Review of Mao, et al. "Speciated Mercury at Marine, Coastal and Inland Sites in New England: Part II. Relationships with Atmospheric Physical Parameters" in *ACPD*, for consideration of publication in *ACP*.

General Comments:

This article describes an exhaustive suite of speciated atmospheric mercury measurements at 3 different sites in New England. The authors analyze the observations of gaseous elemental mercury (Hg^o), reactive gaseous mercury (RGM) and particulate mercury (Hg^P) with respect to other climate variables like temperature, wind direction, humidity, precipitation, and solar radiation. The observations were conducted for an extended period from 2006-2009 at three sites of differing characteristics: marine (Appledore Island), coastal (Thompson Farm) and elevated inland (Pac Moadnock). Comparisons are made between speciated mercury observations at the 3 different sites in how they correlate with the physical variables under study. Several trends are demonstrated by the analyses. The analyses give insight into the seasonality and fluctuations in speciated mercury abundances in comparison to temperature, wind direction, wind speed, relative humidity, precipitation and solar radiation.

The trends and analysis shown in this article represent a significant contribution to the literature with respect to ambient speciated mercury observations for different physical systems. The correlations between RGM and wind speed, temperature, solar radiation, precipitation and relative humidity as a group help to explore mechanisms for production and losses. The observed trends in Hg^P with respect to temperature and precipitation also allude to mechanisms for production or losses and seemingly resilience of this species to persist in the atmosphere. The correlations of Hg^o with temperature and relative humidity begin to constrain the understanding of gaseous elemental mercury abundances, emissions and losses. The differences between the three sites at times help to distinguish variable environments as to their role in mercury partitioning and abundances. However, the total amount of information in this paper is exhaustive and at times the concomitant presentation and interpretation of the data can make the conclusions less clear. This paper would be strengthened by revisions to help streamline the analysis for the reader to better glean from the article the reasoning behind the analyses and give context to the assertions of the authors as to their explanations to rationalize the trends observed. I recommend this article for be considered for publication after revision based on the following:

Specific Comments:

1) Format. The article has 4 main sections which are 1. Introduction, 2. Measurement and Approach, 3. Relationships between Hg^o/RGM/Hg^P and meteorological variables and 4. Summary. Section 3 includes both descriptions of the observed relationships with 3.1 wind, 3.2 solar radiation 3.3 temperature, and 3.4 relative humidity, but also includes a sentence or two about what each trend or relationship 'implies'. The overlap of data interpretation with presentation is at times disjointed or comes across as unsubstantiated (particularly with respect to implications for production and loss mechanisms). I suggest a 4^{th} section (Discussion) that brings together the interpretation of the trends separated in terms of species (Hg^o, RGM and Hg^P) that both contextualizes the observations in terms of how this article explains, shows, elucidates or uncovers relationships not observed

before (with comparison with other literature) and how the observations relate to the atmospheric chemistry of mercury transport, production and/or losses. Some discussion on the observed trends is warranted because it sounds, at times, that the interpretation of individual trends is contradictory (for example, the trend observed in the difference in abundance of RGM versus wind speed is used to explain that transport can dominate the observations of RGM at Thompson Farm (p. 10) but in other analyses transport is not mentioned in relating to diurnal profiles. The complication is that multiple variables could possibly be lumped into the observations and therefore an interpretation of production or loss versus transport may not so easily be distinguished).

2. Interpretation of the results. This goes along with comment 1, where the paper would be stronger if general conclusions are made with respect to how the difference in site location adds to our understanding of mercury chemistry. Specifically, how do the differences in seasonal abundances and site locations help to isolate production, loss or transport mechanisms? This interpretation would be most useful if some of the basics were stated – that RGM is highly susceptible to removal by rainwater, - give the approximate lifetime of RGM in the atmosphere (if it is known, or understood) – what are the different emissions terms for Hg^o at various locations (marine vs inland)? How do the authors distinguish between scavenging efficiency of snow and evasion of mercury during the process of fusion? What is the rationale for the choice of RGM being influenced by production (p16), transport (p21,11) losses (p13) but never dilution from changes in boundary layer height? Can rainfall affect the abundance of oxidants that produce RGM (different from the authors' interpretation on p 21 on residual layer transport)? Can lightning during a heavy storm (p23) influence oxidative capacity of the atmosphere? Each mechanism is usually given or implied in different locations in the paper, but a connection between the physical variables and what can be learned from them would be most helpful early on in the paper so that the leap to interpretation of the data is seamless.

3. Clarity of notation. The abbreviations for the three sites can become burdensome when the reader must continuously remember which is a coastal, marine or inland site. Because the local environment is important in interpreting the results, I would suggest referring to the sites by their environment (coastal, marine and inland) instead of the abbreviations, so as to make the significance of the site clearer.

4. Figure clarity. The information contained within each of the figures could be noted with more specificity so as to highlight the meaning and purpose of the figure or table quickly. For example the notation of Figures 16 and 17 should include the same notation as figures 15 and 18 that includes 'days without rain' 'days with nighttime rain' and 'days with daytime rain'. Also Figures 4 and 5 could include headings like 'coastal' 'marine' and 'inland' sites so that the reader would instantly associate the difference in observations with the different sites without having to read the caption and interpret the site location as to what the difference is with respect to the local environment. There are a lot of references to data/graphs not shown. Because of the wealth of information in this paper, I think it is unnecessary to explain that those relationships are not shown.

