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Interactive comment on "Mercury deposition in southern New Hampshire, 2006–2009" *by* M. A. S. Lombard et al.

Anonymous Referee #1

Received and published: 4 April 2011

General Comments: The paper is well-written and provides new data for a site in a region thought to be affected by Hg deposition. The paper provides useful information regarding the comparison of wet-only event data at this site to other wet-only data, plus the contribution of dry deposition. Perhaps the newest and most interesting finding is that there were different ratios of wet-only to RGM dry deposition in different seasons – data that are lacking in the literature. I suggest that the authors consider re-writing the paper, largely shortening sections that are less well-supported, and focusing on a couple of key findings/conclusions. The data for all analyses are useful and appear to have been collected in a manner that suggests good data quality, but perhaps some material could be moved to appendices or supporting information to focus the reader on the more novel findings. The data set is substantial and collection of both wet and dry data is rare, and very useful for others in the field.

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Specific Comments: Abstract – sentence 3 – suggests that learning more about wetonly deposition can provide understanding of atmospheric cycling and deposition. Rewrite to include dry and occult deposition, which are also crucial to understanding these broader processes.

In the abstract and throughout, avoid using the statistical term "trends". Use "patterns" when a statistical trend analysis has not been performed.

In the abstract and in Section 2, Line 15 – the authors refer to the site as "rural" but they also write that it is located in a heavily-developed "megalopolis". How is "rural" defined? It's unclear that this site should be thought of as rural within this broader urban context.

Introduction (and throughout) – Hg-P is written with a superscript "P", but I believe most other authors are using a subscript P. Please check and change if appropriate.

A reference to consider mentioning in the introduction is the 'state of the science' paper from the Mercury conference in 2006: Lindberg et al. 2007, Ambio Vol. 36, No. 1, February 2007.

P. 4571 Line 9: after "seasonal trends" add "in wet-only deposition" – or specify which are wet-only versus total or dry deposition. Many of the papers referred to are discussing wet-only deposition.

P. 4571, line 29 – Are there no more recent references with information for urban deposition? It seems that there has been more work in this area recently. The authors might scan the publications arising from the 2006 Mercury conference, at least, to check.

P. 4572, Line 19-21: I disagree that MDN sites collect integrated weekly samples – and this sample type ha not been defined. My understanding is that, like NADP collectors, the MDN buckets stay closed unless it is actually raining/snowing. Is this the case? If so, then the collections are wet-only event samples. The generic "integrated weekly" description implies, perhaps, that samplers were continuously collecting. Please pro-

vide more details so readers can determine whether your sampling strategy was reasonable to compare with MDN. If there were different strategies, then there needed to be side-by-side comparisons of the two types to validate your results.

P. 4572, Line 25 – the way this sentence is written, it appears that all (the sum of) events can equal 5-17%, which sounds like a small percentage. What the authors mean is that a single event can contribute between 5-17% – please re-phrase.

P. 4573, Line 9 – Is wet-only, event precipitation flux comparable to dry deposition measurements (which were not event based). I think the authors need to provide more description and rationale up front – in the Introduction or Methods – to define and provide evidence for comparability among the different sample types. Throughfall, total deposition, and occult deposition were not mentioned anywhere in the paper. Perhaps they could be included in an earlier discussion of sample types and what they represent.

P. 4574 – It seems that, if much of this methodology is consistent with MDN, the authors could omit virtually all of this section and refer the reader to MDN methods, simply pointing out in this section any deviations from those methods. Also, please cite an EPA method for cleaning and preservation of sample bottles and samples.

P. 4574, Line 14: Confirm that samples were preserved in their original collection container, as specified in EPA methods. Failure to do so before pouring off aliquots might result in loss of Hg adhered to container walls.

P. 4575, Line 13: Begin first sentence with "Wet-only".

P. 4577, throughout. Often chemical concentration and flux data are skewed left. Before performing statistical analyses, did the authors check data for the assumption of normality, and transform the data accordingly (usually, a log transform works)? If not, then parametric statistical results may not be appropriate. This could be a major issue in the presentation of results and conclusions based on those results.

