# Responses to Reviewers' Comments

**First Measurement of Atmospheric Mercury Species in Qomolangma Nature Preserve, Tibetan Plateau, and Evidence of Transboundary Pollutant Invasion (acp-2018-806)**

Dear editor and reviewer,

We greatly appreciate the useful comments from the editor and reviewers. We think the novelty and importance of this study have been acknowledged by the reviewers. We have revised the original manuscript thoroughly based on the reviewers' comments. Detailed point by point responses are provided as follows. All the revisions have been highlighted in blue color in the manuscript. We hope the revised manuscript could meet the standard of ACP. Thanks again for your considerations.

**Anonymous Referee #2**

**General comment**

The authors present speciated Hg measurements (GEM, GOM, and PBM) at a high altitude station in Tibet near the border to Nepal. They show a pronounced concentration differences between pre-monsoon and monsoon periods and explain them by changing transport patterns encompassing different source regions, especially those in Pakistan, India, and Bangladesh. They also show influence of biomass burning. There are only a few measurements in this part of the world and, thus, they deserve to be published. Their interpretation is sound. Unfortunately, the data presentation is marred by at times awkward wording, imprecise citation of references, uninformative figure captions, etc., and thus it needs a good deal of editing. Some improvements are proposed below.

**Response**

Thanks for your comments and suggestions. We have polished the language of the manuscript, updated the citied references and revised the figure captions accordingly. Please see the revised manuscript. All the revisions have been highlighted in blue. Detailed responses to your comments are provided as follows.

**Specific comments**

**Comment #1**

Section 2.2: This section describes essentially the GOM and PBM measurement but not the measurement of GEM. Sampling time for GEM measurements has to be stated. The reason is that the GEM (with usually 5 min sampling), GOM, and PBM data are probably biased low due to problems with the internal default integration because less than 10 pg was collected for the individual analysis (Slemr et al., 2016; Ambrose, 2017). This problem is especially important at the QNNP station because only flow rates of 0.75 and 7 l(STP) $min^{-1}$ were used for GEM and GOM/PBM measurements, respectively, instead of the usual 1 and 10 l(STP) $min^{-1}$. The authors should mention the bias and assess its average magnitude using Fig. 3 of Slemr et al. (2016). This is needed when the data are compared to measurements at other sites. A definition of standard pressure and temperature would be also helpful.

**Response #1**

Thanks for your suggestion. We agree with the reviewer that a small captured Hg amount would probably lead to the biases of the measurement in QNNP. According to the method by Slemr et al. (2016), the monitoring data with low captured Hg amounts (less than 10 pg) were recalculated. In this case, the monitoring data with GOM or PBM concentrations <23.8 pg $m^{-3}$ was recalculated. The revised average concentrations increase slightly from 21.3±13.5 pg $m^{-3}$ to 21.4±13.4 pg $m^{-3}$ for GOM, and from 25.5±19.2 pg $m^{-3}$ to 25.6±19.1 pg $m^{-3}$ for PBM, respectively. All the data have been updated in the revised manuscript. The GEM sampling time, a definition of standard pressure and temperature is also provided in the revised manuscript (Line 183, 186-187, 193-199 in the revised manuscript).

**Comment #2**

Section 2.4: The use of backward trajectories for identification of the source areas seems to me to be questionable in this particular case. If I understand it properly the trajectory arrival height was set 1500 above the station, i.e. at an altitude of some 5800 m. In addition, the station is located in a very complex terrain (mountains above 8000m) with local winds due to glacier coverage. The question is how well the trajectories are representative for the air analysed at the station? Can the authors say anything about it?

**Response #2**

We fully agree that the complex terrains and local glacial winds could affect the transport of the pollutants, which might cause biases between the real situation and simulated situation. To our knowledge, existing atmospheric Hg models are not able to address the impacts of local terrains (Gustin et al., 2015), which have been evidenced in many previous studies, such as Yin et al.

(2018)'s study in central Tibetan plateau, Zhang et al. (2016)'s study in southwestern China and Fu et al. (2012)'s study in the northeast Tibetan plateau. The local terrains in all these studies have not been addressed. As suggested by another reviewer, in the revised manuscript, we have reset the arrival height of air mass to be 1000 m a.g.l. to reflect the influence of boundary layers. We do appreciate the suggestion from the reviewer, and will explore to model the impacts of local terrains on atmospheric Hg transport in QNNP in the future.

