Interactive comment on "Successes and challenges of measuring and modeling atmospheric mercury at the part per quadrillion level: a critical review" by M. Sexauer Gustin et al.

Response to Anonymous Reviewer 1- ACP_Gustin et al.

We thank this reviewer for his/her comments.

General comment: The authors try to substantiate their claims about higher than reported GOM concentrations by GEM changes, e.g. in the discussion of the results by Weiss-Penzias et al. (2003). Because of changing winds and the diurnal breathing of the boundary layer it is almost impossible to separate photochemistry from transport without measuring special tracers for photochemistry. Mercury studies are notorious for lack of measurements of such tracers. Consequently, most discussions about GEM chemistry from observed GEM changes remain unfounded speculations.

Response: In the discussion of the case study, we have written the text using language that points to points to potential reaction.

Line 40: "We recommend applying a factor of 3 as a conservative correction to reported GOM concentrations." Why not 1.6 or 12, the factors mentioned before? What is the meaning of "conservative"? "Reported GOM concentrations" relate to those measured by Tekran? What about other GOM methods? This recommendation is too sweeping and ambiguous and should be thus deleted.

Response: We have deleted that sentence.

Lines 65-67: An explanation was published by Soerensen et al., 2012. Please correct the sentence accordingly.
Response: Lines 61-66 referring to atmospheric trends have been revised and the phrasing "remains unexplained" has been removed. The sentences now read, " Over the last ~15 years, measured concentrations of atmospheric Hg have been declining (Slemr et al., 2011; Cole and Steffen, 2010; Soerensen et al., 2012; Cole et al., 2010; 2014), despite inventories suggesting global anthropogenic emissions have been relatively flat or increasing (AMAP/UNEP, 2013). This conundrum has challenged our understanding of Hg cycling and emissions, and underscores the need for continued atmospheric Hg monitoring." The reference to Soerensen et al. (2012) has not been added. In order to explain the atmospheric trends, Soerensen et al. (2012) prescribed a 4 pM decline between 1990 and 2010 over the top 1000 m of the entire North Atlantic Ocean based on measured seawater profiles at BATS. There is growing agreement that the early BATS data were likely contaminated (AGU 2014, San Francisco). Also, the change in Hg inputs to the North Atlantic required see a 4 pM change over the top 1000 m of the whole basin exceeds total global Hg mined over the same period by a large margin (see Horowitz et al. (2014) for global estimates of Hg mined since 1850).

Line 109: Please add reference Amos et al., 2012.

Response: This reference has been added.

Line 139-140: Is the variation by 7% a natural or an analytical one.
Response: The sentence has been modified as followed " Lyman et al. (2007; supplemental information) found that TGM measured by Tekran systems side-by- side could vary by 7.0 ± 5.3%.

Line 205: …between 4 and 6 hours a day.
Response: "a day" was added.

Line 209: GOM instead of RGM
Response: This change was made.

Lines 234-235: Previously it was said that the mist chamber collects also PBM. Thus RM instead of GOM should be mentioned here.
Response: This section has been modified to, "The mist chamber could measure RM. Using the mist chamber they showed RM concentrations in Maryland could be up to 500 pg m^{-3} and that GOM could be up to 30% of the TGM."

Line 245: Please correct the reference Huang et al., 2015, this issue. Dtto lines 248, 379, 442, 589.
Response: this has been removed and the reference for Huang and Gustin 2015 EST has been updated.

Lines 304-305 and 316-317: One of the sentences about fly ash is redundant.
Response: One sentence has been removed.

Lines 328-330: What do the authors mean with the sentence about the passivation? What do they mean with "loss of replication"? In a row of measurements there are no measurements replicated.
Response: Thank you for pointing this out. The sentence has been changed as follows, "Passivation of gold surfaces can occur (Barghigiani et al., 1991; Brosset and Iverfeldt, 1989; Gustin et al., 2011; Munthe et al., 1990; Xiao et al., 1991), and when this occurs these surfaces are no longer quantitatively collecting atmospheric Hg."
Line 334: Please delete the reference Brunke et al. Measurements at Cape Point do not provide any information about GOM concentrations. Neither is Cape Point located in tropics.
Response: This information has been removed.

Line 361: Please specify: "Breakthrough of GOM from upstream denuder…"
Response: This sentence has been rewritten as follows, "Breakthrough of GOM from the upstream denuder can result in inadvertent retention of GOM on the PBM collection surface resulting in a PBM measurement that is biased high."

Line 410: What is Aerohead?
Response: This has been clarified with the following information, "In general, the Aerohead or dry deposition sampling system (described above) showed higher deposition for GOM than that calculated using KCl-coated denuder measurements and a dry deposition model."

Lines 448-450: "..GOM compounds in the air vary seasonally and spatially " is a banality and as such does not need to be mentioned. Perhaps the stress should be laid on GOM composition.
Response: The sentence has been removed and emphasis placed on GOM composition.

Line 581: Please be specific: "..to adjust GOM measurements by Tekran in.."
Response: this sentence has been adjusted to read as follows, "Recent papers have used a 3-fold correction factor to adjust the GOM concentrations measured by the Tekran system made in the Western United States and Florida (cf. Huang and Gustin, 2015; Huang et al. 2015)."

Lines 595-596: What has GEM mentioned in the last sentence to do with GOM mentioned in the few paragraphs before? See also the general comment.
Response: We removed this sentence.

Lines 646-649: The consensus among atmospheric chemists about OH, the major atmospheric oxidant, is that it is more or less constant. Halogenes are going down because of Montreal protocol. Thus the sweeping statement about increasing oxidants is wrong. GEM concentrations are primarily dependent on emissions. I do not see any consistent hypothesis in this paragraph.

Response: The paragraph has been removed.