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 in the manuscript. We hope the revised manuscript could meet the standard of ACP. Thanks again for your considerations.

Anonymous Referee #1

General comment

This manuscript by Huiming Lin et al. presents the first record of atmospheric mercury species (GEM, GOM, PBM) during the Indian monsoon transition period in the Qomolangma Nature Preserve, located at the southern edge of the Tibetan Plateau along the border with the Indian subcontinent. Higher GEM concentrations during the monsoon period are attributed to air masses originating from east Nepal and Bangladesh, i.e. transboundary transport of Hg. Given the projected increase in Hg emissions in South and South-East Asia, monitoring data from downwind remote sites are essential. I think that this manuscript could make a valuable addition to the literature. However, and in agreement with reviewer #2, I strongly suggest an update of the references list (imprecise citations throughout the manuscript) along with other edits (see below).

Response

Thanks for the helpful comments and suggestions. We have updated the reference list and addressed other concerns from the reviewer in the revised manuscript. A detailed point by point responses to the comments have been provided as follows.

Specific comments

Comment #1

Lines 38-40 (and throughout the manuscript): Could you please add standard deviations every time you refer to a mean concentration? Additionally, did you perform a statistical test to demonstrate that there is indeed a significant difference between ISM and non-ISM

concentrations?

Response #1

In the revised manuscript, we have added the standard deviations with the mean concentrations, and statistical test results have been added throughout the manuscript when necessary. The GEM concentrations in the ISM period were significantly higher than that in the PISM period, the GOM and PBM concentrations in the ISM period were significantly lower than those in the PISM period ($p<0.001$, ANOVA test). We have also checked the whole manuscript and added the statistical results when necessary. Please see the revised manuscript.

Comment #2

Lines 42-44: I don't think that GOM concentrations of ~20 pg/m^3 are "considerably" higher than values in other clean or polluted regions. Concentrations of 1-20 pg/m^3 are often reported at background/remote sites (e.g., Sprovieri et al. 2016) while hundreds of pg/m^3 have been reported at urban/polluted sites (e.g., Duan et al. 2017; Han et al. 2018; Guo et al. 2017; Das et al. 2016).

Response #2

Thanks for the comment. We totally agree that there are some monitoring sites with higher GOM concentrations than the measured values in QNNP. However, if we compared the GOM concentrations (35.2±18.6 pg m^{-3} during PISM period and 19.3±10.9 pg m^{-3} during ISM period) in QNNP with the monitored values from other monitoring sites in China, we found the values in QNNP were still high considering its low GEM concentrations (as shown in Table 2). For instance, the reported GOM concentrations in Beijing and Shanghai, which have been polluted by quick industrial development for a long time, were 10.1±18.8 and 21±100 pg m^{-3} (Zhang et al., 2013;Duan et al., 2017). In the background monitoring sites such as Waliguan (Fu et al., 2012) and Ailaoshan (Zhang et al., 2016), the measured GOM concentrations were 7.4±4.8 pg m^{-3} and 2.3±2.3 pg m^{-3}, respectively. However, we acknowledge that the word "considerably" could cause misunderstanding by readers, and we have revised this sentence as follows: "**Relative to the low GEM concentrations, GOM concentrations (with a mean value of 21.3±13.5 pg m^{-3}) in this region were relatively high compared with the measured values in some other regions of China.**" (Line 42-45 in the revised manuscript).

Comment #3

Lines 49-52: To me, GEM concentrations reported in this study are at the lower end of

concentrations reported in the Northern Hemisphere (Sprovieri et al. 2016). However, I do agree that international cooperation to limit Hg emissions is of utmost importance.

Response #3

We agree that, in general, the GEM concentrations in QNNP are relatively low compared with other monitored values in Northern Hemisphere (Wan et al., 2009;Fu et al., 2012;Sprovieri et al., 2016). From our study, we found that the atmospheric GEM concentrations could increase significantly from the PISM period (1.31±0.42 ng m^{-3}) to the ISM period (1.44±0.36 ng m^{-3}) in QNNP ($p<0.001$). We have revised the sentence as follows: "**The atmospheric Hg concentration in QNNP in the Indian Summer Monsoon period was significantly influenced by the transboundary Hg flows. This sets forth the need for a more specific identification of Hg sources impacting QNNP and underscores the importance of international cooperation for global Hg controls.**" (Line 50-53 in the revised manuscript).

Comment #4

Line 61: I think reference to a review paper on Hg chemistry and atmospheric cycle is more appropriate here (e.g., Selin 2009).

Response #4

We have added the reference accordingly. Please see Line 60-62 in the revised manuscript.