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P. 4577, Line 9. Awkward wording – after "...differences", change to "among years for the same season..."

P. 4578, Line 9-18: There are many plausible reasons for elevated deposition at the TF site. Those listed here make sense, but the section is quite speculative and could be shortened – or, provide references from the literature that support some of the hypothesized mechanisms.

P. 4579 Line 5-10: Does the solar radiation investigation make sense, if the authors collected using MDN-like samplers that keep the sample bottle stored inside the collector (in the dark)? Why would solar radiation affect Hg in those bottles stored away from light? Consider omitting this section – it is again fairly speculative and not a key finding of the study.

P. 4579, Line 18-19 – The reported correlation between precipitation and deposition is not particularly meaningful – deposition includes precipitation in its calculation, and thus, they are not independent variables. In fact, many (non-Hg) studies have shown that we expect greater concentrations of chemicals such as SO4 in events with less precipitation, due to a washout effect later in rainstorms. The authors might peruse the literature to seek out such references and adjust their expectations and hypotheses to reflect these types of mechanisms. Also see page 4580, line 10+ - this explanation could account for some of the weak observed negative correlations. In general, the discussion of these various correlations is not strong – I suggest shortening it significantly.

P. 4581, Line 4+: The literature has made much of "ineffective scavenging" by snow in winter. This apparent phenomenon must be very carefully approached. At least one previous article on the topic misinterpreted an earlier publication and was subsequently widely cited. Also, it is crucial to define the form of Hg – and the authors have done a nice job doing so here, pointing out RGM specifically in the scavenging discussion. HgP could be affected by other mechanisms in winter. See Nelson et al. 2008 (Applied

Geochemistry 23, pp. 384-398), which also used data from the ME98 MDN site referenced in the current manuscript. Importantly, this current manuscript provides more support for the concept that there can be significant deposition in winter, despite the "ineffective scavenging" hypothesis having led to many scientists dismissing winter deposition as unimportant. Again, this is new information that provides crucial evidence in that debate.

P 4582 Line 20-24 – The information about RGM's seasonal pattern, and comparing it to the seasonal wet deposition pattern, is the key contribution of the paper in this reviewer's opinion. The work that supports it (comparing to other sites, to MDN) is useful and important to ensure data comparability, but if those sections were downscaled to focus on this important work, the paper would be stronger.

P. 4584, around Line 15: Here the authors have an opportunity to be more specific in their summary – note that RGM dry deposition was greatest in winter, rather than simply saying there was seasonality. Make the difference between wet-only and RGM clear.

Tables: Table 1: is there a way to use bold font or some other visual technique to group similar seasons? Much of the paper focuses on differences among seasons, but the table (though informative and necessary), makes is difficult to compare.

Note that the mean versus median suggests the data are indeed skewed left and might need to be transformed prior to statistical analyses that require the assumption of normality be met.

Table 2: can symbols (*, etc) be used to indicate significance at a certain p level, rather than listing all the p values?

Table 3: significant digits should be checked throughout the table. The table is useful, but another way to present the wet + dry information could be a stacked line graph. It would be visually appealing, but it would not be possible to include the ratio data – so

C1487

this is a "take it or leave it" recommendation.

Figures: Figure 1: Can the figure be made more information-rich by perhaps putting a callout at each site that lists annual concentration and deposition of Hg for the project years? This would allow a reader to see how the sites compare, on an annual scale, at a glance.

Figure 4: This graph demonstrates that log transforming the data would be much more appropriate and provide more statistically defensible results.

Figure 5: For display clarity, it would be useful to put the Y gridlines on the graph, perhaps in gray or as dotted lines. It can be difficult to read values from a log scale without grid lines.

Figure 6: This is an interesting way to present the ratio data. Perhaps this figure obviates the need for the ratio column in Table 3, which might then be better presented as a stacked line graph (or stacked bar).

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 4569, 2011.