Reviewer #2:

The authors present results of a box-model simulation of Hg chemistry at three sites in southern New Hampshire, USA. The sites are located in different environments (marine, coastal, and elevated), which allows the authors to examine the similarities and differences in Hg chemistry in these environments. The authors conclude that Br and BrO dominate Hg oxidation during the day and H_2O_2 at night at the marine site, while O_3 and OH are dominant at the coastal and inland sites. I found the comparison in Hg chemistry between the sites interesting. Atmospheric Hg chemistry remains one of the least understood processes controlling Hg cycling in the environment. Studies like this that use models to interpret in situ Hg observations in different environments are necessary to fully characterize the oxidation of Hg in the atmosphere. However, I have a number of major concerns that the authors should address to make their study convincing.

We thank the reviewer for their detailed, thoughtful, constructive comments and suggestions. The manuscript has been revised carefully. Below we addressed the review point by point.

Major comments: 1) The authors examine the oxidation of GEM with the set of gas phase reactions listed in Table 1. There is high uncertainty in these reaction rates, up to a factor of 10 for reactions of GEM with Br and BrO. The recent review by Ariya et al. (2015) has a compilation of all previously reported estimates for GEM oxidation reaction rates. The authors perform one sensitivity study addressing the uncertainty in the GEM+O_3 rate, but seem to ignore the uncertainty in the remaining reactions. A discussion of the effect of these uncertainties are necessary before any conclusion can be reached about the dominant GEM oxidation pathways at the studies sites.

The major oxidation reactions of GEM are GEM + O_3 and GEM + Br in our box model. For the GEM + Br reaction, Ariya et al. (2002) yielded a rate constant of 3.2×10^{-12} cm^3 $molecule^{-1}$ s^{-1} using a relative rate method. However, Ariya et al. (2002) used one single rate reference only, which largely limited the accuracy of their results (Hynes et al. 2009). Moreover, large amounts of cyclohexane (an OH scavenger) used in Ariya et al. (2002)'s experiment may lead to an enhancement in the absorption of reactants on the cell walls (Hynes et al. 2009). A number of studies (Spicer et al. 2002; Donohoue et al. 2006; Sumner et al. 2011; Subir et al., 2011; Goodsite et al., 2004, 2012) showed a narrow range of $(3.0 – 6.4)\times10^{-13}$ cm^3 $molecule^{-1}$ s^{-1} for the rate coefficient of GEM + Br, from which we used a temperature dependent rate coefficient of $3.7\times10^{-13}(T/298)^{-2.76}$ cm^3 $molecule^{-1}$ s^{-1} from Goodsite et al. (2004; 2012). To further investigate the GEM + Br rate coefficient sensitivity on GOM simulation, we added a new sensitivity scenario using Ariya et al. (2002) rate coefficient (section 3.4.1 in the revised manuscript). As a result, using the greater rate coefficient of Ariya et al. (2002) produced a factor of 3 or higher GOM mixing ratios than the base scenario.

We added the following discussion on the effect of reaction kinetics uncertainties on model simulations in section 3.4.1 (lines 522 - 527) of revised manuscript:

"Using a slower rate coefficient of GEM + O_3 (Hall, 1995) had similar effects as not including the GEM + O_3 reaction, i.e. decreasing GOM mixing ratios, especially at nighttime, and brominated GOM species becoming dominant. The GEM + OH reaction was not as important as GEM + O_3 or Br. The use of a higher GEM + Br rate coefficient derived from the study by Ariya et al. (2002) caused more than a factor of 3 higher GOM and PBM resulting in overestimated GOM for most cases."

2) How were the concentrations of the species that weren't measured set? How were the concentrations of Br/Cl/I species determined at the three sites? The authors briefly mention this in Section 3.4.1. This is a key aspect of the study and should be discussed in detail in Section 2.

For species that were not measured, we use the chemical mechanism to calculate their concentrations. Initial concentrations of most unmeasured species were set as the values in similar environments from the literature if available. Br/Cl/I concentrations were all calculated from the model given initial concentrations of 1 pptv (e.g. Finley et al., 2008; except for AI) for Br_2, Cl_2, and I_2 species. At AI, we set the Br_2 concentration to be constant during simulations and used Saiz-Lopez et al. (2006)'s values to constrain [BrO]. At TF and PM, the initial concentrations of Br_2, Cl_2, and I_2 were not sensitive factors for the simulated concentrations of Br/Cl/I, because during the simulations, Br_2, Cl_2, and I_2 were rapidly depleted without sources in inland environments.

We have added such information in section 2.1.2 (lines 203 - 207 in the revised manuscript) as follows:

"Br/Cl/I concentrations were all calculated from the model given initial concentrations of 2 pptv (Finley et al., 2008; except for AI) for Br_2, Cl_2, and I_2 species. At AI, the Br_2 initial concentration was set to be constant during simulations and used Saiz-Lopez et al. (2006)'s values to constrain [BrO]. Detailed information can be found in Section 3.3.1."