Comment #5

Line 63: Recent modeling studies suggest a shorter lifetime in the atmosphere: 0.3-1 year (Selin 2009; Horowitz et al. 2017).

Response #5

We have updated the information about the lifetime of GEM in the revised manuscript. Please see Line 63-66 in the revised manuscript. Thanks for your suggestions.

Comment #6

Line 64: Again, reference to Fang et al., 2009 is not appropriate here. Cite the original paper or a review paper.

Response #6

We have deleted the reference in the revised manuscript. The following references are added: (Selin, 2009;Horowitz et al., 2017;Travnikov et al., 2017).

Comment #7

Lines 73-74: Add Sprovieri et al. (2016) here.

Response #7

We have added this reference in the revised manuscript (Line 74-75).

Comment #8

Line 110: You could also briefly discuss future projections here (e.g., Pacyna et al. 2016).

Response #8

Thanks for your suggestions. We have reviewed some previous studies (Burger Chakraborty et al., 2013;Giang et al., 2015;Pacyna et al., 2016;Wu et al., 2018) and added more descriptions about the future atmospheric Hg emissions in China and India, as follows: "**China is predicted to become the largest economy in the world in the next 20-50 years, and India is predicted to catch up with the Euro area before 2030 (Pacyna et al., 2016). China is predicted to become the largest economy in the world in the next 20-50 years, and India is predicted to catch up with the Euro area before 2030 (Pacyna et al., 2016). With the implementation of control strategies, the atmospheric Hg emissions is forecasted to be about 242 tonnes in China in 2020 (Wu et al., 2018). With the implementation of control strategies, the atmospheric Hg emissions is forecasted to be about 242 tonnes in China in 2020 (Burger Chakraborty et al., 2013).**" Please see Line 112-119 in the revised manuscript.

Comment #9

Lines 120-122: I agree that this is the first study in the QNNP, but not the first one on the impact of the monsoon on Hg concentrations in Asia (e.g., Sheu et al. 2010; Yin et al. 2018; Wang et al. 2018; Zhang et al. 2014, 2016). This should be more clearly stated.

Response #9

We have revised this sentence as follows: "**To the best of our knowledge, the present work is the first study regarding Hg monitoring and source identification in the QNNP covering both the period preceding the Indian Summer Monsoon (PISM) and during the Indian Summer Monsoon (ISM).**" (Line 130-134 in the revised manuscript).

Comment #10

Section 2.2: What is the time resolution of GEM measurements (e.g., 5 or 15 minutes)? If 5 minutes, concentrations are most likely biased low and should be adjusted upwards (Slemr et al. 2016; Ambrose 2017).

Response #10

Thanks for your suggestion. In this study, the time resolution of GEM measurements is 5 minutes. We agree with the reviewer that the small captured Hg amount would probably cause the bias of the measurement. In the revised manuscript, the monitoring data with the low captured Hg (with a Hg amount lower than 10 pg) was adjusted based on the method of Slemr et al. (2016). All the data has been updated in the revised manuscript.

Comment #11

Line 202: Why did you use an arrival height of 1500 m a.g.l.? According to lines 159-161, the height of the boundary layer is ~2000 m during the day and ~ 350 m at night. This means that your back trajectories are well within the convective boundary layer during the day, but above the nocturnal boundary layer. Surface measurements at night are likely decoupled from what is happening in the residual layer and have a fairly restricted footprint. I am worried that these night-time trajectories may not be a good indication of source regions, especially given the complexity of the site. It is of common practice to use a height of 0.5 PBL.

Response #11

Thanks for your suggestion. According to your suggestions, we have reset the arrival height of air masses at 1000 m a.g.l. (0.5 PBL) in the revised manuscript. All simulations were recalculated according to the new arrival height of air masses. All the results have been updated in the revised manuscript. Please see the revised manuscript.

Comment #12

Lines 254-258: I agree with the overall PBM decrease but you should perhaps add a sentence here saying that higher PBM concentrations during ISM2 will be addressed later in the manuscript (Section 3.3.2).

Response #12

Thanks for the suggestion. The following sentence has been added into the revised manuscript, as follows: "**Reason for the higher PBM concentrations during ISM2 is discussed in Section 3.3.2.**". Please see Line 277-278 in the revised manuscript.

Comment #13

Line 261: Add Sprovieri et al. (2016) here.

Response #13

We have added the suggested reference in the revised manuscript (Line 283-284).

