

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

Anonymous Referee #2

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General Comments: In general I found the article needing support information on many of the assumptions drawn by the authors. Important discussion points need to be fleshed out. Less speculation is merited. Patterns are not trends, and the limited data presented in this manuscript only suggest potential patterns. The data and methods are provided without full documentation. This is a problem. The statistics used in the manuscript need to reflect the nature of the parameters investigated. Many of the variables are auto-correlated. Most of the data are likely skewed. Data transformation or the use of nonparametric tests may be more appropriate. The explanation of how dry deposition of mercury was calculated based on ambient air concentrations of various forms of mercury, and the assumptions (and short comings) associated with that calculation (based on H and the average concentration of RGM), needs to be much better discussed. The linchpin of this study is the estimation of RGM dry deposition. The

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reader needs to be fully informed of the difficulties in estimating dry deposition. The loose use of the term dry deposition should be avoided if possible. Comparison data from the literature appear to be cherry-picked (only 2 authors in the Table 3 comparison). A more scholarly approach/discussion on the subject of total mercury deposition is recommended to place this research in context. The authors are encouraged to provide more meaning to the important discussion points in the manuscript. Thank you for the opportunity to review this manuscript.

Specific Comments: It is suggested that the authors eliminate the first two sentences of the abstract and please rewrite the third sentence as a simple declarative statement.

Page 4570, line 9: change the word occurred to was measured.

Page 4570, line 11: Inter-annual differences of what? Please specify. Be careful of conclusions drawn in this abstract based on potential auto-correlations. Eliminate the last sentence of the abstract or develop that line of discussion better in the main body of the manuscript.

Page 4571, line 3-5: Please re-write to clarify statement.

Page 4571, line 12: Other explanations include differences in weather patterns/storms, oxidative potential of the atmosphere, proximity to various emission sources, etc, etc. Please include a more full discussion. The patterns of the various species of mercury in ambient air depend on many variables.

Page 4571, line 25: The sentence starting “Contradictory. . .” is confusing. Please re-write.

Page 4572, line 2: needs a reference.

Page 4572, line 5: This paragraph needs to provide better information. The Bow Power Plant, locate NW of the monitoring site has SCR for NO_x control. This results in substantial RGM emissions. Once the plant undergoes additional pollution controls the RGM emissions will be significantly reduced. The Bow plant is an important emission

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source that may influence deposition.

Page 4572, line 12: Please include in the main discussion comparative information from these MDN sites. Historical data provide windows to understanding current patterns.

Page 4572, line 20: Neither the NADP/NTN nor the MDN networks provide weekly integrated samples.

Page 4572, line 23: I believe that the Underhill site also measured (and still measures) ambient air mercury species. Contact Eric Miller. There are other similar sites located in NJ (operated by Rutgers). I also believe that additional sites exist in Canada and elsewhere in the United States.

Page 4573, line 1: Why did you pick the Maine sites only? Please explain.

Page 4573, line 24-25: How do you define an event. You use a 12 hour no precipitation period to define two separate events. Does a warm front followed by a cold front six hours later comprise one event? Are they two events with two separate meteorological characteristics? This needs to be clarified in the text since you may be sampling two separate events, but reporting only one event. [see Page 4575.]

Page 4573: How efficient were the ppt samplers during the winter snows? Sample bottle preparation appears to be different from those used by MDN. How likely are these differences to cause bias? What method was followed by bench chemists when analyzing precipitation for mercury? Was the protocol used by Frontier Geochemical used and if so were there differences in methods?

Page 4575: Sigler et al., 2009 indicate that RGM measurements began in the winter of 2006. The manuscript indicates that RGM measurements began in November 2006. Does the change in sampling time (120 minutes to 175 minutes) for Hg(p) introduce any bias to the data?

Page 4575: The NO_y and CO measurements could be used more extensively to help
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explain changes in boundary conditions and dry deposition of RGM. Please explain why these were not used.

Page 4575: Please compare the single precipitation event data with the two precipitation event data to determine how this may change your statistics.

Page 4575: You may wish to discuss the high deposition storms more. The MDN site at Casco Bay did not receive this amount of deposition during the week July 22- July 29. It recorded 0.27 ug/m² and a concentration of 7-8 ng/l. This would argue the potential for a very isolated precipitation event (convective storm?). You may wish to look at the wind vectors (wind roses) for this event (and similar events) and run the HYSPLIT back trajectories.

Page 4576-4577: Please let the reader know how you define your seasons. The variability in the recorded seasonal average concentration of mercury in precipitation among the MDN and TF sites (for the limited number of summers that were compared) do not indicate that the TF concentration for summer 2008 was particularly out of the ordinary. One should expect such inter-annual and seasonal differences. If you compared inter-annual season differences for precipitation, you might find a similar pattern (statistical differences). Please define "the northeastern United States". What geographic area does this cover? You may want to clarify what you mean by "numerous conditions affecting deposition".

Page 4578: Please avoid conjectures without supporting evidence such as is the case in "possibly due to...precipitation". The statement "First...indicate that more Hg is available..." needs to be more fully explained. It is not clear what you mean by more Hg is available. This may be re-written to clarify what you mean. Also, the sentence "Thus it is reasonable to hypothesize...due to anthropogenic emissions" needs additional clarification. What other factors could contribute to this? The reader is left with the feeling that these statements are supported by weak evidence. Please strengthen the lines of thought.

Page 4580: The section covering Anthropogenic Influences needs a lot of additional information. The data here should be used to help understand the mechanisms associated with diurnal and daily changes in RGM (and GEM) in the next section. Do you see patterns among the various measured mercury species and the other conservative and reactive gases that are measured? There is a wealth of information here which will help explain the patterns you see in the RGM (and GEM). Here, as is the case with precipitation and wet deposition, you should be careful running inappropriate statistics on variables that potentially auto-correlate. It seems logical that photochemistry plays a very important role in RGM and GEM concentration patterns (as is also the case with halogenated and other radicals). Additional discussion of this are warranted. Page 4581: There needs to be additional data/discussion presented about the interplay among the various variables that influence the estimated Vd. The derivation and use of this equation is the most important part of your argument about RGM deposition. Please provide a more robust discussion on its derivation, and the uncertainties associated with its calculation. Comparisons with other data sets (sa Miller et al., 2005 and Engel et al., 2010) need to be discussed within the context that these authors used different methods/components to estimate dry deposition of mercury. It is not clear that these comparisons are entirely appropriate. Miller et al. (2005) stated in their study that they have low confidence in their RGM deposition estimates. The importance of snow scavenging of mercury has yet to be determined. The research done at TF can provide very useful insights to this. Please expand on this.

Page 4582: The authors use the phrase "total Hg deposition flux". However, this paper only looks at wet deposition (as precipitation) and RGM dry deposition. Please avoid this since measurements of other types of mercury deposition are not covered in this paper. Also, the (estimated) ratios of wet mercury deposition (precipitation measured) to dry RGM deposition (estimated), may be plausible, but they need to be discussed in terms of the uncertainty of the variables. How certain is the Vd estimate? Please provide additional information.

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Page 4583-4584: Were the data used in Table 4 for Engles et al. (2010) study derived (calculated) by the authors, or were they taken from the article? This is unclear. Additional evidence needs to be provided to assist the reader in understanding the importance of the discussion about ratios (wet mercury deposition in precipitation [only] and dry deposition of RGM).

Page 4596: Figure 4. One would expect auto-correlation of these variables.

Page 4598 Figure 6 – There is no reference to this figure in the text.

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

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