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

Chen, P., Gao, Y., Lee, A. T., Cering, L., Shi, K., and Clark, S. G.: Human–carnivore coexistence in Qomolangma (Mt. Everest) Nature Reserve, China: Patterns and compensation, Biological conservation, 197, 18-26, 2016.

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Gay, D. A., Schmeltz, D., Prestbo, E., Olson, M., Sharac, T., and Tordon, R.: The Atmospheric Mercury Network: measurement and initial examination of an ongoing atmospheric mercury record across North America, Atmospheric Chemistry and Physics, 13, 11339-11349,'D'O'I:' 10.5194/acp-13-11339-2013, 2013.

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Selin, N. E.: Global biogeochemical cycling of mercury: a review, Annual Review of Environment and Resources, 34, 43-63, 2009.

Slemr, F., Weigelt, A., Ebinghaus, R., Kock, H. H., Bödewadt, J., Brenninkmeijer, C. A., Rauthe-Schöch, A., Weber, S., Hermann, M., and Becker, J.: Atmospheric mercury measurements onboard the CARIBIC passenger aircraft, Atmospheric Measurement Techniques, 9, 2291-2302, 2016.

Sprovieri, F., Pirrone, N., Bencardino, M., D'Amore, F., Carbone, F., Cinnirella, S., Mannarino, V., Landis, M., Ebinghaus, R., and Weigelt, A.: Atmospheric mercury concentrations observed at ground-based monitoring sites globally distributed in the framework of the GMOS network, Atmospheric Chemistry and Physics, 16, 11915-11935, 2016.

Tong, Y., Yin, X., Lin, H., Wang, H., Deng, C., Chen, L., Li, J., Zhang, W., Schauer, J. J., and Kang, S.: Recent Decline of Atmospheric Mercury Recorded by Androsace tapete on the Tibetan Plateau, Environmental science & technology, 50, 13224-13231, 2016.

Travnikov, O., Angot, H., Artaxo, P., Bencardino, M., Bieser, J., D'Amore, F., Dastoor, A., Simone, F. D., Diéguez, M. d. C., and Dommergue, A.: Multi-model study of mercury dispersion in the atmosphere: atmospheric processes and model evaluation, Atmospheric Chemistry and Physics, 17, 5271-5295, 2017.

Wu, Q., Wang, S., Li, G., Liang, S., Lin, C.-J., Wang, Y., Cai, S., Liu, K., and Hao, J.: Temporal trend and spatial distribution of speciated atmospheric mercury emissions in China during 1978–2014, Environmental science & technology, 50, 13428-13435, 2016.

Yin, X., Kang, S., Foy, B. d., Ma, Y., Tong, Y., Zhang, W., Wang, X., Zhang, G., and Zhang, Q.: Multi-year monitoring of atmospheric total gaseous mercury at a remote high-altitude site (Nam Co, 4730 m asl) in the inland Tibetan Plateau region, Atmospheric Chemistry and Physics, 18, 10557-10574, 2018.

Zhang, H., Fu, X., Lin, C., Wang, X., and Feng, X.: Observation and analysis of speciated atmospheric mercury in Shangri-La, Tibetan Plateau, China, Atmos. Chem. Phys, 15, 653-665,

2015.
Zhang, H., Fu, X., Lin, C.-J., Shang, L., Zhang, Y., Feng, X., and Lin, C.: Monsoon-facilitated characteristics and transport of atmospheric mercury at a high-altitude background site in southwestern China, Atmospheric Chemistry & Physics, 16, 2016.