Comment #14

Lines 275-277: See previous comment; GOM concentrations are "at the upper end of" (and not "much higher than") values in clean regions and are not higher than concentrations reported in polluted regions (e.g., Duan et al. 2017; Han et al. 2018; Guo et al. 2017; Das et al. 2016).

Response #14

We agree with the reviewer that "much higher than" may cause misunderstanding to the readers, and we have revised it in the manuscript, as follows: "**However, despite its low GEM concentration, GOM concentration (with a value of 21.4±13.4 pg m^{-3}) in QNNP was relatively high compared with the values in the clean regions (usually lower than 10 pg m^{-3}, Table 2) or even some polluted regions of China (such as the suburban area of Beijing (10.1±18.8 pg m^{-3}), Shanghai (21±100 pg m^{-3})(Zhang et al., 2013;Duan et al., 2017)) (Table 2).** " (Line 297-301 in the revised manuscript).

Comment #15

Lines 307-311: Please add standard deviations. I would like to see something like the 95 % confidence interval for the mean on Figure 3.

Response #15

We have provided the standard divisions for all the mean values throughout the manuscript. We have added another figure in the revised manuscript (Figure S3) and 95% CI has been added, since too many colored lines are in the original Figure 3, and they are difficult to identify.

Comment #16

Line 343: Add here what's written lines 159-161 ("the height of the atmospheric boundary layer changes significantly in one day from ~350 m above ground level during the night to ~2000 m during the day").

Response #16

We have added the following sentence in Line 363-365 of the revised manuscript: "**The height of the atmospheric boundary layer could vary significantly, from ~350 m above ground level to ~2000 m in one day.**"

Comment #17

Lines 344-363: I am not really convinced by the arguments here. Do you expect higher GEM

concentrations in the afternoon to be due to local emissions? Have you checked whether you have such an increase every day, i.e., no wind direction influence? Or more or less emissions under more or less radiation? You seem to have all the data needed to perform a more thorough analysis. Could it be due to the boundary layer height? Is the boundary layer lower during the monsoon period? Is there any correlation with radiation or temperature? You could perhaps investigate the correlation between delta-GEM and delta-temperature or something like that.

Response #17

Thanks for your comments and suggestions. We totally agree with the reviewer that many factors could contribute to the diurnal variations of GEM besides the local emissions. such as wind directions, light radiation, boundary layer height, temperature and some other factors (Li et al., 2006;Selin, 2009;Horowitz et al., 2017;Travnikov et al., 2017). The Hg(0) reemission from glaciers caused by the high temperature and light radiation might be one of the potential explanation for this change (Faïn et al., 2007;Holmes et al., 2010). We have added more discussions about other possible factors which might affect the diurnal changes of GEM: "**With the increase of ambient temperature and radiation from April to August, the reemission of GEM from glaciers could increase as well. As the snow coverage in the QNNP decreased significantly from the PISM to the ISM period (Figure S4), some of the released Hg may become a source of new GEM from the initial ISM to the final stage of the ISM period. More GEM could be released due to the higher temperature and stronger radiation in the afternoon. However, some other factors such as changes in the PBL heights and in wind directions could also be partly responsible for the diurnal variations of GEM concentrations (Li et al., 2006;Selin, 2009;Horowitz et al., 2017;Travnikov et al., 2017).**" (Line 384-393 in the revised manuscript).

Comment #18

Lines 378-388: In Figure 4, could you please use something else than shades of green. It is hard to tell the difference between 1.5 ng/m3.

Response #18

Thanks for your suggestion. We have changed the color in Figure 4. Please see the revised manuscript.

Comment #19

Lines 410-412: How can you explain that GEM concentrations in air masses originating from the Tibetan Plateau were the highest?

Response #19

Thanks for the comment. We think that the high GEM concentrations in air masses originated from the Tibetan Plateau might be caused by some local residential emissions. As we can see from Figure 5(b), the cluster 2 originated from or passed through the central Tibet, China, where the majority populations in Tibet live in. The local residents usually use the biomass (i.e., yak dung) for cooking and heating. Previous studies have pointed out that the atmospheric Hg emissions from burning of yak dung could be an important Hg source in Tibet (Rhode et al., 2007;Chen et al., 2015;Xiao et al., 2015;Huang et al., 2016). We have added this information into the revised manuscript, as follows: "**GEM levels in cluster 2 (23%) were the highest (1.52 ng m^{-3}), which originated from or passed through the Tibetan Plateau. The high GEM concentrations could possibly result from the Hg emissions from the burning of yak dung (Rhode et al., 2007;Chen et al., 2015;Xiao et al., 2015;Huang et al., 2016)**" (Line 440-443 in the revised manuscript).

