors state here that they do not use a priori emissions. This raises a few broad concerns.
  - The results of their inversion are presented in terms of scaling factors. If a priori emissions are not used, to what are these scaling factors referring? I have no idea what the meaning of a scaling factor of 0.2 or 4 means given that I don't have an a priori emission to multiply this by in order to infer an actual absolute emissions.
  - If the inverse model is started from zero a priori emissions, then the results obtained are not absolute estimates of emissions, rather they are estimates of the emissions which contribute to the measurements in Milwaukee. My understanding in this light is in sync with the authors description of the inversion results as "contributions" and "impacts" in many places. However, they also in many other places (such as comparison to other Hg inventories) refer to their findings as emissions, not contributions or impacts. Yet these results have no bearing on emissions within the modeling domain that lead to Hg concentrations in locations other than Milwuakee. Hence direct comparison to total inventories is misleading, as is concluding that particular emissions sources are fundamentally over or underestimated. I thus have an issue with all of the discussions and conclusions related to absolute emissions, as well as the wording of the title (i.e., replace identification with contribution or impacts).

3 Minor comments

- Throughout: suggest Eulerian grid model or chemical transport model rather than 'grid simulations'.

- 40: The overview of previous region studies in areas other than Wisconsin begins to feel a bit extensive and could be condensed.
- 41.9: Why is the focus on the GEM measurements alone?
- 42.15: What is meant by "eliminate all deposition effects" and why is this desirable? Is deposition not an important sink of mercury?
- 42.25: I don't understand what is meant by the sentence beginning "It is important . . . " In inverse modeling, it is important to have the number of variable parameters be matched by the number of observations plus regularization constraints. If there is one variable per grid cell, then I suppose this aspect could be translated into a statement regarding grid resolution, but that wasn't clearly the intention. The resolving power of an observing system is also not trivial to estimate; how the authors determined what is an optimal grid size or modeling domain (i.e., number of variable parameters) is not apparent.
- 46.10: It wasn't clear here how the inversion parameter vector and sensitivity matrix are constructed. I think after finishing the article I had a better idea, but it might be useful to more explicitly define things here. From presentation of the results it seems that there are variable parameters (scaling factors) for each of several biomass burning regions, one global background region, and many grid-specific sources. But here x_{CAMx} is defined as "scaling factors on the concentrations", so how these are used to invert for emissions scaling factors is not clear. Or are these used to rescale initial conditions? Then what are background values? So I guess I still don't understand what is x_{CAMx} vs the other elements of x.
- 48.17: The wording here is odd, as we don't know what is meant by "times" until the subsequent sentence.

- 49: This entire section is a bit strange. CFA wasn't really explained much. These results also don't seem to fit into the rest of the paper. They are not mentioned in discussion, conclusions or abstract. Overall, I would suggest skipping this analysis or incorporating it more completely into the paper.
- 50: The synthetic inversion test is a great way to assess the capabilities of an inverse model, and I'm glad the authors included this. However, there are a few aspects of their synthetic inversion that were not clear.
  - Like the real inversion, were these also performed using zero a priori?
  - Was the location with the X the only source?
  - This sentence: "Because the model simulates . . . " does not make sense. I don't think the model domain is the single reason. I think the inversion tends to overestimate emissions far away because there are no other observation locations to constrain such sources.
  - The last sentence of the first paragraph at first seemed at odds with the third to last sentence. It took my a while to see where 68% comes from (82 out of 121). Maybe this could be stated more clearly.
- White is used in Figures 3, 7, 8, 11, 13 but it isn't in the color bar.
- 52.17: It would be useful to also plot the time series of the observations that are actually being fit in the inversion.
- 54.23: The text would imply that Table 3 compares model values to those from the inventories shown in Fig 8, but it does not, it only contains model values, and only from fire emissions. Suggest expanding this table for better inter comparison of model with inventory (assuming this is even possible, given above discussion regarding contributions vs emissions).

- 55.4: would be expected
- 58.17: This assessment is a bit vague; can the authors be more quantitative in their summary?

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 12935, 2012.