As we discussed in section 3.3.2, during ISM2 period, the trajectories and potential source region analysis could well present the influence of biomass burning from north Indian. When the source regions have frequent biomass burning (fire hotspots), the GEM and PBM concentrations in QNNP would correspondingly increase. This may indicate that the trajectories can still well represent the air analysis under complex terrain in QNNP.

**Comment #3**

Section 3.1: Averages and standard deviations should always be given with the number of measurements since only with it the significance of the differences can be determined. Are the difference of GEM, GOM and PBM concentration between PISM and ISM periods statistically significant?

**Response #3**

We have provided the number of measurements and statistical information in the revised manuscript. Please see the revised Section 3.1. Thanks for your suggestion.

**Comment #4**

Lines 278-283: Subsidence is probably only a part of the explanation; lack of precipitation could be another part.

**Response #4**

We agree that rare precipitation in QNNP could be an important reason for the high GOM in this region. As stated in the section of Methods and materials, the annual precipitation in QNNP is only 270.5 mm (Chen et al., 2016). We have provided the following information in the revised manuscript, "**Low wet deposition of GOM caused by rare precipitation in QNNP (~270mm) (Chen et al., 2016) could be another reason for the high GOM concentration (Prestbo and Gay, 2009)**". (Line 307-309 in the revised manuscript).

**Comment #5**

Table 2 claims to summarize global measurements of GEM, GOM, and PBM which is far from being true. Outside of Asia only three US sites are listed which is only a small fraction of all measurements (Sprovieri et al., 2010, 2017; Gay et al., 2013). In addition, these three US sites are not mentioned in the text. Since a comprehensive list would fill several pages I would recommend to concentrate on the measurements in Asia and for comparison with worldwide concentrations only to refer to above references.

**Response #5**

Thanks for your suggestions. Yes, we agree that it would be better to focus on the atmospheric Hg monitoring in Asia. In the revised manuscript, we have removed the monitoring sites outside of Asia from the Table 2. Please also see the revised manuscript (Line 279-280).

**Comment #6**

Section 3.2: In the text a sum of GOM and PBM is discussed but in the legend of Figure 3 symbols are declared as PBM or GOM. Please correct. The caption of Figure 3 reads as if the presented diurnal variations were representative of different periods, i.e. as averages of several days, but the reader has an impression that diurnal variations on a single day are presented. Are the diurnal variations measured on a single day (which one?) or do they represent an average of several days? If latter, how many days were averaged and what are the standard deviations or errors of the means? If averages are presented – are their differences. i.e. the average diurnal variation statistically distinguishable and different for different periods?

**Response #6**

Thanks for your suggestion.

//In the original Figure 3, GOM and PBM were displayed by using hollow and solid blue dots, respectively. We have added a new label to make it clear for readers.

//The data presented in Figure 3 is the average value of the monitoring data in each period (PISM, ISM 1-5), and this has been clarified in the caption of revised Figure 3. Number of days to calculate the average in each period is also provided. Please see the revised Figure 3.

//We agree that it would be better to provide standard deviations of different monitoring data in Figure 3. However, there are many colored lines in the original Figure 3. Hence, we have added a Figure S3 in the revised manuscript to describe the uncertainty in atmospheric Hg monitoring data. Please see Figure S3 in the revised manuscript.

**Comment #7**

Lines 500-504: Cai et al. (2007) mentions only a transport from upper level but not from stratosphere. Lelieveld et al. (2018), on the contrary, mentions a flux from the troposphere into the stratosphere in the region but not from stratosphere in the troposphere. Please refer correctly to cited literature.

**Response #7**

Thanks for your suggestion. We have revised this sentence as follows: "**As showed in other studies in the northern or eastern Tibetan Plateau, the glacier wind can pump down air masses from upper level to the surface in QNNP (Cai et al., 2007). The pump movement is remarkably efficient at transporting air masses (Zhu et al., 2006), and could bring significant amount of pollutants to QNNP.**" (Line 536-540 in the revised manuscript).

**Comment #8**

Lines 506-507: "Atmosphere Hg has been reported to have strongly declined. . ." reads as a universal downward trend. That is generally not true – the downward trend has been observed only in North America and Europe in the last 10 – 20 years. Hg concentrations decreased in the southern hemisphere between 1996 and 2004, increased between 2007 and 2012 and remained nearly constant since. The records for East Asia are mostly too short to allow a general statement – see also the cited work by Tang et al. (2018). In this discussion, I would recommend to use emission inventories and their temporal change instead of trends Hg concentrations.