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

241 are very few literatures about them. Some publications have reported that the air quality in
242 Bangladesh is very bad (Mondol et al., 2014;Islam et al., 2015;Rana et al., 2016;Ommi et al.,
243 2017;Rahman et al., 2018). So we think it is possible that the atmospheric Hg emissions in
244 Bangladesh might also be underestimated similar to Nepal. We have added the following sentences
245 into the revised manuscript: "**Considering the heavy air pollutions in Nepal (Forouzanfar et al.,**
246 **2015;Rupakheti et al., 2017) and Bangladesh (Mondol et al., 2014;Islam et al., 2015;Rana et**
247 **al., 2016;Rahman et al., 2018), Nepal and Bangladesh might be the underestimated Hg source**
248 **regions in the modeling and should be taken into consideration in further study**." (Line 485-
249 489 in the revised manuscript).
250 //The discussion about the higher GEM in the ISM is provided in Line 437-439, as follows:
251 "**During the ISM period (Figure 5b-5f), the transport pathways of atmospheric Hg changed**
252 **signally with the onset of the monsoon and differed strongly from the PISM period.**". We think
253 that frequent fires in the source regions could be an important cause.
254 **Comment #22**
255 Line 464: Could you please add the dates for ISM2 here and/or add ISM2 in Figure S3?
256 **Response #22**
257 Thanks for the suggestions. The information has been provided in the revised figure.
258 **Comment #23**
259 Lines 464-466: Large amounts of PBM "may have been released". In this section and throughout
260 the manuscript, please use the conditional tense to express conjectures/hypotheses.
261 **Response #23**
262 Thanks for your suggestion. Revisions have been made accordingly (Line 499-501).
263 **Comment #24**
264 Line 471: The discussion is about PBM here, not GOM. Remove reference to GOM.
265 **Response #24**
266 We have removed GOM information from the manuscript. Thanks.
267 **Comment #25**
268 Line 478: Can you explain this high value? Where did the air masses come from?
269 **Response #25**
270 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:

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First Measurement of Atmospheric Mercury Species in Qomolangma Nature Preserve, Tibetan Plateau, and Evidence of Transboundary Pollutant Invasion

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Abstract

Located in the world's 'Third Pole' and a remote region connecting the Indian Ocean plate and the Eurasian plate, Qomolangma National Nature Preserve (QNNP) is an ideal region to study the long-range transport of atmospheric pollutants. In this study, gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM) and particle-bound mercury (PBM) were continuously measured during the Indian monsoon transition period in QNNP. A slight increase in GEM concentration was observed from the preceding the Indian Summer Monsoon period (1.31±0.42 ng m^{-3}) to the Indian Summer Monsoon period (1.44±0.36 ng m^{-3}), while significant decreases were observed in GOM and PBM concentrations, with concentrations decreasing from 35.2±18.6 to 19.3±10.9 pg m^{-3} ($p<0.001$) for GOM and from 30.5±12.5 to 24.9±19.8 pg m^{-3} ($p<0.001$) for PBM. A unique daily pattern of GEM concentration in QNNP was observed, with a peak value before sunrise and a low value at noon. Relative to the low GEM concentrations, GOM concentrations (with a mean value of 21.4±13.4 pg m^{-3}, n=1239) in this region were relatively high compared with the measured values in some other regions of China. A cluster analysis indicated that the air masses transported to QNNP changed significantly at different stages of the monsoon, and the major potential Hg sources shifted from north India and west Nepal to east Nepal and Bangladesh. Because there is a large area covered in glaciers in QNNP, local glacier winds could increase transboundary transport of pollutants and transport polluted air masses to the Tibetan Plateau. The atmospheric Hg concentration in QNNP in the Indian Summer Monsoon period was influenced by 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.

Keywords

Indian summer monsoon; atmospheric mercury; trans-boundary transport; glacier winds; Qomolangma National Nature Preserve

1. Introduction

Understanding atmospheric mercury (Hg) concentrations in remote regions is vital to understand the global atmospheric Hg cycling processes (Zhang et al., 2016a;Angot