3) The authors performed several sensitivity studies with the box-model by varying different physical and chemical parameters. However, these sensitivity studies seem out of place. The authors do not specify why they chose to vary the parameters listed in Table 3, and not others. Secondly, the presentation of the results of the sensitivity studies is not thorough. There is no discussion of how the results of the sensitivity studies affect the overall conclusions. Section 3.3.3 addresses Br chemistry in the MBL. I do not think this fits in this study, considering that there were no BrO measurements that could be used to compare with the model results.

The ranges of parameters in sensitivity studies were based upon the varying range of each parameter from observations and the literature. The liquid water content range was derived from Hedgecock et al. (2003). The temperature range was based on the magnitude of observed average temperature diurnal cycles. We added such information in section 3.4.1 of revised manuscript (lines 487 - 490).

More discussion on the effect of these sensitivity tests was added (lines 518 - 530 in the revised manuscript):

"In summary, the parameters used in gas-particle partitioning processes including solar radiation values, temperature, and the rate coefficients of major GEM oxidation reaction, could all affect simulated GOM mixing ratios but with varying degrees. Aerosol properties were suggested to play a very important role in the partitioning of ambient GOM and PBM species and thus should be better represented in future Hg model simulation studies. Using a slower rate coefficient of GEM + O_3 (Hall, 1995) had similar effects as not including the GEM + O_3 reaction, i.e. decreasing GOM mixing ratios, especially at nighttime, and brominated GOM species becoming dominant. The GEM + OH reaction was not as important as GEM + O_3 or Br. The use of a higher GEM + Br rate coefficient derived from the study by Ariya et al. (2002) caused more than a factor of 3 higher GOM and PBM concentrations resulting in overestimated GOM for most cases. GOM and PBM production appeared to favor lower temperature during daytime and higher temperature at night, and simulated GOM concentrations were not as sensitive to temperature change as to solar radiation and gas-particle partitioning."

Regarding section 3.3.3 (section 3.4.3 in the revised manuscript), in our opinion, this is one of the original contributions this study offers. Considering the importance of halogen chemistry in Hg cycling, we think halogen chemistry needs to be interactive with Hg chemistry. Constraining the BrO simulations using the observations from Saiz-Lopez et al. (2006), our box model results suggested that Br and BrO are two key compounds in determining GOM mixing ratios in the MBL. Section 3.4.3 in the revised manuscript includes theoretical analysis and discussion of the important bromine reactions that could affect Br and BrO simulations, which has vital importance in this study and can provide guidance for future Hg studies.

4) Hg^{2+} reduction reactions were included in the model. Is reduction an important control on GOM mixing ratios? Some discussion of the effect of the reduction pathways on GOM and PBM would be valuable. The Tekran 2537/1130/1135 typically measures GEM, RGM, and PBM. The PBM measurements are not discussed in the manuscript. Can the PBM measurements be used to constrain the reduction rates?

A table with aqueous Hg reactions used in our model was added as Table S1.

Aqueous Hg reduction is one of the major sources of GEM in the atmosphere. Therefore, aqueous Hg reactions is supposedly a factor controlling GEM mixing ratios and further influence GOM mixing ratios. However, in this study aqueous Hg reduction was not an important control on GOM simulations. This is because GEM mixing ratios in the model were fixed using observed values. The uncertainties associated with aqueous Hg reactions would not influence GEM mixing ratios and therefore have minor effects on simulated GOM mixing ratios.

It is true that high quality GOM and PBM measurements would be of great help for modelling studies to evaluate the schemes such as gas-particle partitioning process as well as to constrain the aqueous reduction rate. However, the inlet of the Tekran speciation sampling system had an elutriator inlet with an acceleration jet to remove aerosols > 2.5 μm so that only PBM on fine particles was measured. The PBM calculated from the box model does not include size fractionation, thus Tekran $PBM_{2.5}$ measurements could not be used to constrain our simulations and the reduction rate.

5) In Section 1, the authors briefly discuss previous studies of Hg chemistry by Hedgecock et al., Holmes et al., and Wang et al. The present study of Ye at al. is very similar to these previous studies. All of them examine the diurnal cycle of oxidized Hg in the mid-latitude marine boundary layer using a box-model. The authors should include a discussion of how their results compare with the findings in these previous studies.

Section 1 was revised and expanded to reflect the aspects of this study that differentiate it from previous studies.

Minor comments: Page 4, line 9: "Hg in the MBL cycles differently in coastal or inland areas." The difference needs to be expanded upon as this is directly related to the present study. How is the cycle different?

The major differences of Hg cycles between MBL and coastal or inland areas are reflected in the magnitude and speciation of GOM, which are due to different chemical, meteorological and atmospheric conditions such as halogen radical mixing ratios, boundary layer height, and atmospheric particles size and properties. More detailed discussion can be found in section 3.1 and section 3.4.2 in the revised manuscript.

Page 5, line 23: Please add a list of reactions and their rates as a supplement, given that a few of the reactions do not seem to follow the JPL Report #17 recommendations.

We have 424 reactions in total, which is too many to be included in the publication. We would be happy to provide the reactions upon request.

Halogen reactions listed in Table 4 were following the halogen chemistry reviews by Atkinson et al. (2004; 2008). We added this information in section 2.

