Answers to Reviewer #2:

We want to thank reviewer #2 for pointing out weaknesses of the presented manuscript. We improved our terminology and added a section on the total mercury burden in the atmosphere to address this review.

1) Please clarify whether the heights mentioned throughout the manuscript are referred to the height above sea level (asl) or above ground level (agl). If the height refers to als, the authors should also compare their modeling results with observations at high-altitude peaks worldwide (e.g., Mount Bachelor Observatory, USA, 2700 m asl; Storm Peak, USA< 3200 m asl; Lulin Atmospheric Background Station, Taiwan, 2862 m asl; Pic du Midi Observatory, France, 2877 m asl and Mt. Leigong, China, 2178 m asl. These observations are in the free troposphere and can be compared with the modeling results).

A: The hight levels refer to agl (above ground level) as all models use sigma-hybrid levels for the vertical coordinate. This makes it difficult to compare the model results to mountain stations. This is especially true for the global models which use quite low horizontal resolutions. Thus, we did not compare modeled concentrations against observations from mountain stations.

2) Line 480: Please clarify the mean of ‘source regions’. Are they related to anthropogenic or natural sources (GOM and PBM formation in the atmosphere)?

A: We clarified this: “… even in source regions with high anthropogenic emissions (e.g. coal fired power plants).”

3) Line 480: the citation should be Fu et al., 2016.

A: We added: “In China, PBM concentrations up to 1000 pg/m^3 and GOM concentrations up to 100 pg/m^3 have been observed, however no aircraft observations in the PBL and the lower free troposphere are available for this region. (Fu et al., 2016).”

4) The authors modeled the vertical concentrations of GEM, GOM, and PBM in the troposphere and stratosphere. Will it be possible to give the total quantity (Mg) of GEM, GOM, and PBM in the PBL, lower free troposphere, middle free troposphere, upper free troposphere and stratosphere?

A: We added a short section investigating the total atmospheric mercury burden as calculated by the four global models:

3.4 Total atmospheric mercury burden

We investigated the total atmospheric mercury burden as predicted by the four global models. We found that all models give a similar relative global mercury distribution with 53% to 55% of the TM in the northern hemisphere. Looking at the vertical distribution the models predict 22% to 34% inside the PBL, 54% to 60% in the free troposphere, and 9% to 16% in the stratosphere. However, the absolute numbers show a large variability. ECHMERIT (1800 Mg) gives the lowest total atmospheric mercury burden, followed by GEOS-Chem (3700 Mg), GLEMOS (6200 Mg) and GEM-MACH-Hg (6300 Mg) (Fig. 15). On average the models give 4500 Mg which is close to the estimate of 5300 Mg by Amos et al. (2013). The average vertical distribution in the model ensemble is PBL (1300 Mg), FT (2600 Mg), and stratosphere (600 Mg).

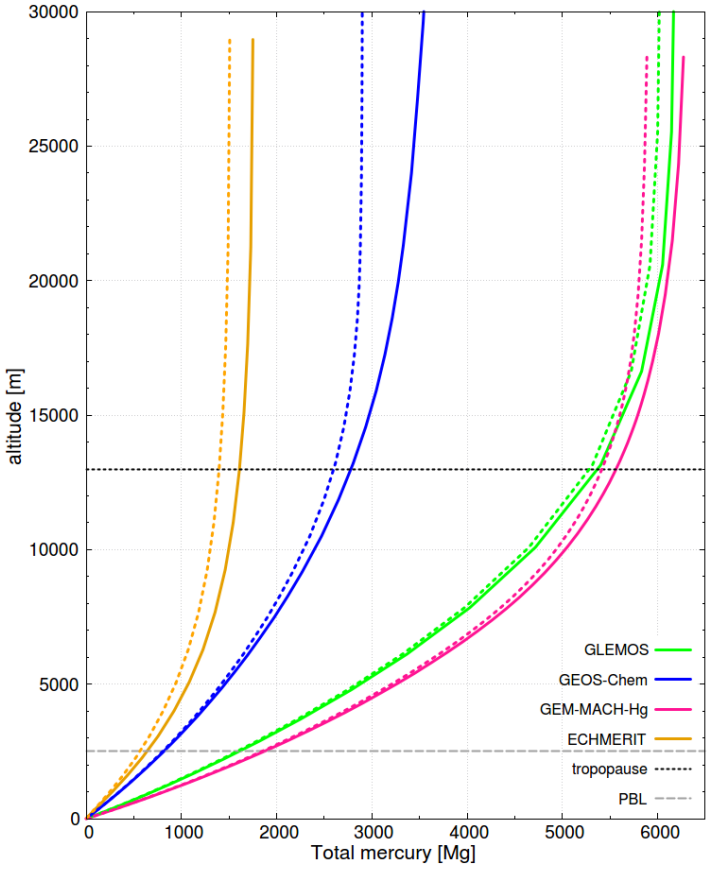

Figure 15: Global cumulative total mercury (solid) and gaseous elemental mercury (dashed line) integrated from surface to model level for each of the four global models. The model ensemble gives a total 4500 Mg of mercury in the atmosphere with 1300 Mg inside the PBL, 2600 Mg in the free troposphere, and 600 Mg in the stratosphere.

5) The atmospheric physicochemical properties over the oceans and continents are generally different. I suggest the authors should also calculate the average vertical distributions of atmospheric Hg species over oceans and continents.

A: This is a interesting idea, however we find that the paper is already extremely long and thus did not add this to the revised manuscript.

6) The measurements of GOM and PBM have many uncertainties. As mentioned the in the paper, previous studies for GOM and PBM measured utilized several different techniques. The authors should introduce the uncertainties of these observations and the effects on their comparisons.

A: The observations used for this evaluation are all based on Tekran instruments. Thus, all observations are comparable to each other. We added more discussion on the representativeness of the data and the impact of ozone and humidity for the retrieval of oxidized mercury species by the Tekran instruments.
(please see also answers to reviewer #1)

7) Line 1418, Shah et al., 2015 should be Shah et al., 2016.

A: we corrected this typo throughout the manuscript.

8) GEM, GOM, and PBM (generally the Hg bounded with fine particulates) are the three major forms of atmospheric Hg. In many parts of the paper, the authors used TM (total atmospheric Hg) and RM (reactive atmospheric Hg), and this is not completely right under some situations. Hg bounded with coarse particulates could represent a large fraction of total particulate Hg in the PBL. Also, GEM, GOM, and PBM could be transformed to other Hg species including Hg in cloud vapor, fog, etc.. These Hg species in the atmosphere sometimes represents an important fraction of atmospheric Hg. Should we define these species as RM? Have the authors taken these species into the modeling? I think this might be an important element influencing the comparisons between observations and modeling

A: We agree with the reviewer that the name RM is misleading and not the correct term to use. We now use OM (for oxidized mercury) instead throughout the manuscript.
In the manuscript, OM is defined as the sum of all oxidized mercury species including the aqueous phase. Thus, OM = TM – GEM

At the end of the introdcution we now state:
“The speciation of mercury is thus operationally defined as GEM, GOM, and PBM (Gustin et al., 2015). In the following we will address the sum of all oxidized mercury species, including mercury in the aqueous phase, as OM (oxidized mercury).”