et al., 2016;AMAP/UNEP, 2013). Generally, atmospheric Hg can be divided into three major types: gaseous elemental Hg (GEM), gaseous oxidized Hg (GOM) and particle-bound Hg (PBM) (Selin, 2009). Over 95% of atmospheric Hg exists in the form of GEM (Ebinghaus et al., 2002;Huang et al., 2014). Due to its stable chemical properties and long lifetime in the atmosphere (approximately 0.3 to 1 year), GEM can be transported over long distances (Horowitz et al., 2017;Travnikov et al., 2017;Selin, 2009). In contrast, GOM and PBM could deposit quickly from the atmosphere, exposing local environments to significant impacts (Lindberg and Stratton, 1998;Seigneur et al., 2006;Lynam et al., 2014). To understand the global and regional cycling of atmospheric Hg, different Hg monitoring networks and sites have been established in recent decades, such as the Atmospheric Mercury Network (AMNet) (Gay et al., 2013) and Global Mercury Observation System (GMOS), which contains over 40 ground-based monitoring stations distributed in the world (Sprovieri et al., 2016). Generally, atmospheric Hg background concentrations range between 1.5 to 1.7 in the northern hemisphere and 1.1 to 1.3 ng m^{-3} in the southern hemisphere (Lindberg et al., 2007;Slemr et al., 2015;Venter et al., 2015;Sprovieri et al., 2016). However, existing studies are still far from sufficient to obtain a full understanding of long-range Hg transport because of insufficient monitoring data in remote and less-populated regions (Zhang et al., 2015a;Fu et al., 2012a).

The trans-boundary and long-range transport of pollutants have attracted considerable attentions in the northeastern and southeastern regions of the Tibetan Plateau (Yang et al., 2018;Li et al., 2016;Zhang et al., 2015b;Pokhrel et al., 2016). The transboundary flows of atmospheric pollutants to the Tibetan Plateau have been identified for pollutants such as persistent organic pollutants and black carbon (Yang et al., 2018;Li et al., 2016;Zhang et al., 2015b;Pokhrel et al., 2016). It was reported that smoke from biomass burning in the Indian subcontinent could pass over natural barrier of the Himalaya (Wang et al., 2015;Pokhrel et al., 2016). HCHs, DDTs and PCBs were all found to have their highest concentrations in the southeast Tibetan Plateau during the monsoon season (Wang et al., 2018). Similar conditions have also occurred for black carbon (Li et al., 2016). However, studies of the trans-boundary transport of Hg on the

Tibetan Plateau are still limited. The existing Hg monitoring data is affected to varying extents by local emission sources (Fu et al., 2012a; Zhang et al., 2015; Zhang et al., 2016). 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. At Shangri-La, located on the southeastern edge of the Tibetan Plateau, the atmospheric Hg sources were reported to be Southeast Asia, India and mainland China (Zhang et al., 2015a). Nevertheless, studies are still lacking on trans-boundary transport of Hg in the Qomolangma National Nature Preserve (QNNP), which directly connects the Indian Subcontinent and Eurasia. The detailed pollutant transport pathways and seasonal or daily patterns of atmospheric Hg concentrations in this region are still not clear.

QNNP, located on the southern edge of the Tibetan Plateau, is considered one of the world's cleanest regions (Qiu, 2008). With an average altitude of ~4,500 m a.s.l., QNNP is a remote region with sparse human population and rare industries (Qiu, 2008; Yao et al., 2012b; Li et al., 2016). However, it is surrounded by two large potential pollution sources: the populated and developed eastern China region, which has experienced about 30 years of rapid industrial development, and South Asian developing countries (e.g., India, Nepal, and Bangladesh), which have also been developing rapidly in recent years (Streets et al., 2011;Zhang et al., 2015b;Yang et al., 2018). China and India are reported as the largest coal consumers in the world (BP Statistical Review of World Energy 2018), and coal combustion is the largest source of atmospheric Hg emissions globally, accounting for ~86% of Hg emissions (Chen et al., 2016a). 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). The rapidly growing economies have led to rapid increases in energy demands and hence increasing domestic Hg emissions (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). However, atmospheric Hg emissions in India are expected to increase to about 540 tonnes Hg by 2020 (Burger Chakraborty et al., 2013). Because

QNNP is located on the pathway of air mass transport due to the Indian Summer Monsoon (ISM) (Li et al., 2016), meteorological conditions in QNNP vary significantly during the monsoon transition period (Wang et al., 2001). The monthly average precipitation can range from less than 50 mm in the non-ISM period to 950 mm in the ISM period (Panthi et al., 2015). In addition to the monsoon, the glacial coverage in QNNP is approximately 2,710 km^2 (Nie et al., 2010). Glacier winds could therefore have direct effects on the local pollutant transport because downslope glacier winds can transport polluted air from the upper levels to the land surface (Cai et al., 2007). The atmosphere in QNNP is therefore vulnerable to surrounding pollution sources (Li et al., 2016;Xu et al., 2009).

