Interactive comment on “Seasonal and diurnal variations of Hg° over New England” by H. Mao et al.

Anonymous Referee #2

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Long term measurements of ambient mercury concentrations are relatively scarce and this manuscript presents a novel analysis in the trends in ambient mercury concentrations and correlations with other atmospheric constituents that contribute to the community’s knowledge of mercury pollution. The data presented in this manuscript show strong warm season diel trend and season trends in the ambient concentrations. Revisions showing the variability, uncertainty, and significance in measurements are needed to better support the conclusions in this manuscript. Further analysis of the warm season diel trends is needed to support the conclusion that nighttime deposition is driving the nighttime depletion of ambient concentrations.

General Comments
More information should be provided supporting the assumption that the reactive gaseous mercury (RGM) fraction of the total gaseous mercury (TGM) concentration is negligible at the three sampling sites. This can be done by expanding on the RGM measurements. Was the RGM/TGM at a maximum during midday summer periods? What about cool season periods? Are the sample lines heated? Low RGM concentrations relative to TGM do not negate the accumulation of RGM on sample lines and possible subsequent evasion as gaseous elemental mercury (GEM).

The authors liberal use of the word ('')significant('') may be misleading. A measure of significance should be provided in cases where this wording is used. A measure of the variability and significance in correlations, diurnal and annual trends should be presented.

More data is needed to support the nighttime reductions in mercury and ozone concentrations by dry deposition. Is this supported by the diurnal variability in the wind direction? The Thompson Farm site is located near anthropogenic, natural terrestrial and oceanic sources/sinks of elemental mercury. Presumably a shift in the wind speed would greatly change the concentrations as air would be advected over sources or sinks of differing strengths. Further analysis is needed to show that the observed reduction in nighttime concentrations is from dry deposition and not seasonal shifts in the diurnal wind direction. Perhaps the lack of a nighttime drop and the muted nighttime drop in GEM concentrations at the Appledore Island and Pac Monadock are due to those sites relative homogeneity in surrounding land cover type.

The anomalies of GEM versus CHBr$_3$ are interesting but only scatter plots are presented. The diel variability of CHBr$_3$ may be useful in interpreting the data collected at Thompson Farm and Appledore Island. Given the discussion on oceanic influences on the mercury concentrations the relationship between ambient concentrations and wind direction at the Thompson Farm and Appledore Island site should be quantified with a scatter plot or concentration rose.
Changes in seasonal synoptic weather patterns and the seasonal variability in the boundary layer mixing depth should be discussed in the interpretations of the changes in the GEM mixing ratios. Could some of the changes in the ambient GEM concentrations arise from changes in the boundary layer depth or changes in the source of air parcels advected to the study sites.

Specific comments

1. Page 17215 lines 21-23: Given the reported annual and seasonal variability in this manuscript, the time of year of the measurements preferably the general characteristics of the measurement site, i.e. coastal, taiga, etc., by Poissant et al (2004) should be included.

2. Page 17216 lines 18-19: CMAQ Hg models the dry deposition of GEM using a resistance model that is temporally and spatially dynamic, as reported in Lin et al (2006). Defining a seasonal and domain mean as the deposition velocity of GEM is misleading. A range of values, preferably for forested or coastal land cover types, would be more applicable to the comparisons presented in this manuscript.

3. Page 17218 lines 12-13: Please specify (')standard additions of Hg0 ('). Was this done using the internal permeation source of the Tekran? Were these spiked air samples? What were the concentration or loading of these (')standard additions(')?

4. Page 17220 lines 10-11: A measure of the significance of the (')significant almost daily downward propagation in its mixing ratio(') should be reported.

5. Page 17221 line 6: A measure of the significance of the summer trends in the PM site GEM mixing ratios should be reported.

6. Page 17221 lines 11-13: (')considerable interannual variability(') should be quantified. How did the interannual variability compare to the seasonal variability? Are the seasonal values from different years significantly different?

7. Section 5: additional information is required to support the hypothesis that the night-
time depletion of GEM at TF is a result of dry deposition. Section 6 acknowledges that TF is located near a multitude of natural and anthropogenic sources and the analysis of GEM verses CHBr₃ implicitly implies a predictable diurnal shift in wind directions. Estimates of scatter in mean values and significance in relationships in the measurement data should be presented in this section.

8. Page 17225 line 14: Please quantify the ('')significant day-to-day variation('')

9. Page 17227 second to last paragraph in section 5: Nighttime deposition velocities are typically much lower than daytime values. Are there published estimates of nighttime deposition velocities as high as the 0.17 cm s⁻¹? This is larger in magnitude than the peak mid day deposition velocity reported in the modeling study of Lin et al. (2006).

10. Page 17228 line 9: Please quantify or change the wording in ('')deposition processes for Hg⁰ are in fact very significant(''). ('')very('') can be eliminated from the sentence as it is not quantitative and adds little value.

11. Page 17229 second full paragraph: Was there no measurement of wind speed and direction? A wind direction filter would be a much better indicator of onshore transport.

12. Page 17230 lines 16-17: Please quantify the ('')significantly lower Hg⁰ levels('')

13. Page 17231 lines 10-12: Weekly scale variances in the concentrations may be indicative of a dependence on synoptic weather patterns.

14. Page 1732 line 13: Please quantify the ('')significant decreases in the maximum mixing ratios('')

15. Page 1732 lines 25-26: Please quantify or provide a citation for ('')near normal conditions in 2004 and 2005 to significantly warmer than normal conditions in 2006 and 2007('')

16. Page 17236 line 8: Please quantify ('')overall significantly lower Hg levels and steeper decreasing trend('')
17. Figure 2: Error bars should be added to the mean quantities in the figures.

18. Figures 4 through 6 and 11: A p value should be added to the figures or in the text describing the figures.