Page 6, line 11: Please include a table with the reaction rates and references for the aqueous-phase reactions.

A table showing aqueous Hg reactions in our model was added as Table S1.

Page 7, line 1: Not all previous modeling studies have used simple approximations. The model of Hedgecock et al. (2003, 2005) uses detailed MBL chemistry.

The sentence has been revised.

Page 7, line 11: How are the wind speed measurements used in the box-model?

Wind speed measurements were used for case selection, not input for the box model. The text was revised to reflect this.

Page 7, line 19: "...were set to be constant during a simulation." Please specify the length of a simulation. Was it one day or one hour?

The length of a simulation is one hour. The sentence was revised to include this.

Page 9, line 20: It would be interesting to see how the source regions of the air masses at the three sites differed. The back trajectories for only the AI site are discussed in the text.

The back trajectories for the PM and TF sites (Fig. 1) showed air masses source regions. Air masses reaching PM originated from inland areas west to north of the site, while air masses at TF half came from northwestern to northern inland areas and half from the marine boundary layer. However, we did not find correlation between source regions and GOM mixing ratios at TF and PM. This is why the origin of air masses at the two sites was not discussed.

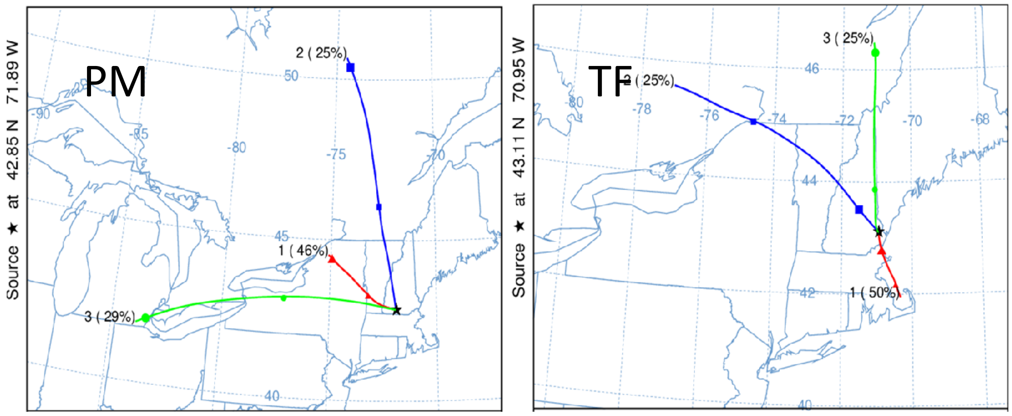

Figure 1. Clustered 24-hour backward trajectories of air masses in all cases at PM and TF.

Page 10, line 21: The LOD for the GOM observations was 0.1 ppqv at AI, but it appears to be much lower at PM. Figure 2 shows most GOM observations at PM below 0.05 ppqv. Please specify the LOD for GOM at PM.

The GOM detection limit for all three instruments were derived as ~0.1 ppqv, based on three times the standard deviation of the averaged blank (Sigler et al., 2009; Mao and Talbot, 2012). We added this information in section 2.1.2.

Page 12, line 12: HgO is considered a GOM species here, although the authors state in the Introduction (page 3, line 22) that “a consensus has emerged that GEM+O_3 reaction most likely occurs with solid-phase products...”

The experimental study by Pal and Ariya (2004) measured 1% of HgO produced by GEM + O_3 on an aerosol filter. Snider et al. (2008) showed HgO(s) production in their kinetic and product study. A theoretic study of Schroeder et al. (1998) suggested HgO would not exist as an isolated molecule at a decomposition temperature of +500 °C. However, the GEM + O_3 reaction and decomposition temperature (Schroeder et al., 1998) could also be impacted by the presence of other ambient gases (Snider et al., 2008; Gustin et al., 2013; Seigneur et al., 1994). Moreover, a recent study by Huang et al. (2013) observed gas-phase HgO using nylon and cation exchange membranes. Overall our knowledge about this reaction remains nebulous. We added this discussion in the introduction (section 1).

Page 12, line 19, I was surprised not to see $HgBrNO_2$ as one of the more abundant GOM species. I expected $HgBrNO_2$ to be produced faster than HgBrOOH and Hg-BrOBr, given the typically higher concentrations of NO_2.

In checking reactions forming $HgBrNO_2$, we found a mistake in NO_x input. We should have fixed NO_x concentrations in the input for the simulations but it was mistakenly left unfixed. In this revised version, we have rectified the mistake. As a result, the dominant brominated GOM species was changed to $HgBrNO_2$, and following with HgBrO; other brominated GOM species were negligible. However, Hg+Br reaction is so slow compared to further HgBr oxidation reactions that Hg+Br is the rate-limiting step for these two steps of reactions. Therefore, the change in the total GOM production was minor, and major conclusions remain unchanged (See Section 3.2 in the revised manuscript).

Page 13, line 14: Why were HgO and $Hg(OH)_2$ more sensitive than halogenated GOM species to gas-particle partitioning?