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). We performed continuous measurements of GEM, GOM and PBM concentrations for 2 weeks during the onset of the monsoon and for 3.5 months during the monsoon itself. To identify the detailed sources, we combined the real-time Hg monitoring data with a backward trajectory analysis, clustering analysis and potential source contribution function (PSCF) analysis. We further discuss the effects of local glacier winds, caused by the large spatial extent of QNNP glaciers, on the trans-boundary transport of pollutants. This combined monitoring and modeling study could help researchers and government managers to better understand the global Hg cycling processes and potential impacts from the rapidly developing countries in South Asia on the atmospheric Hg concentrations in QNNP.

2. Materials and methods

2.1 Atmospheric Hg monitoring site

Atmospheric Hg monitoring was conducted at the “Atmospheric and Environmental Comprehensive Observation and Research Station, Chinese Academy of Sciences on Mt. Qomolangma” (latitude: 28°21’54” N, longitude: 86°56’53” E) in QNNP, at an altitude of 4,276 m a.s.l. (Figure 1). In QNNP, Mt. Qomolangma spreads from east to the west along the border between the Indian subcontinent and the Tibetan Plateau

(Figure 1). Due to its high altitude, QNNP is naturally isolated from the populated regions, and only rare local Hg emission sources have been observed (AMAP/UNEP, 2013). The most populated region near this monitoring site is Tingri County (with a population density of 4 persons per km^2), located ~40 km to the southwest of the monitoring site. The average annual temperature is 2.1 °C and the total annual rainfall is 270.5 mm in QNNP (Chen et al., 2016b). QNNP is located along the air mass transport pathway of the ISM (Li et al., 2016), and the meteorological conditions in QNNP have significant variations between the PISM and ISM periods (Wang et al., 2001). During the transition period, the temperature in the Tibetan Plateau and South Asia changes from "southern warm - northern cool" to "northern warm - southern cool" (Wang et al., 2001). This reversal leads to a significant increase of diabatic heating over South Asia and the southern slope of the Tibetan Plateau (Ge et al., 2017), which further affects the wind directions and speeds. Local glacier winds could also affect the transport of air masses in QNNP. Glaciers cover ~2,710 km^2 in QNNP (Nie et al., 2010), and most of the glaciers are located on the northern slope of the mountain (Figure 1) (Bolch et al., 2012). The glacier wind is a continuous downslope wind blowing from glacier surfaces down to the foothills of the mountain throughout the day. Hence, the transport of air masses in this region is a combination of atmospheric circulation (monsoon) and local weather conditions (glacier winds). The structure of the boundary layer over QNNP is also significantly affected by glaciers (Li et al., 2006). The height of the atmospheric boundary layer follows a diurnal profile ranging from ~350 m above ground level during the night to ~2000 m during the day (Li et al., 2006).

2.2 GEM, GOM and PBM monitoring

To describe the changes of atmospheric Hg concentrations during the PISM and ISM periods, real-time continuous measurements of GEM, GOM and PBM concentrations were carried out using the Tekran 2537B, 1130 and 1135 instruments (Tekran Inc., Toronto, Canada) from 15 April, 2016 to 14 August, 2016. During the operation of the Tekran instruments, ambient air was introduced into the instrument for 60 minutes through an impactor, a KCL-coated annular denuder, and a Quartz Fiber Filter (QFF). All the Hg species were converted into Hg(0) and then measured by cold vapor atomic