Comment #20

Lines 415-417: "The clusters were similar to most of the clusters during the PISM period; however, the GEM concentrations in these clusters were higher than those during the PISM period". Could you explain why?

Response #20

Thank you for the comment. As discussed in Section 3.3.2, the higher GEM concentrations during ISM 2 were likely related with the frequent fire hotspots in the source region. Large amounts of Hg were released from the biomass burning (Finley et al., 2009), leading to the higher GEM concentration in ISM 2. We have added the following sentence, "**The clusters were similar to most of the clusters during PISM period; however, the GEM concentrations in these clusters were higher than those during the PISM period, which might be caused by the large Hg emissions from frequent fires in the source region during ISM 2 (Finley et al., 2009) (Figure S5).**" (Line 447-450 in the revised manuscript).

Comment #21

Lines 452-454: What about Bangladesh? Additionally, you don't really explain why GEM

concentrations increase during the ISM period.

Response #21

//Thanks for the comments. We have carefully reviewed the recent publications about atmospheric Hg emission and pollutions in Bangladesh from the Web of Science. However, there are very few literatures about them. Some publications have reported that the air quality in Bangladesh is very bad (Mondol et al., 2014;Islam et al., 2015;Rana et al., 2016;Ommi et al., 2017;Rahman et al., 2018). So we think it is possible that the atmospheric Hg emissions in Bangladesh might also be underestimated similar to Nepal. We have added the following sentences into the revised manuscript: "**Considering the heavy air pollutions in Nepal (Forouzanfar et al., 2015;Rupakheti et al., 2017) and Bangladesh (Mondol et al., 2014;Islam et al., 2015;Rana et al., 2016;Rahman et al., 2018), Nepal and Bangladesh might be the underestimated Hg source regions in the modeling and should be taken into consideration in further study**." (Line 485-489 in the revised manuscript).

//The discussion about the higher GEM in the ISM is provided in Line 437-439, as follows: "**During the ISM period (Figure 5b-5f), the transport pathways of atmospheric Hg changed signally with the onset of the monsoon and differed strongly from the PISM period.**". We think that frequent fires in the source regions could be an important cause.

Comment #22

Line 464: Could you please add the dates for ISM2 here and/or add ISM2 in Figure S3?

Response #22

Thanks for the suggestions. The information has been provided in the revised figure.

Comment #23

Lines 464-466: Large amounts of PBM "may have been released". In this section and throughout the manuscript, please use the conditional tense to express conjectures/hypotheses.

Response #23

Thanks for your suggestion. Revisions have been made accordingly (Line 499-501).

Comment #24

Line 471: The discussion is about PBM here, not GOM. Remove reference to GOM.

Response #24

We have removed GOM information from the manuscript. Thanks.

Comment #25

Line 478: Can you explain this high value? Where did the air masses come from?

Response #25

Thanks for your comment and suggestion. We checked the trajectory of the high value, and the trajectory passed through the north of India. This sentence has been revised as follows: "**During the whole monitoring period, the highest GEM concentration reached 3.74 ng m^{-3} (with trajectories passing through the north of India), ~2.5 times higher than the average concentration in the Northern Hemisphere (~1.5-1.7 ng m^{-3}) (Lindberg et al., 2007;Slemr et al., 2015;Venter et al., 2015).**" (Line 512-515).

Comment #26

Lines 484: As mentioned above, 1.3 ng/m^3 is at the low end of GEM concentrations reported in the Northern Hemisphere. I agree that there is indeed an influence from South Asia, but concentrations on the QNNP are still fairly low during the PISM. I feel like you should slightly nuance your position.

Response #26

We agree that, in general, the GEM concentrations in QNNP are relatively low compared with other monitored values in the background regions of Northern Hemisphere. We have revised the sentence as follows: "**Compared with the ISM period, the GEM concentrations in the PISM period were significantly lower, with a value of 1.31±0.42 ng m^{-3}. This value during PISM is not high compared with other background monitoring data in the Northern Hemisphere**." (Line 518-521).

Comment #27

Lines 487-495: Could you possibly add a comparison between PISM and ISM periods in Figure 7? This comparison is the core of your manuscript.

Response #27

We have added a comparison between PISM and ISM periods, as follow: "**During the ISM period, the transboundary transport of atmospheric Hg could be strengthened by both monsoon and glacial winds. However, this effect seems to be weaker during the PISM period.**" (Line 530-532 in the revised manuscript).

Comment #28

Line 503: “significant” rather than “considerable”.

Response #28

We have corrected the word accordingly. Please see Line 538-540 in the revised manuscript.