The difference between sensitivity of HgO/$Hg(OH)_2$ and halogenated GOM species to gas-particle partitioning was caused by higher molar mass of halogenated GOM species than HgO/$Hg(OH)_2$. When taken into calculations, compounds with smaller molar mass had a higher gas-to-particle rate based on the scheme described in section 2.1.3. The Henry's constant values of $Hg(OH)_2$ and halogenated GOM species are large enough to be not as sensitive as the molar mass of the compounds is to gas-to-particle partitioning.

Page 24, line 25: I do not see an order of magnitude difference between the peaks in Figure 5. I see a factor of 2-3 difference.

Corrected.

Page 19, lines 12 onwards: Could entrainment from the free troposphere explain this inverse relation between GOM and RH? Entrainment from the free troposphere was not treated explicitly, yet the boundary layer height at the TF site varied diurnally. Was it assumed that this entrainment does not change GOM mixing ratios in the boundary layer?

In this study, we selected clear-sky and calm wind conditions, usually accompanied by strong stability with a strong inversion layer at the top of the daytime convective PBL layer based on measurements from the literature (e.g., Hogan et al., 2009). Minimal entrainment at the top of the boundary layer was thus expected.

We agree with the reviewer's point that at TF, the GOM in the remnant layer could be mixed down to the surface in the morning when the boundary layer rises. The observed daytime GOM mixing ratio peak is around 0.8 ppqv, and the contribution of downward mixing from the remnant layer at TF was estimated by Mao et al. (2006) to be about ~23% in the time window of after sunrise and 10 am local time. Under such circumstances, the contribution from the preceding convective boundary layer to the morning GOM mixing ratios would at most be ~0.2 ppqv. Moreover, even though GOM in the remnant layer at night did not deposit to the surface, it could be lost by deposition to aerosols and via other unknown mechanisms. Taking these into consideration, that 0.2 ppqv contribution from the remnant layer would be the upper limit. As the day progresses and solar radiation gets stronger, the GOM mixing ratio is mostly driven by photochemical production.

Page 21, line 29: "Clearly, the under-estimation case occurred under the strongest Bermuda High influence..." It isn't clear to me. Can the authors explain a bit more why it is the influence of the Bermuda High, and not just a transient high-pressure system?

The under-estimation cases were 06/13/2008 and 08/22/2007, the meteorological conditions of these days were illustrated using the NCEP 1° x 1° meteorological reanalysis data (Fig. 2). The observed GOM concentrations peaked at 14:00 LT on 06/13/2008 and 16:00 LT on 08/22/2007 respectively. On 13 June 2008, the Bermuda high pressure system covered almost the entire eastern US coastline, where our sites are located. This high pressure system lasted 4 days (10 – 14 June 2008). On 22 August 2007, the continental part of the Bermuda high pressure system was over the southeastern US extending to the northeast. These lasting high pressure systems caused regional buildup pollutants, explaining the observed high mixing ratios of GOM in the two cases. We will include these figures in the supplemental material.

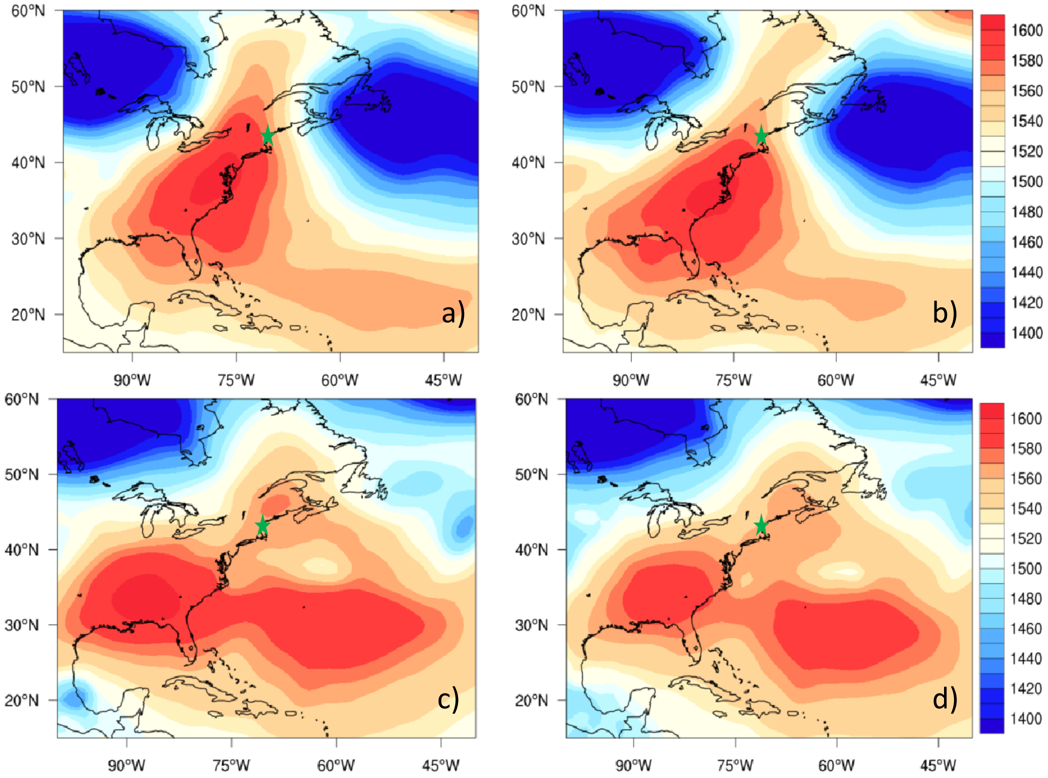

Figure 2. Geopotential height for a) 06/13/2008 08:00 EDT, b) 06/13/2008 14:00 EDT, c) 08/22/2007 14:00 EDT, and d) 08/22/2007 20:00 EDT at 850 hPa, the green star shows the location of TF site.