fluorescence spectroscopy (CVAFS). The collected PBM and GOM were desorbed in succession to Hg(0) at the temperature of 800 °C and 500 °C, respectively. Hg-free air was used to flush the 1130 and 1135 systems to introduce the desorbed PBM and GOM into model 2537B for analysis. The GEM was collected at 5-minutes intervals. The sampling inlet was set at ~1.5 m above the instrument platform (shown in Figure S1). To mitigate the impacts of low atmospheric pressures on the pump's train, a low air sampling rate of 7 L min^{-1} for the pump model and 0.75 L min^{-1} (at a standard pressure of 1013 hPa and temperature of 273.14 K) for model 2537B was applied (Swartzendruber et al., 2009;Zhang et al., 2015a;Zhang et al., 2016a). The Tekran 2537B analyzer was calibrated automatically using the internal Hg permeation source inside the instrument every 23 h, and the internal source was calibrated before and after the monitoring by an external Hg source using a syringe. The Tekran ambient Hg analyzer has been described in more details in the previous publications (Landis et al., 2002;Rutter et al., 2008;de Foy et al., 2016). Recent studies have suggested that there may be a low bias of GOM and PBM concentrations for small sample loads of Hg(e.g. less than 10 pg) (Slemr et al., 2016;Ambrose, 2017). Hence, the monitoring data with GOM or PBM concentrations below 23.8 pg m^{-3} were recalculated by the method of Slemr et al. (2016). The updated GOM concentrations increased slightly from 21.3±13.5 pg m^{-3} to 21.4±13.4 pg m^{-3} and from 25.5±19.2 pg m^{-3} to 25.6±19.1 pg m^{-3} for PBM.

2.3 Meteorological data

Throughout the sampling period, the meteorological information was recorded using the Vantage Pro2 weather station (Davis Instruments, USA) with a 5-minute resolution. The monitored parameters included the temperature (with a precision of 0.1°C), relative humidity (with a precision of 1%), wind speed (with a precision of 0.1 m s^{-1}), wind direction (with a precision of 1°), air pressure (with a precision of 0.1 hPa), solar radiation (with a precision of 1 W m^{-2}) and UV index (with a precision of 0.1 MEDs). The snow cover data was obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS) instrument on board the Terra and Aqua satellites (MOD10A1, Hall et al., 2010) with a daily 0.05° resolution.

2.4 Backward trajectory simulation

To identify the atmospheric Hg sources, the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model was applied to perform a backward trajectory simulation (Stein et al., 2015;Chai et al., 2016;Chai et al., 2017;Hurst and Davis, 2017). The HYSPLIT model, known as a complete and mature system for modeling air parcel trajectories of complex pollutant dispersion and deposition, was developed by the US National Oceanic and Atmospheric Administration (NOAA). Global Data Assimilation System (GDAS) data with 1°×1° latitude and longitude horizontal spatial resolution and 23 vertical levels at 6-hour intervals was used for the backward trajectory simulation. All the trajectory arrival heights were set to 1000 m above ground level. Every backward trajectory was simulated for 72 hours in 6-hour intervals, and the air mass transport regions covered China, Nepal, India, Pakistan and majority of west Asia. Backward trajectories during the whole monitoring period were calculated, and cluster analysis was carried out to identify the Hg transport pathways. The cluster statistics summarize the percentage of back trajectories in each cluster, and the average GEM concentrations are linked with each cluster. The clustering algorithm utilized in this study is based on Ward's hierarchical method (Ward Jr, 1963), which minimizes angular distances between corresponding coordinates of the individual trajectories. By averaging similar or identical pathways from existing air mass pathways to the receptor site, clusters can help identify the mean transport pathways of air masses and provide the primary directions of pollutants transported to the measurement site.

