Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-725-RC3, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Deciphering the Chemical Forms of Gaseous Oxidized Mercury in Florida, USA" *by* Jiaoyan Huang et al.

Anonymous Referee #1

Received and published: 15 December 2016

The manuscript presents field observations of ambient concentrations of speciated mercury and other air pollutants at a costal site in Florida. GOM were monitored with Tekran and membranes. GOM dry deposition fluxes were calculated using Tekran concentrations (with a correction factor) and dry deposition velocities, and monitored using surrogate surfaces. Some GOM forms were observed on some membranes, and those possible forms were used to estimate dry deposition fluxes with various correction factors. Other analyses include back trajectory. The authors conclude that "the chemical forms of GOM in the atmosphere at OLF varied by season as suggested by Gustin et al. (2012)." However, it is unclear whether this manuscript presents results at additional sites, during a different time period, with different methodologies, or of different findings /conclusions from those reported in Gustin et al. (2012), Peterson et al. (2012), and the large number of publications (as listed in the reference section

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of the manuscript) by the two primary authors (i.e. Huang and Gustin) in the past few years on the topic of the chemical forms of GOM. Furthermore, as a standalone publication, this manuscript should provide more data and theoretical support that "the chemical forms of GOM in the atmosphere at OLF varied by season." My specific comments/suggestions are listed below.

1. Major concerns

1.1 There is a lack of support to conclude that there are different forms of GOM in different seasons.

Due to the small number of samples and inconsistent identification of most GOM forms between samples, there is a large uncertainty in identification of most GOM forms. The same can be said with Tekran and passive samplers. Similarly, there are many factors contributing to the deviation between measured and model estimated fluxes with different adjustment factors. Furthermore, the notion of more knowledge of GOM forms would lead to a better model-measurement agreement is debateable, because it was built upon the assumption of only one GOM form present in each dry deposition or air sample. Therefore, the reviewer is not convinced that disagreement between the different methods is largely due to limitations of the Tekran system and different GOM forms at different times of the year, as the authors suggest, due to insufficient data supporting this.

In other words, the limitations of the sampling method and study design are inadequate to justify the arrival of the conclusions. By leaping from not observed to not present (in concentrations), then from poor model-measurement agreement (in fluxes) to the presence of existing but not observed forms, the chosen approach seems to be convenient rather than fully based on science. The authors may want to provide an in-depth discussion of any observational or theoretical considerations supporting seasonal variation of chemical forms of ambient GOM at OLF. For example, the changes in any of the following factors with season, 1) prevailing wind/air mass directions, 2) oceanic or

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meteorological conditions, e.g. ambient temperature and ocean currents, 3) major local Hg sources, 4) major upwind Hg sources, 5) ambient concentrations of major reactants related with the GOM forms observed or hypothesized, and 6) any other factors that may lead to different GOM forms at OLF in different seasons. Furthermore, the authors may need to explain why any of those seasonal variations would lead to different chemical forms of ambient GOM at OLF. Perhaps the phrase "different predominant forms" is more conservative.

1.2 Section 3.4 Elevated Pollution Event: the selection of one elevated event raised many questions instead of establishing a valid association between the observed concentration levels and the observed GOM forms.

The analyses in this section rely on a few two-week samples and 72-hr back trajectories to "hypothesize this is a common event in the spring that represents free troposphere transport". The questions that need further explanation include: 1) why free troposphere transport is limited to spring if it is responsible for elevated concentrations, 2) if the prevailing air mass directions support the trans-pacific transport of air pollutants, why it only caused elevated concentrations in spring, 3) given the proximity of the sampling site to the ocean, why HgBr2 was only observed once in spring during the one year study period, and 4) a prolonged period of elevated pollutant concentrations is often associated with local emissions and/or local/regional weather conditions. What evidences suggest that this site was under "the impact of subsidence of air from the stratosphere/troposphere" for 4-weeks with little impact of synaptic weather conditions which could lead to strong mixing hence lower concentrations. The authors may want to include analyse of other periods when GOM forms were observed or omit this section considering the limitations of using two-week samples and 72-hr back trajectories to explain long range transport (beyond three days) and transformations of Hg (in the time scale of hours to days).

2. Editorial comments and suggestions

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1) “Tekran®, and nylon, and cation exchange membranes”, please clarify.

2) L187-192, this paragraph is less relevant to section 3.1, could be omitted or placed in another section.

3) “Overall, air concentrations measured by the Tekran® system in this study were similar to those measured in 2010 (Peterson et al., 2012).” and L250, same site?

4) L200-204, please rephrase.

5) L205, please provide the two forms.

6) L236-238, please provide the forms at the rural site and traffic site.

7) “Modeled GOM dry deposition fluxes were calculated”, “measured Hg dry deposition fluxes were similar to those modeled simulations with modeled GOM dry deposition α=β=2”, please rephrase.

8) L282-283, please rephrase.

9) “the profile was a gradual increase”, please rephrase.

10) “In order to improve our understanding of Hg air-surface exchange, and measure GOM physiochemical properties of different GOM compounds need to be understood”, please rephrase.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-725, 2016.