Page 23, line 7: "It was hypothesized that..." This was not substantiated in the study, and does not belong in the conclusions.

These hypotheses were developed based on the modeling and analysis work in the paper. The text was revised to reflect the logical steps to take to arrive at the hypotheses.

Page 23, line 24: The authors allude to problems in GOM measurements using the Tekran instrument. If the measured GOM is indeed biased low by a factor of 2 or 3 under certain conditions, how does it affect this study's conclusions? This is important and needs to be discussed in a little more detail.

Page 23: The authors should also point out to the reader that, in the absence of speciated measurements of oxidized Hg compounds, the results of a modeling study cannot be used to conclusively identify the dominant oxidants of Hg in the atmosphere.

The reviewer raised excellent points here. We agree that without measurements of speciated GOM, modeling results cannot be used to conclusively identify the dominant oxidants of Hg, as well as dominant GOM species in that matter, in the atmosphere. Indeed the potential uncertainty in ambient Hg measurements especially GOM is a major concern in the community. We had some discussion on the effect of uncertainty in GOM measurements on our interpretation of measurements data. With the reviewer's suggestion in mind, the discussion was expanded to discuss the potential effect of biased low GOM measurements on our conclusions in the last section (lines 623 - 640 in the revised manuscript).

That being said, it is unlikely to put any range on the bias of our GOM concentrations considering our own GOM measurements and the literature. Recent laboratory experiments and reviews (Lyman et al., 2010; Jaffe et al., 2014; McClure et al., 2014; Huang and Gustin, 2015; Gustin et al., 2015) reported O_3 and relative humidity (RH) interferences on mercury halides for KCl-coated denuder, which was a part of Tekran 1130 unit used for GOM field measurements commonly in the community as well as the observations of this study. Huang and Gustin (2015) suggested a linear relationship between RH and GOM loss (in %) in GOM measurements, i.e., RH = 0.63 GOM loss % + 18.1, $r^2 = 0.49$, $p < 0.01$, at RH range of 21 to 62%. In our GOM measurements, the interferences of RH at our sites should have largely been eliminated since we used a custom-built refrigerator assembly and a canister of drierite to cool and dry air streams before entering into the 1130 pump module (Sigler et al., 2009). As a result, the RH of air streams was kept < 25%, therefore the upper limit of GOM loss cause by RH was < 10% using Huang and Gustin (2015)'s equation.

With regard to O_3 interference, the experimental study (Lyman et al., 2010) showed 3 to 37% reduction on the collection efficiency of $HgCl_2$, and the proposed reaction was $HgCl_2 + 2O_3 \rightarrow Hg^0 + 2O_2 + ClO$. However, the quantitative extent of the bias caused by O_3 in field GOM measurements was yet derived (Lyman et al., 2010). Huang et al. (2013) showed lower collection efficiency of KCl denuders compared to nylon membrane and the cation exchange membrane for $HgBr_2$, $HgCl_2$, HgO, $HgSO_4$, and $Hg(NO_3)_2$ in laboratory tests. However, for field measurements (Huang et al., 2013; Gustin et al., 2013), since GOM and PBM could not be distinguished from total reactive mercury using nylon membrane and cation exchange membrane chambers, the quantitative bias extent derived for total reactive mercury could not be directly used for GOM. Moreover, Huang et al. (2013) suggested that in their marine boundary layer site and highway impacted site, ambient GOM most likely existed in forms other than the laboratory tested species. Therefore, bias low GOM collection efficiency of KCl-coated denuders in field measurements remains speculative at this point.

Quality measurement data are used as ground truth for atmospheric Hg modeling studies, notwithstanding their limitation. Better instrumentation and/or solidly quantified bias for current instruments are in urgent need and are of essential importance to atmospheric Hg modeling. Nevertheless, even if models did not perfectly reproduced observations, the information derived from model simulations and sensitivity studies could provide insight into how the mechanisms work.