**Response #8**

Thanks for your suggestions. We agree with the reviewer that the downward trend of atmospheric Hg concentrations was only observed in North America and Europe (Gay et al., 2013;Sprovieri et al., 2016). In 2016, we published a paper to describe the changes of atmospheric Hg between 2006-2015 in Tibet (Tong et al., 2016). Through the analysis of leaves of *Androsace tapete* that represent growing periods spanning the past decade, we found that there was a significant decrease of atmospheric Hg since 2010 in Tibet. Based on the reviewer's suggestion, in the revised manuscript, we have provided the description about historical change of atmospheric Hg emissions in China, as follows: "**According to the recently updated emission inventory in China (Wu et al., 2016), anthropogenic Hg emissions in China reached a peak amount of about 567 tonnes in 2011 and have decreased since then. In 2014, the anthropogenic Hg**

**emissions decreased to 530 tonnes. This was also confirmed the concentration of plant Hg from a sampling site near QNNP, which recorded the decrease of atmospheric Hg concentration in Tibet since the year of 2010 (Tong et al., 2016)**.” (Please see Line 548-553 in the revised manuscript).

**Comments #9**

Line 42: Why “unexpectedly”? Increase of GOM concentrations with altitude is predicted by some models and evidenced by observations such as at Mount Bachelor.

**Response #9**

We have deleted this word accordingly.

**Comments #10**

Line 62-63: The term “half-life” is unusual in atmospheric chemistry. “Lifetime” is usually used and clearly defined. A lifetime of 1- 2 years is somewhat long, current global models estimate GEM lifetime as short as several months. Please add references.

**Response #10**

//We have replaced “half-life” with “lifetime” in the revised manuscript.

//We have updated the information of GEM lifetime. After reviewing previous studies (Selin, 2009;Horowitz et al., 2017;Travnikov et al., 2017), we think ~0.3-1 year might be appropriate. Please see Line 63-66 in the revised manuscript.

**Comment #11**

Line 80: “invasions” reads like a military term, “flux” or “import” may be more appropriate.

**Response #11**

We have replaced the word with “import” accordingly. Thanks for the suggestion.

**Comment #12**

Lines 90-92: “The. Hg concentrations. . . originated from. . .” is incorrect because as a consequence of the long GEM lifetime nobody can say where Hg came from. “The air masses carrying high Hg concentrations originated or, better; passed over. . ..” would sound more appropriate.

**Response #12**

We have revised the sentence as follows: “**Fu et al. (2012a) report that air masses with high Hg concentrations passed over the urban and industrial areas in Western China and**

**Northern India, and influenced the atmospheric Hg concentrations in Waliguan on the northeastern edge of the Tibetan Plateau.**" (Line 92-95 in the revised manuscript).

**Comment #13**

Lines 120-122: "This monitoring site.." repeats a statement in lines 96-97. One of these statements is redundant.

**Response #13**

We have deleted this sentence from the manuscript.

**Comment #14**

Line 124: Why "comprehensive" when GEM, GOM and PBM are listed?

**Response #14**

We have deleted this word accordingly

**Comment #15**

Line 254: "significantly" – at which level of significance?

**Response #15**

This sentence has been revised as follows: "**Figure S2 shows that GEM concentrations increased significantly with the development of ISM ($p<0.001$ between ISM1 and ISM4), while decreases of GOM and PBM concentrations were observed during the study period ($p<0.001$, between ISM1 and ISM5), with decreases of 37.9% (from 20.3±7.38 pg $m^{-3}$ to 12.6 ±8.82 pg $m^{-3}$) and 48.1% (from 21.2±7.38 pg $m^{-3}$ to 11.0±5.85 pg $m^{-3}$), respectively**". Please see Line 272-277 in the revised manuscript.

**Comment #16**

Line 542-543: "air masses passed over Himalaya" is more credible than "air masses passed through Himalaya".

**Response #16**

We have replaced it accordingly (Line 587-589 in the revised manuscript).

**Comment #17**

Lines 566-567: "Atmos." Instead of "Atoms." Dtto lines 560, 572, 588, 606, 608, 734, etc. Page numbers?

**Response #17**

We have replaced this word and the whole manuscript has been checked and revisions have been made.

**Comment #18**

Figure captions contain generally too few information about what the figures display. A figure with its caption should be understandable without reading the paper.

