

General Comments

This manuscript details the seasonal and annual variation in gaseous elemental mercury (Hg₀), reactive gaseous mercury (RGM), and particulate mercury (Hg_P) at 3 sites in New England. Initially I thought this was very new and innovative work. However, upon closer examination I am a little concerned that this may be a “data mining” expedition, presenting little if any new valid information. The “data mining” will likely result in another similar paper in the near future since the study is ongoing. There is a distinct lack of differentiation between this and their 2008 study. I am not sure that anywhere in the paper does it say “unlike in our 2008 study” (or anything similar), if there is I am sorry it just didn’t stand out. I am also concerned that the trends may not be as significant as the authors point out due to reasons discussed below. Unfortunately based on the criteria set forth by AC&P I am not sure this paper is publishable as a standalone article. All in all I think the relevant data from this article could be distilled and presented in one of the other manuscripts currently being published in this series (Part 2 or 3). If it is to remain as a standalone article major revisions are needed in order to prove that new, novel information is being presented here and the trends are real, and not just a data mining expedition. Also the presentation needs to be streamlined and the number of figures reduced to less than 13.

Specific Comments

- 1) There are very few differences between this paper and their 2008 paper. Also, if monitoring is continuing at these sites, what is the real point of this paper? Would it not be more advantageous to wait until more data is collected to actually identify trends at all sites?
- 2) The authors report a significant negative trend in Hg₀ concentrations that is below the detection limit of their analyzer (10 ppqv). This is potentially the most significant result of the study, but no indications of the statistical significance of this relationship are given. Therefore, I am not convinced that this trend is real. Much more explanation needs to be given on the possible factors (i.e. climatology, emission inventories, etc.) that may be influencing these trends. See Steffen, A., W. Schroeder, et al. (2005). "Mercury in the Arctic atmosphere: An analysis of eight years of measurements of GEM at Alert (Canada) and a comparison with observations at Amderma (Russia) and Kuujuarapik (Canada)." *Science of the Total Environment* 342(1-3): 185-198.
- 3) I have problems with the discussion of Hg₀ increase or decrease rates in “warm” and “cold” seasons (p. 32308 line 1). This discussion is confusing and vague. How were the rates calculated? Were they simply calculated with the single maximum and minimum concentrations then “scaled” to a daily rate? Were they calculated from single data points, daily means on single days, points from a moving average, etc.? The calculation of these rates was also not discussed in the 2008 paper. Depending on the values used these rates may be misleading. Does a single 5 minute point “high” or “low” point influence the “rate”? If daily means (or medians) are used how does this affect the rate. Before I can believe conclusions drawn from these “rates” I need to be convinced of exactly what they are representing. There are also no statistics presented to show whether the differences in these “rates” are actually statistically different by site. Significant further explanation is needed. Also if only two points in the calculation of this rate, the entire discussion of this rate is focusing on only two points in a very large

dataset collected each year at each site, therefore may not be an actual representation of the temporal variability at the sites.

- 4) Is there a precedent for removing mercury data at “CO below its 25th percentile?” How much data is removed by doing this? What happens if this data is left in? The decreasing “trend” in Hg⁰ concentrations is probably the most significant finding of the article but very little time is devoted to proving that this trend is real. No statistics, etc. It is completely buried and “glossed over.”

Technical Suggestions:

I suggest the authors give the manuscript a significant editing. Possibly having an outside person read the document. Here are my suggestions but there may be more needed:

- 1) P. 32302 line 2. Should be “reactive gaseous mercury” not reactive mercury.
- 2) p. 32302 line 5. What is meant by elevated? Elevated concentrations? Elevation?
- 3) P. 32308 line 1. The choice of the authors to refer to a “warm” and “cold” season is very ambiguous and confusing, since the “warm” and “cold” seasons are defined by mercury concentrations. I think. This needs more clearly explained.
- 4) P. 32308 line 27: Add “of” between “decline rate” and “0.6”
- 5) P. 32308 line 28: Add “rate of” between “minimum” and “0.1”
- 6) P. 32308 line 28: Add “in” between “resulting” and “a total”
- 7) P. 32309 line 20-21: How much of the data was “parsed out”? Were there significant differences between the entire dataset and parsed set? More discussion and justification is needed here.
- 8) P. 32310 line 6-7: Upper range of mixing ratios for where? Global? Regional?
- 9) P.32310: no statistics are presented on the differences among the sites. So discussions of the differences among sites and their causes may not be valid.
- 10) P. 32311 line 5: “Hg⁰” – bad form and does not fit with the style of the Seasonal and annual variations section
- 11) P. 32313 line 18: “RGM:” see above
- 12) P. 32314 line 13: “HgP:” see above
- 13) Figures 6 through 13 could be streamlined by only including the panels that are worthy of discussion.