The discussion added in the Summary section is as follows (lines 623 - 640 in the revised manuscript):

"It should be noted that without measurements of speciated GOM, modeling results cannot be used to conclusively identify the dominant oxidants of Hg, as well as dominant GOM species in that matter, in the atmosphere. Indeed, the potential uncertainty in ambient Hg measurements especially GOM is a major concern in the community. That being said, it is unlikely to have a quantitative understanding of the bias of our GOM concentrations. Recent laboratory experiments and reviews (Lyman et al., 2010; Jaffe et al., 2014; McClure et al., 2014; Huang and Gustin, 2015; Gustin et al., 2015) reported O_3 and relative humidity (RH) interferences on mercury halides for KCl-coated denuder, the part of Tekran 1130 unit commonly used for GOM field measurements. As stated in Section 2, in our GOM measurement the RH effect was minimized by adding refrigeration to remove excess of water in the airsteam. O_3 interference and bias low GOM collection efficiency of KCl-coated denuders were limited to a handful of GOM species in laboratory experiments and remain untested in field measurements. If the measured GOM concentrations were indeed biased low by a factor of 2 or 3 under certain conditions as previous studies speculated, the matching cases at AI and TF would be reduced from 50% of the total cases to 30%, and the model would potentially underestimate GOM concentrations in the remaining cases (70%) by a factor of 3 to 4. It is however hard to speculate the effect at PM since most GOM observations there were below the LOD. This suggested even greater unknowns in our understanding of Hg chemistry."

Table 2: Please include the standard deviation of the observed variables.

Added the standard deviation values for observed variables in Table 2.

Figure 1: Please add some geographical context to the map. May be show the latitude/ longitude girds, and the land/ocean boundary.

Plotted a new map for Figure 1 (Fig. 3 showing below) with latitude/longitude grids and land/ocean boundary showed.

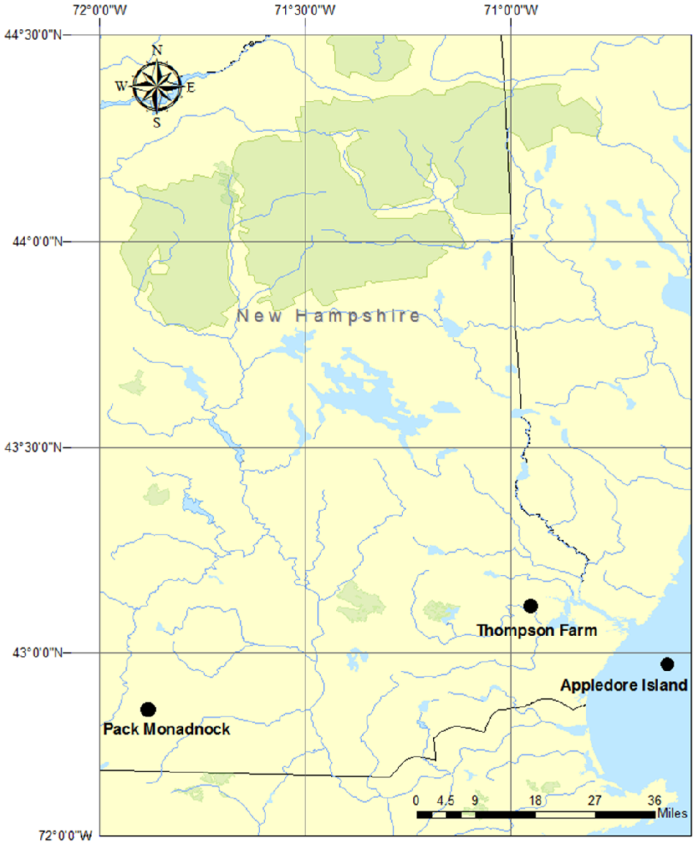

Figure 3. New Hampshire site map: Appledore Island (marine), Thompson Farm (coastal), and Pack Monadnock (inland elevated).

Figure 10: The back trajectories suggest strong regional influence at the AI site. Can the authors reconcile this with their assumption for the box-model that regional transport is negligible?

The trajectories were used to identify the origin of the air mass reaching AI. GEM was long-lived enough to originate from the same source region of the air mass. However, GOM in the air masses did not necessarily originate from the same source region due to its short lifetime. Under the conditions of strong atmospheric stability as selected in this study, GOM would likely be in-situ, photochemically produced.

Technical comments: Page 1, line 14: May be the title can specify that the study focuses on the summertime.

Upon the reviewer's suggestion the title was changed to "Investigation of processes controlling summertime gaseous elemental mercury oxidation at mid-latitudinal marine, coastal, and inland sites".

Page 1, line 14: The term Hg(II) is not needed here.

Deleted.

Page 3, line 7-8. "GOM and PBM are...subject to dry and wet deposition..." GEM is also subject to dry deposition.

Added.

Page 3, line 27: The sentence starting with "In the MBL..." needs to be rephrased for clarity.

Revised.

Page 5, line 10: "...initial GEM mixing ratios...were set to be constant mimicking GEM emission flux." This sentence is unclear and should be reworded. I think removing the clause "and were set..." may help.

Revised.

Page 10, line 4: FB is fractional bias.

Changed.

Page 11, line 26: Reference to Section 3.2.2. Should this refer to Section 3.4?

In this sentence, we meant that the reasons of large variations of GOM daytime peaks between AI, TF, and PM. We have discussed this in Section 3.4.2 of revised manuscript. We have corrected this.

Page 14, line 15: It seems TF_AIdry, PM_AIdry, TF_AIaero, PM_AIaero are not discussed any further. It would be better to not introduce them here.

