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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Concentration-weighted trajectory approach to identifying sources of Speciated Atmospheric Mercury at an Urban Coastal Site in Nova Scotia, Canada" *by* I. Cheng et al.

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We greatly appreciate the reviewer for providing valuable comments which have helped us to improve the paper. Our point-by-point responses are detailed below.

Original comment: Positives: 1) There are some good aspects of this paper, including a nice literature survey and statistical methods using back trajectories. 2) The discussion and explanation of CWTs and how they differ from PSCF is very good. 3) The paper is well written and easy to follow. It is well referenced. 4) The Hg measurements are

first rate. There are not many reports of speciated Hg data in maritime Canada, an important region being that it is downwind of point sources in Canada and the U.S.

Response: Thank you for your feedback on the strengths of the paper. We hope we have addressed the following weaker points in the revised paper.

Original comment: Negatives: 1) The fact that the trajectory model showed there were speciated Hg sources in the Atlantic Ocean and elsewhere calls into question the accuracy and efficacy of using this kind of trajectory approach for speciated Hg.

Response: GEM is emitted from oceans due to evasion from the ocean surface. Oceans make up the largest natural source of Hg globally and includes primary emissions and emissions of previously-deposited Hg originating from natural and anthropogenic sources. GOM production over the ocean is also possible by in situ GEM oxidation involving oxidants, e.g. reactive Br (since the ocean is a major source). This supports the source areas in the Atlantic Ocean for GEM and GOM in the CWT plots (Fig. 2 and 3 in the ACPD paper). For PBM (<2.5μm), there are no emission sources from the ocean, which is consistent with the lower CWTs for PBM from the ocean (more yellow areas in Fig. 4 ACPD paper).

As mentioned in the Introduction, previous studies had applied similar trajectory approach to identify source areas contributing to speciated atmospheric Hg, but few studies had tried to assess model accuracy. We explained how comparing the CWT modeled source areas with mercury emissions inventory is not the best approach to verify accuracy because of uncertainties and limitations with the current mercury emissions inventory. CWT source areas were compared (using statistical analysis) with known industrial Hg emissions because this data is updated annually and precise locations were available, unlike Hg emissions data for non-point sources. Given the uncertainties on non-point Hg sources, the model needs to be accurate about non-source areas; our results showed there were minimal industrial Hg emissions in areas with low CWTs. This was verifiable through a method like CWT because other similar trajectory methods are

applying a threshold concentration (i.e., examining the model results only above a certain concentration instead of all concentrations). In the revised paper (section 3.7), a more detailed discussion on the model uncertainties and limitations have been added.

Original comment: 2) Because of the uncertainty of the output results (the CWT plots) can the authors say whether speciated Hg originates globally, regionally, or locally? I don't think they can. Example of conjecture is lines 292-296. Just because the trajectories gave a result, should we then conclude that these areas are sources of Hg? It seems too simplistic to me.

Response: There are sources of uncertainty from back trajectory modeling and mercury emissions inventory. There is more emphasis on the former in the revised paper (section 3.7). The CWT model in the study was intended to identify regional source areas (not global or local); therefore, a geographical domain was chosen based on locations of regional Hg point sources as mentioned in the ACPD paper on p. 4190 lines 5-7. Using a regional geographical domain allows us to use smaller grid sizes (0.5o x 0.5o) and more grid cells (2400), so that we can be more precise about the source areas. Had a global domain been chosen, there would be more uncertainties because GOM and PBM deposits close to their sources of emission and only coarser grid sizes would have been computationally feasible but a potential drawback could be less precise modeling of source areas. We did not use CWT results to identify local sources because the meteorological data resolution of 40 km for the back trajectory model was too coarse, so we conducted the analysis with local wind direction data (CPF method in section 3.6 of ACPD paper).

Even though the CWT model gave a result, it does require interpreting the results based on available industrial Hg emissions data (ACPD paper section 3.3). Section 3.4 applied statistical analysis to compare modeled source areas with known industrial Hg emission from each grid cell. Source areas were also compared with non-point sources reported in previous literature findings and 2008 National Emissions Inventory (ACPD paper section 3.5). In the revised paper (section 3.6), we compared CWT

results over the ocean with those from a recent modeling study. We do not consider the way the sources were interpreted as conjecture.

The results might have the impression that it is simplistic if it was only based on trajectories. The CWT model is a function of two factors: residence time of back trajectories and Hg concentration (ACPD paper on p. 4189 lines 9-11); therefore, a large CWT (potential source area) resulted from having a long residence time in a grid cell accompanied by high Hg concentration. The addition of a concentration weighting factor in the model makes it more complex than only a trajectory model.

Original comment: 3) What if a trajectory endpoint segment resided in a grid cell at 5 km altitude? Would it still be counted in the CWT? This seems like a real problem because how could an air mass at such a high altitude pick up pollutants emitted at the surface? Can you filter the CWTs to include only those that are < 1 km altitude for example? Section 3.4 is kind of interesting and suggests the trajectories could be telling us something. Section 3.5 seems like conjecture and may be superfluous Section 3.6 has a lot of conjecture as well.

Response: There were no trajectory segment endpoints at a 5 km altitude. We verified that all the trajectory segment endpoints were at an altitude below the model ground level of 1.6 km. It is not appropriate to conclude that the trajectory segment endpoints below a certain altitude (e.g., < 1 km) were emitted at the surface because there are uncertainties about vertical mixing and turbulence in the boundary layer. In the revised paper (section 3.7), a more detailed discussion on the model uncertainties and limitations have been added. Vertical mixing occurs throughout the day and was shown to affect diurnal patterns of atmospheric Hg (Sigler et al., 2009; Nair et al., 2013).

Section 3.4 of the ACPD paper applied statistical analysis to compare modeled source areas with known industrial Hg emission from each grid cell. Source areas were also compared with non-point sources reported in previous studies and 2008 National Emissions Inventory (section 3.5 and 3.6 in ACPD paper). But we could not conduct statis-

tical analysis between the modeled source areas and non-point sources of Hg due to uncertainties and limitations with Hg emission from non-point sources (see section 3.7 in revised paper). In the revised paper (section 3.6), we compared CWT results over the ocean with those from a recent modeling study.

Sigler, J.M., Mao, H., and Talbot, R.: Gaseous elemental and reactive mercury in Southern New Hampshire. Atmos. Chem. Phys., 9, 1929-1942, 2009.

Nair, U. S., Wu, Y., Walters, J., Jansen, J., and Edgerton, E. S.: Diurnal and seasonal variation of mercury species at coastal-suburban, urban, and rural sites in the southeastern United States. Atmos. Environ., 47, 499–508, doi:10.1016/j.atmosenv.2011.09.056, 2012.

Original comment: Overall impression: 1) Nice to have more speciated Hg data in the literature, but I'm not sure the authors have satisfied their objectives which were: "identify regional source areas contributing to speciated atmospheric Hg measurements at an urban site in Dartmouth, Nova Scotia, Canada" and "address potential local sources of Hg"

Response: The objectives of the study were to identify potential regional and local sources contributing to speciated atmospheric Hg measurements at an urban site using the CWT model. The CWT model and similar trajectory statistical models have been used in the literature to model advection of pollution and examine potential source-receptor relationships. The revised paper has emphasized that these are potential sources that may be affecting the site and there are uncertainties and limitations with the CWT model in achieving the objectives of the study (section 3.7 in the revised paper). We also modified the title of the revised paper by adding the word "potential" to better reflect its content.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 4183, 2013.