Comment #29

Line 507: Not true everywhere (e.g., Martin et al. 2017).

Response #29

We have revised the sentence as follows: “**Atmospheric Hg concentration has been reported to have continuously declined (~1–2% y^{-1}) at the monitoring sites in North America and Europe from 1990 to present (Zhang et al., 2016b).** ”. Please see Line 542-544 in the revised manuscript.

Comment #30

Line 516: Do you know if India, Nepal and Bangladesh have ratified the Minamata Convention on Hg? Check here: http://mercuryconvention.org/Countries/Parties/tabid/3428/language/enUS/Default.aspx. Hg emissions are projected to increase in India (Pacyna et al. 2016), what about Nepal and Bangladesh? You can perhaps strengthen the discussion here.

Response #30

Thanks for the information.

//We have reviewed the information in the website carefully. We found that India, Nepal and Bangladesh have signed the convention, but only India has ratified the convention so far.

//As we replied in Response #21, we have reviewed the recent publications carefully on Web of Science, but there are very few publications about the Hg emission and Hg concentration in Bangladesh and Nepal. Some publications have reported that the air quality in Bangladesh is very bad (Mondol et al., 2014;Islam et al., 2015;Rana et al., 2016;Ommi et al., 2017;Rahman et al., 2018). So we think it is possible that the atmospheric Hg emissions in Bangladesh might also be underestimated, similar to Nepal. We have added this information into the revised manuscript. Please see Line 562-563.

Comment #31

Lines 526-528: Is there a significant difference?

Response #31

Yes, in the manuscript, we have performed the statistical analysis to compare the atmospheric Hg concentrations between PISM and ISM periods, and the results show that there are significant differences between two periods ($p<0.001$). We have added the statistical information in the revised manuscript. Please see Line 265-271.

Comment #32

Lines 544-546: Again, concentrations reported here during PISM are at the low end of concentrations reported in the Northern Hemisphere. Additionally, concentrations are similar to those recently reported at Nam Co station on the Tibetan Plateau (Yin et al. 2018).

Respond #32

We agree with the reviewer and we have deleted this sentence from the manuscript.

Comment #33

Figure 1: I assume that the red star within the QNNP is the location of the monitoring station. What about the two other red stars (Lhasa and Xigaze)? Do they represent cities and potential emissions? You should perhaps use a different type of star (monitoring site vs. cities) and make it clear in the caption.

Response #33

Yes, Lhasa is the largest city in Tibet, and Xigaze is the second. We have marked these two places in a different symbol to help readers understand the locations of QNNP. Please see the revised Figure 1. Thanks for your suggestions.

Comment #34

Figure 2: Could you please add on this Figure the different periods (ISM1-5) you're referring to in Table 1?

Response #34

We have highlighted different ISM periods in the revised Figure 2.

Comment #35

Figure 3: I can't read the yellow axis, it is too bright. Please use another color. Additionally, what do you mean by GOM or PBM? Is this GOM, PBM, or the sum of the two? It is hard to see the dots and the diurnal cycle for GOM/PBM.

Response #35

We have adjusted the color of yellow axis. The hollow and solid dots (in blue) represent the

monitored GOM and PBM concentrations, respectively. We have clarified this point in the revised Figure 3.

Comment #36

Figure 4: Which one is GOM, which one is PBM? Add a), b), c) on the Figure and caption.

Response #36

We have added the labels accordingly. Thanks.

Comment #37

Figure 6: Could you please explain in the caption what these values are? Probability of air passes passing through these regions?

Response #37

The figure caption has been revised as: "**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.**" Please see the revised Figure 6.

Comment #38

Table 2: I think you can focus on Asian sites or refer to Figure 1 in Yin et al. (2018). The concentration reported for the Nam Co station is incorrect (Yin et al. 2018).

Response #38

In the revised manuscript, we removed the atmospheric Hg monitoring sites out of Asia, which is also suggested by another reviewer. Please see Line 279-280 and the revised Table 2.

Comment #39

Figure S4: Could you please add PISM, ISM1-5? Additionally, instead of April-August, is it possible to plot fires during PISM, ISM1-5? It would make it easier to identify whether fires are indeed more frequent in the area of interest during ISM2.

Response #39

We have highlighted different ISM periods in Figure S5 and added the fire information as well. Thanks for the suggestions.

Reference:

Burger Chakraborty, L., Qureshi, A., Vadenbo, C., and Hellweg, S.: Anthropogenic mercury flows in

India and impacts of emission controls, Environmental science & technology, 47, 8105-8113, 2013.
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