Deleted.

Page 17, line 1: I think it would be more appropriate to place the model evaluation section before the sensitivity studies.

Agreed and done.

Page 19, line 27: "It was thus hypothesized that certain processes..." This sentence is vague. Please reword.

Revised.

Page 23, line 15: “The updated chemical mechanism largely improved GOM simulations...”. Improved with respect to what?

Revised to “The updated chemical mechanism largely improved the simulation of the magnitude and pattern of GOM diurnal variation at the coastal and inland sites.”

Figure 2: What do the “filled circles” represent? Expand the site abbreviations in the caption. The font size is too small.

The figure was revised.

Figure 3: Font size is too small. In the caption: do the bars “represent” the range of simulated GOM?

The figure was revised and the font size was increased for better presentation. Now the bars represent standard deviations of simulated GOM.

Figure 4: Please change “Other_RGM” to “Other GOM species”. Please increase font size. It is hard to distinguish between the lines in Figure 3(a).

Changed.

Figure 8: Caption: “(“Observed”, red, “Simulated”, triangle)”. Please correct typographical error.

Corrected.

Figure 9: It is difficult to distinguish between the Simulated_under-estimated, Simulated_matching, and Simulated_over-estimated lines. It would be also be helpful to maintain consistency between the figures in what is represented by the error bars.

We revised the figures and used error bars for standard deviation only.

REFERENCE

Ariya, P. A., Khalizov, A. and Gidas, A.: Reactions of gaseous mercury with atomic and molecular halogens: Kinetics, product studies, and atmospheric implications, Journal of Physical Chemistry A, 106(32), 7310–7320, 2002.

Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson, R. F., Hynes, R. G., Jenkin, M. E., Rossi, M. J., Troe, J. and Wallington, T. J.: Evaluated kinetic and photochemical data for atmospheric chemistry: Volume IV – gas phase reactions of organic halogen species, Atmos. Chem. Phys., 8(15), 4141–4496, doi:10.5194/acp-8-4141-2008, 2008.

Donohoue, D. L., Bauer, D., Cossairt, B. and Hynes, A. J.: Temperature and pressure dependent rate coefficients for the reaction of Hg with Br and the reaction of Br with Br: A pulsed laser photolysis-pulsed laser induced fluorescence study, Journal of Physical Chemistry A, 110(21), 6623–6632, 2006.

Finley, B. D. and Saltzman, E. S.: Observations of Cl_2, Br_2, and I_2 in coastal marine air, J. Geophys. Res., 113(D21), D21301, doi:10.1029/2008JD010269, 2008.

Goodsite, M. E., Plane, J. M. C. and Skov, H.: A Theoretical Study of the Oxidation of Hg^0 to $HgBr_2$ in the Troposphere, Environmental Science and Technology, 38(6), 1772–1776, 2004.

Goodsite, M. E., Plane, J. M. C. and Skov, H.: Correction to A Theoretical Study of the Oxidation of Hg^0 to $HgBr_2$ in the Troposphere, Environ. Sci. Technol., 46(9), 5262–5262, doi:10.1021/es301201c, 2012.

Gustin, M. S., Amos, H. M., Huang, J., Miller, M. B. and Heidecorn, K.: Measuring and modeling mercury in the atmosphere: a critical review, Atmos. Chem. Phys., 15(10), 5697–5713, doi: 10.5194/acp-15-5697-2015, 2015.

Gustin, M. S., Huang, J., Miller, M. B., Peterson, C., Jaffe, D. A., Ambrose, J., Finley, B. D., Lyman, S. N., Call, K., Talbot, R., Feddersen, D., Mao, H. and Lindberg, S. E.: Do we understand what the mercury speciation instruments are actually measuring? Results of RAMIX, Environmental Science and Technology, 47(13), 7295–7306, 2013.

Hall, B.: The gas phase oxidation of elemental mercury by ozone, Water Air Soil Pollut., 80(1-4), 301–315, 1995.

Hedgecock, I. M., Pirrone, N., Sprovieri, F. and Pesenti, E.: Reactive gaseous mercury in the marine boundary layer: Modelling and experimental evidence of its formation in the Mediterranean region, Atmos.Environ., 37(SUPPL. 1), S41–S49, 2003.

Hogan, R. J., Grant, A. L. M., Illingworth, A. J., Pearson, G. N. and O'Connor, E. J.: Vertical velocity variance and skewness in clear and cloud-topped boundary layers as revealed by Doppler lidar, Q.J.R. Meteorol. Soc., 135(640), 635–643, doi:10.1002/qj.413, 2009.

Holmes, C. D., Jacob, D. J., Mason, R. P. and Jaffe, D. A.: Sources and deposition of reactive gaseous mercury in the marine atmosphere, Atmospheric Environment, 43(14), 2278–2285, 2009.

Huang, J. and Gustin, M. S.: Uncertainties of Gaseous Oxidized Mercury Measurements Using KCl-Coated Denuders, Cation-Exchange Membranes, and Nylon Membranes: Humidity Influences, Environ. Sci. Technol., 49(10), 6102–6108, doi:10.1021/acs.est.5b00098, 2015.