Technical Corrections:
p.3 The authors refer to the acronym TGM before defining it.
p.4 end parentheses after Hg^o needs to not be in the superscript
p.4-5. The sentence "Brooks et al (2010) found that peaks of Hg^p … showed distinct and consistent relationships with the average planetary boundary layer dynamics enhanced by a shallow nocturnal boundary layer …" is confusing. Were the peaks enhanced by the shallow nocturnal boundary layer, or were the dynamics enhanced? (it appears that it should be the former, but the sentence reads as though it were the latter)
p. 5 The first sentence of the second paragraph is misleading and makes the paragraph awkward. I would suggest starting with something like, "While long-term studies of TGM have been investigated …". The last 2 sentences are also somewhat troublesome in that they make it harder to discern how different Part I and Part II are from each other. I recommend a more parallel structure for describing the two different papers (both describe the differences at locations with distinct geographical characteristics, but Part I focuses on difference in abundances with respect to site location and diurnal and interannual time scales).
p. 6 last paragraph: Describe the instrument as Tekran 1135 for measuring Hg^p instead of "1135 Hg^p Tekran"
p 7. the sentence that starts with "There were a significant fraction…" is awkward. I would suggest starting with "When there were…" and reformat appropriately
p. 7 first sentence of section 3.1. "One of ***the most*** significant…"
p. 11: note please refer to Fain et al (2009) "High levels of reactive gaseous mercury observed at a high elevation research laboratory in the Rocky Mountains" **ACP** 8, 8049-8060 and references therein to show other evidence of anomalous RGM observations at remote sites.
p. 15 and afterwards use 1) or 1. but not both
p 16 last paragraph "At AI there was only one month ***of*** data…"
p 17 why is (Hg^p) in parentheses in the second paragraph?
p 18 1st paragraph: the data points did not suggest a trend with the time of day (instead of preference, the data can't prefer anything)
p 18 3rd paragraph. The first sentence is very awkward
p 19. last paragraph: This paragraph is very difficult to determine the reason behind the authors' description, so it would be useful to give context to difference of this observation with respect to other sites (which is implied in the interpretation of the results on p 20) – why do the authors comment only on observations about 2 ppqv? Is it because this site had an anomalously high number of points above 2ppqv?
p. 20 When the authors refer to Table 1, there is some discussion of how many points were above the LOD but there is nothing in the table to describe that. Either give a number (how many points were below LOD versus the total points given in Table 1) or include something about observations below LOD in Table 1. Because the authors have data that is above the LOD for days with precipitation, it would be helpful to understand how much of the data is like that (only 10%? 50%?)
p. 20 The authors' interpretation of the trends in RGM with respect to precipitation seems over interpreted. The only significant difference is for winter, whereas the ratio of RGM in dry weather vs. wet weather is something on the order of 38-2 for spring and summer

and only reaches a lower value (2-1) for winter. Spring has higher general abundances during dry days and the wet days seem to correspond to that overall abundance.
p. 22 Last paragraph: The start to the paragraph is very awkward. The paragraph is about integration of RGM over a 6-hour interval. The first sentence should address that first and justify the reason for doing it that way second.
p. 23 remove the hyphen after night
p. 24 2st paragraph: Start the paragraph with the explanation that the seasonal observations may reflect the sensitivity of the instrumentation to particle size.
P 25. I am surprised that if transport was the primary source of RGM to the location that then other analyses in the paper tend to describe interpreted production and loss terms (instead of dynamic fluctuations in boundary layer height, for example)
p. 25 – why do the authors not mention the relationship between Hg^o and relative humidity?

Tables:
Table 1: The text of the table should include RGM and the units of the data (the authors could give it a mathematical notation of RGM (ppqv) above $\bar{x}$ ($\pm 1\sigma$))
Table 2: The data for RGM are an average? Out of how many points? Units should be given in the table. The difference of the shaded and unshaded regions should be given as a header in the table (Short Rainfall Events / Sustained (>x hours) Rainfall Events)
Table 3: Give RGM and units as labels in the table
Table 4: Give Hg^p and units in the body of the table

Figures:
Figure 1: Figure 1a) appears to be missing the data for wind speed and wind direction that is given in Figure 1b). It may be easier to distinguish some of the data if a variable or two was given as a line.
Use the same order in the legend and same symbol types for the variables between Figures 1a and 1b.
Figure 3. (see comments above). Coastal/Inland sites
Figure 4. (see comments above). The 90 seems to be cut off in the wind roses.
Figure 5. (see comments above). The 90 seems to be cut off in the wind roses.
Figure 6. The data in figure 6 b) above the 90^{th} percentile looks to even to be real. Is it?
Figure 10. The relationship between Hg^p and temperature seems to be wholly captured by Figure 10 a). Are the other 3 sub-figures necessary?
Figure 15. Is the wintertime data really sorted with respect to rain? Or precipitation (snowfall)?
Figures 16 and 17: (see comments above)