**Response #18**

We have updated the figure captions in the revised manuscript, as follows:

"**Figure 1. Location of Qomolangma National Nature Preserve (QNNP). The red star shows the location of the monitoring station in QNNP. The red dots show the locations of two largest cities in Tibet (Lhasa and Xigaze), with the scale bars showing their distances from the QNNP.**

**Figure 2. Time series change of GEM, GOM and PBM concentration during the study period. The time series was split into a Pre-Indian Summer Monsoon (PISM) period (15 April-30 April, 2016) and 5 Indian Summer Monsoon (ISM) periods (1 May–12 May (ISM1), 13 May–4 June (ISM2), 5 June–20 June (ISM3), 21 June–10 July (ISM4), 11 July–14 August (ISM5)).**

**Figure 3. Diurnal variations of GEM, GOM and PBM concentrations during the Pre-Indian Summer Monsoon (PISM) period (15 April–30 April, 2016) and 5 Indian Summer Monsoon (ISM) periods (1 May–12 May (ISM1), 13 May–4 June (ISM2), 5 June–20 June (ISM3), 21 June–10 July (ISM4), 11 July–14 August (ISM5)). The concentrations represent the daily average values during each period.**

**Figure 4. Concentration roses of GEM, GOM and PBM from different wind directions. The length of each spoke describes the frequency of flow from the corresponding direction.**

**Figure 5. Clusters of the Back trajectories analysis from the Qomolangma National Nature Preserve (QNNP) monitoring site during the Pre-Indian Summer Monsoon (PISM) period and the 5 Indian Summer Monsoon (ISM) periods. The cluster statistics summarize the percentage of back trajectories for each cluster. The background color shading represents the global Hg emissions from anthropogenic sources (UNEP, 2013).**

**Figure 6. Potential source regions and pathways of GEM using the Potential Source Contribution Function (PSCF) method before and during the Indian Summer Monsoon (ISM). PSCF values represent the probability that a grid cell is a source of Hg.**

**Figure 7. Conceptual map of transboundary transport of atmospheric Hg in the Himalaya region. Arrows show the impacts of the Indian Summer Monsoon, upward winds and glacial winds on the transboundary transport of Hg.**

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**Comment #19**

Fig. 3: Solar radiation is difficult to discern, please correct.

**Response #19**

We have regulated the color of solar radiation in Figure 3, and please see the revised figure.

**Comment #20**

Fig. 4: What are the units of wind speed? Please add to the figure or state in the figure caption.

**Response #20**

Fig. 4 describes the frequency and concentration distribution of atmospheric Hg at different wind directions. The length of each spoke describes the frequency of atmospheric Hg concentration at certain wind direction. So, this value is irrelevant with the wind speeds.

**Comment #21**

Fig.5: It would be desirable if the caption contained some information about what the authors understand under “back trajectories analysis”.

**Response #21**

The figure caption has been revised as follows: “**Figure 5. Clusters of the Back trajectories analysis from the Qomolangma National Nature Preserve (QNNP) monitoring site during the Pre-Indian Summer Monsoon (PISM) period and the 5 Indian Summer Monsoon (ISM) periods. The cluster statistics summarize the percentage of back trajectories for each cluster. The background color shading represents the global Hg emissions from anthropogenic sources (UNEP, 2013).**” Please see Line 899-904 in the revised manuscript. All the figure captions in the manuscript have been revised.

**Comment #22**

Fig. 6 - caption: What “concepts” are shown by the maps?

**Response #22**

The caption of Fig. 6 has been revised as follows: “**Figure 6. Potential source regions and pathways of GEM using the Potential Source Contribution Function (PSCF) method before**

**and during the Indian Summer Monsoon (ISM). PSCF values represent the probability that a grid cell is a source of Hg.**"

**Comment #23**

Fig S2 – caption: What do the diagrams show? Presumably averages, medians, some percentiles – but what is what?

**Response #23**

We have added a legend in the revised Fig S2. Please see the revised manuscript.

**Comment #24**

Figure S3: The capture states "Changes of snow cover rate and diurnal index..." Why rate when the y-axis is called snow coverage? What is the diurnal index? In both cases, the percents are of what?

**Response #24**

We have replaced the "snow cover rate" with "snow coverage percentage" in the revised manuscript. To avoid the misunderstanding, we have deleted the diurnal index in the revised figure.

**Comments #25**

Figure S4: The caption does not mention the diagram.

**Response #25**

The figure caption has been revised as follows: "**Changes of snow coverage in QNNP during the study period (data from MODIS, MOD10A1)**" Please see the revised Figure S5.

**Reference**

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