Huang, J., Miller, M. B., Weiss-Penzias, P. and Gustin, M. S.: Comparison of gaseous oxidized Hg measured by KCl-coated denuders, and nylon and cation exchange membranes, Environmental Science and Technology, 47(13), 7307–7316, 2013.

Hynes, A. J., Donohoue, D. L., Goodsite, M. E. and Hedgecock, I. M.: Our current understanding of major chemical and physical processes affecting mercury dynamics in the atmosphere and at the air-water/terrestrial interfaces, Mercury Fate and Transport in the Global Atmosphere: Emissions, Measurements and Models, (Journal Article), 427–457, 2009.

Jaffe, D. A., Lyman, S., Amos, H. M., Gustin, M. S., Huang, J., Selin, N. E., Levin, L., ter Schure, A., Mason, R. P., Talbot, R., Rutter, A., Finley, B., Jaeglé, L., Shah, V., McClure, C., Ambrose, J., Gratz, L., Lindberg, S., Weiss-Penzias, P., Sheu, G.-R., Feddersen, D., Horvat, M., Dastoor, A., Hynes, A. J., Mao, H., Sonke, J. E., Slemr, F., Fisher, J. A., Ebinghaus, R., Zhang, Y. and Edwards, G.: Progress on Understanding Atmospheric Mercury Hampered by Uncertain Measurements, Environ. Sci. Technol., 48(13), 7204–7206, doi:10.1021/es5026432, 2014.

Lyman, S. N., Jaffe, D. A. and Gustin, M. S.: Release of mercury halides from KCl denuders in the presence of ozone, Atmos. Chem. Phys., 10(17), 8197–8204, doi:10.5194/acp-10-8197-2010, 2010.

Mao, H. and Talbot, R.: Speciated mercury at marine, coastal, and inland sites in New England-Part 1: Temporal variability, Atmospheric Chemistry and Physics, 12(11), 5099–5112, 2012.

Mao, H., Talbot, R., Nielsen, C. and Sive, B.: Controls on methanol and acetone in marine and continental atmospheres, Geophys. Res. Lett., 33(2), L02803, doi:10.1029/2005GL024810, 2006.

McClure, C. D., Jaffe, D. A. and Edgerton, E. S.: Evaluation of the KCl Denuder Method for Gaseous Oxidized Mercury using HgBr2 at an In-Service AMNet Site, Environ. Sci. Technol., 48(19), 11437–11444, doi:10.1021/es502545k, 2014.

Pal, B. and Ariya, P. A.: Studies of ozone initiated reactions of gaseous mercury: Kinetics, product studies, and atmospheric implications, Physical Chemistry Chemical Physics, 6(3), 572–579, 2004.

Saiz-Lopez, A., Shillito, J. A., Coe, H., and Plane, J. M. C.: Measurements and modelling of I_2, IO, OIO, BrO and NO_3 in the mid-latitude marine boundary layer, Atmos. Chem. Phys., 6, 1513–1528, doi:10.5194/acp-6-1513-2006, 2006.

Schroeder, W. H. and Munthe, J.: Atmospheric mercury - An overview, Atmospheric Environment, 32(5), 809–822, 1998.

Seigneur, C., Wrobel, J. and Constantinou, E.: A chemical kinetic mechanism for atmospheric inorganic mercury, Environmental Science and Technology, 28(9), 1589–1597, 1994.

Sigler, J. M., Mao, H. and Talbot, R.: Gaseous elemental and reactive mercury in Southern New Hampshire, Atmos. Chem. Phys., 9(6), 1929–1942, doi:10.5194/acp-9-1929-2009, 2009.

Snider, G., Raofie, F. and Ariya, P. A.: Effects of relative humidity and CO(g) on the O_3-initiated oxidation reaction of Hg^0(g): Kinetic & product studies, Physical Chemistry Chemical Physics, 10(36), 5616–5623, 2008.

Spicer, C. W., Satola, J., Abbgy, A. A., Plastridge, R. A. and Cowen, K. A.: Kinetics of Gas-Phase Elemental Mercury Reaction with Halogen Species, Ozone, and Nitrate Radical under Atmospheric Conditions, (Journal Article), 2002.

Subir, M., Ariya, P. A. and Dastoor, A. P.: A review of uncertainties in atmospheric modeling of mercury chemistry I. Uncertainties in existing kinetic parameters - Fundamental limitations and the importance of heterogeneous chemistry, Atmos.Environ., 45(32), 5664–5676, 2011.

Sumner, A. L., Spicer, C. W., Landis, M. S. and Stevens, R. K.: Kinetics of gaseous elemental mercury oxidation reactions under conditions of relevance to the atmosphere, Atmos.Environ., 2011.

Wang, F., Saiz-Lopez, A., Mahajan, A. S., Martín, J. C. G., Armstrong, D., Lemes, M., Hay, T. and Prados-Roman, C.: Enhanced production of oxidised mercury over the tropical Pacific Ocean: A key missing oxidation pathway, Atmospheric Chemistry and Physics, 14(3), 1323–1335, 2014.