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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Gaseous elemental and reactive mercury in southern New Hampshire" *by* J. M. Sigler et al.

J. M. Sigler et al.

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Response to anonymous reviewer #2

We thank the anonymous reviewer for agreeing to read our paper and taking the time to provide commentary and criticism. The reviewer raises a couple of important issues, one, our use of wind roses in section 5.3, and two, some general comments on the need for focused studies of trace gases/marine halogens at our field sites and further discussion and presentation of observations of marine biogenics, seasonality of halogen compounds and their relation to Hg speciation.

We agree with the reviewer that trajectory modeling and Potential Source Contribution Function (PSCF) would be an effective means for determining source areas of elemental Hg. We have used these methods (see Sigler and Lee, 2006, which examined

interannual variation (or lack thereof) to determine source areas of TGM in the north-east using measurements at a Connecticut field site). However, this paper serves as an overview of the extensive measurements. More technical papers will follow in the future to tackle more specific scientific topics using these measurements. For instance, the reviewer’s suggestion will make a focused task to identify/quantify regional sources and sinks of Hg° and RGM. Therefore we felt that a detailed PSCF analysis was somewhat beyond the scope of our paper.

Our intention in using wind roses was primarily to show that there is a directional component to observed high RGM and SO2 events at Thompson Farm, and some elevated RGM observations at Pac Monadnock. Minor variations in these directional dependencies strongly indicate the importance of local combustion sources, particularly the Merrimack power station and other sources in the Manchester, NH area, which are within ˜50 km of the sites and are known to contribute to biogenic Hg “hotspots” (Evers et al., 2007). Assuming average wind speed near the surface is ˜5 m s-1 (on the higher end, Mao and Talbot (2004)), it would take at most ˜ 3 hours for pollutants to travel from the hotspots to our monitoring sites. Even using the 3-hour and 48 km EDAS datasets, which has the finest time and spatial resolution amongst all choices of input datasets, to drive HYSPLIT to calculate trajectories, it is hard to obtain information on our desired temporal and spatial scales. In the case of Appledore Island, the directional dependence and the nearby presence of Boston is fairly straightforward. Given the locality of these sources and the regional character of these sites, as well as the short lifetime of RGM in the atmosphere, we feel that the wind roses in Figures 9-11 are appropriate for what we intend to show here, are important to our study and we would prefer to include them in the paper. This is similar to the wind rose analysis used for RGM measurements in the Midwest U.S. by Yatevelli et al. (2006) and Manolopoulos et al. (2007). We also note that previous studies at our Thompson Farm site by Mao and Talbot (2004a/b) used wind sector analysis for other pollutants (O3 and CO) at several AIRMAP sites and found similar results; i.e., that southerly and southwesterly flow transports pollutant-rich air to NH from southern

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New England and that wind direction measurements at TF are well-representative of the region. In light of the reviewer comments, we will add this point to our revised manuscript. In any case, we feel that Figures 9-11 are appropriate for what we wish to communicate in this overview paper.

The reviewer points out that trajectory modeling would be beneficial for areas with complex terrain and in studies of more distant sources. This point is well-taken. We will revise our text in section 5.3 to include this point, particularly given that we suspect there may be influence from areas that are greater than 50 km distant (ie, Albany, urban areas of Massachusetts and Connecticut, etc).

We also note that although they are not included in the paper, HYSPLIT back trajectory simulations (48 hours) were performed for the many of the high RGM events shown in Figure 11. These basically re-affirmed the directional bias of these pollution events, which is one reason we opted not to delve into trajectories in this manuscript.

The reviewer noted that it would be useful if a more detailed study of trace gases, including marine halogens, could be conducted at all three sites for a more prolonged period. In fact, we have observations of many key pollutants including O3, CO, NO, NOy, SO2 and other trace gases at these and several other AIRMAP sites in NH dating back in some cases to 2001 (these data are available in archive form and in near real-time at airmap.unh.edu). Detailed studies of O3, CO, aerosols, elemental Hg, VOCs and various hydrocarbons and halogen species at various field sites have been conducted (see airmap.unh.edu/publications). Our studies of marine halogens are currently limited to Thompson Farm and Appledore Island. This research is new and on-going, and two recent papers (Zhou et al., 2005, Zhou et al., 2008) make initial headway in characterizing the levels of and controls on CHBr3 and CH2Br2 at these sites, and some initial interpretation of seasonal variation.

The reviewer also raised the point that the paper would benefit from a more clear discussion of seasonality in halogen compounds (and possibly in accord with changes in

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sea surface temperature and algal biomass), if available, and the possible influence on Hg speciation. A “spring bloom” in algae is observed in the Gulf of Maine, typically in late March. Thus far, we have seen no influence on Hg speciation based on our limited data. One reason for on-going, year-round Hg observations at Appledore is to capture any possible link between Hg speciation in the marine boundary layer and marine biogenics. As noted above, Zhou et al. (2005) provided some initial information on seasonality of various marine halogen compounds at TF, including CHBr3 and CH2Br2. Those results are too limited to formally establish a relationship with Hg speciation at TF. However, the 2007 time series in Figure 2a indicate a significant drop in Hg° at TF during summer and an increase in the colder months. This pattern is repeatable from year to year at TF (see Mao et al., 2008) and we speculate that seasonal variation in halogen chemistry may play a role (for example, higher levels of CHBr3 at TF during summer, accompanied by stronger photochemical dissociation, could lead to lower Hg0 levels). We propose to add a statement to this effect in section 3.1 in our revised manuscript. But we stress that at this point, this is speculation.

Technical correction: p17768:methods: “denuders are replaced a 10-day basis at TF and PM, and more often on a 2-3 week basis at AI…” As the reviewer notes, we should replace “more often” with “typically” to make this statement more clear.

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