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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Quantitative assessment of upstream source influences on total gaseous mercury observations in Ontario, Canada" *by* D. Wen et al.

**Anonymous Referee #1**

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General Comments:

A highly-modified version of the Stochastic Time-Inverted Lagrangian Transport (STILT) model has been used to simulate atmospheric transport of total gaseous mercury (TGM). STILT was originally developed to study atmospheric transport of inert tracers. The authors have modified STILT to account for wet and dry deposition and elevated releases of TGM. However, TGM is composed of elemental mercury vapor (Hg0) and various gaseous mercury compounds collectively defined as reactive gaseous mercury (RGM). These two modeling species for gaseous mercury have widely varying atmospheric properties. The authors have treated them separately in



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their estimation of the effects of wet and dry deposition. However, there appears to be no treatment for the chemical conversion of one form to the other. Chemical transformation of mercury between Hg0 which is slowly deposited and various oxidized forms that are rapidly deposited is poorly understood and remains a primary source of error and uncertainty for all atmospheric mercury models. I fear this failure to address chemical transformation detracts greatly from the information to be gained by this work.

The modified STILT model has been applied here to estimate the sources of TGM for three locations in southern Ontario. Four 10-day time periods are analyzed, each having been chosen for season coverage (winter, spring, summer, autumn), but also to cover periods where another model (CMAQ) proved inaccurate. The authors suggest that the poor accuracy from CMAQ was mostly due to errors in simulated near-field transport that are inherent in all Eulerian-type models and that the Lagrangian transport analysis of STILT provides improved accuracy in this regard. If near-field sources are a major influence on TGM concentrations, the obvious remedy for Eulerian-type modeling is to use a finer horizontal grid. If I understand correctly, both STILT and CMAQ used the same emissions and meteorological data resolved at 36 km on the horizontal scale. In this case, the only real advantage STILT would have over CMAQ is a precise location for the receptor site. The errors in transport simulation due to horizontal smoothing of the wind data would be similar for both models, would they not? The same is true for errors related to the definition of emission sources. A common argument for Lagrangian transport modeling over Eulerian techniques is that it eliminates numerical diffusion. The use of finer grid sizes and non-dispersive advection schemes can well address this difficulty in Eulerian modeling.

The authors' results do show superior agreement to observed TGM air concentrations with STILT. However, as the manuscript explains, the simulation periods were chosen to include instances where CMAQ exhibited poor performance in reproducing observed TGM air concentrations. As mentioned above, STILT does not address chemistry. CMAQ simulates chemical transformations of mercury between Hg0 and RGM and



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physical transformations of mercury between gaseous and aerosol forms. CMAQ may have provided an inaccurate simulation of these mercury transformations during the test periods selected. I do not believe the evidence provided here should be used to conclude that Lagrangian trajectory modeling as applied in STILT has a fundamental advantage over Eulerian-type modeling in the assessment of source influences on TGM air concentrations. This sort of conclusion could only be drawn by applying these models over a long period of time that is not chosen to put either model at a disadvantage.

I am also concerned about the modification made to STILT to account for deposition. Once deposition comes into play, how does that affect the theoretical demonstration of time-reversibility in Thomson (1987)? For mercury, wet deposition is brought about by more than just sub-cloud scavenging by precipitation. In addition to water solubility, reduction-oxidation chemistry in cloud droplets is also important. For dry deposition, especially that of RGM, definition of the surface layer depth is very important. Rapid dry deposition of RGM to the surface leads to rapid depletion when the surface layer is shallow. It appears that a constant surface layer depth of approximately 75 meters was used in the STILT modeling. If so, I would expect the dry deposition flux for RGM to be overestimated for nighttime conditions when the true surface layer depth is less than 75 meters and underestimated in daytime where convective mixing extends much higher than 75 meters.

Besides the shortcomings mentioned above, the ecological and human health risks from mercury contamination are very weakly associated with air concentrations of TGM. It is the atmospheric deposition of mercury to sensitive aquatic systems that is important. Most of this deposition occurs specifically from RGM air concentrations. TGM is much easier to measure than its rather small and elusive RGM fraction. For this reason, TGM measurements are much more available. However, TGM air concentrations, whether measured or simulated, provide little useful information for risk assessment or risk mitigation related to mercury exposures.



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Interactive Comment



Specific Comments (referenced to the printer-friendly PDF format of the manuscript)

p. 28758, lines 7-10: The text states that the particles simulated in STILT are not tied to grids, so they can resolve sub-grid scale influences. However, the wind data used to define their transport are certainly gridded since the same wind data used by CMAQ were also used in STILT. How is it that particles traveling between discrete points offer any advantages when the wind information used to define their movement is gridded? Explain how STILT can resolve influences below the length scale of the meteorological data used to define transport and diffusion, especially when the emissions across the derived "footprint" and the precipitation driving wet deposition are also gridded to that same scale.

p. 28759, lines 14-15: The text states that the authors modified CMAQ V4.3 to include dry deposition of Hg0 and RGM. However, CMAQ has always treated dry deposition of RGM since it was originally adapted for atmospheric mercury simulation. Did you simply add a treatment for Hg0 dry deposition? Maybe the treatment for RGM dry deposition was modified. Please explain.

p. 28765, lines 19-21: The small subset of particles that did not reach the boundary should not be used to estimate boundary influences. Extrapolating to the boundary regardless of how far the particle endpoint is from that boundary seems dreadfully haphazard.

Section 3.1 and Figure 2: Upon first glance, Fig. 2 appears to show a very strong reduction in sampling error from 50 to 1000 particles, with little improvement offered from additional particles. However, the vertical scale for sampling error does not extend to zero. This gives the false impression that the adopted particle number of 3000 offers a nearly complete reduction of sampling error. This figure should either show the sampling error scaled all the way to zero or provide a secondary scale on the right showing the fraction of error reduction from the worst case (50 particles).

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 28755, 2010.