The Potential Source Contribution Function (PSCF) model is a hybrid receptor model using the calculated backward trajectories to estimate the contributions of different emission sources in upwind regions and has been applied in many previous studies (Kaiser et al., 2007;Fu et al., 2012b;Kim et al., 2005;Zhang et al., 2013). The PSCF calculation is made based on counting the trajectory segments that terminate within each cell to determine the values for the grid cells in the study domain (Ashbaugh et al., 1985). In this study, the PSCF model was used to identify the possible sources of atmospheric GEM. The study domain was separated as i × j cells. Then, the PSCF value for the ij^{th} cell is defined as follows:

$$PSCF_{ij} = \frac{M_{ij}}{N_{ij}}$$

where N_{ij} is the total number of endpoints that fall into ij^{th} cell during the whole simulation period, and M_{ij} is the number of endpoints for the same cell that correspond to GEM concentrations higher than a set criterion. In this study, PSCF values were calculated based on the average GEM concentration during the whole sampling campaign. The PSCF value stands for the conditional probability that the GEM concentration at the measurement site is larger than the average GEM concentration if the parcel passes through the ij^{th} cell before it reaches the measurement site.

To account for and reduce the uncertainty due to low values of N_{ij}, the PSCF values were scaled by an arbitrary weighting function W_{ij} (Polissar et al., 1999). While the total number of the endpoints in a cell (N_{ij}) is less than ~three times the average value of the end points for each cell, the weighting function will decrease the PSCF values. In this study, W_{ij} was set using the following piecewise function:

$$W_{ij} = \begin{cases} 1.00 & N_{ij} > 3\,N_{ave} \\ 0.70 & 3\,N_{ave} > N_{ij} > 1.5N_{ave} \\ 0.42 & 1.5N_{ave} > N_{ij} > N_{ave} \\ 0.05 & N_{ave} > N_{ij} \end{cases}$$

We used the PSCF analysis to evalutate the effects of biomass burning regions using the MODIS fire data. MODIS fire spots data from 1 April 2016 to 31 August 2016 was obtained from the Fire Information for Resource Management System (FIRMS) operated by the National Aeronautics and Space Administration (NASA) of the United States (Giglio et al., 2003; Davies et al., 2004).

3. Results and discussion

3.1 Comparisons of atmospheric Hg concentrations between PISM and ISM

The GEM, GOM and PBM concentrations at the sampling site were 1.42±0.37 ng m^{-3} (n=15180), 21.4±13.4 pg m^{-3} (n=1239) and 25.6±19.1 pg m^{-3} (n=1237), respectively, during the whole study period (Figure 2 and Table 1). GEM accounted for over 95% of all the atmospheric Hg species. Figure S2 shows a comparison of the GEM, GOM and PBM concentrations during the PISM and ISM periods. During the PISM period, the average GEM, GOM and PBM concentrations were 1.31±0.42 ng m^{-3} (n=2001),

35.2±18.6 pg m^{-3} (n=167), and 30.5±12.5 pg m^{-3} (n=168), respectively, while during the ISM period, the average GEM, GOM and PBM concentrations were 1.44±0.36 ng m^{-3} (n=13179), 19.3±10.9 pg m^{-3} (n=1072), and 24.9±19.8 pg m^{-3} (n=1069), respectively. The concentrations of GEM, GOM and PBM are statistically significant different ($p<0.001$) between PISM and ISM period. We further compared the Hg concentrations at different ISM stages. 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. Reason for the higher PBM concentrations during ISM2 is discussed in Section 3.3.2.

Table 2 summarizes GEM, GOM and PBM concentrations and diurnal variations of GEM measured by the Tekran system in some previous studies in Asia. Generally, the GEM concentration in the QNNP was approaching the reported values in the Northern Hemisphere (~1.5 to 1.7 ng m^{-3}) and was higher than those in the Southern Hemisphere (~1.1 to 1.3 ng m^{-3}) (Lindberg et al., 2007;Slemr et al., 2015;Venter et al., 2015;Sprovieri et al., 2016). Among the global Hg monitoring sites, the EvK2CNR monitoring site on the southern slope of the Tibetan Plateau, Nepal, is the nearest station (at a straight-line distance of approximately 50 km) from the monitoring site in this study (Gratz et al., 2013). The average GEM concentration at EvK2CNR (1.2±0.2 ng m^{-3}, from Nov. 2011-Apr. 2012) was slightly lower than that in the QNNP (1.31±0.42 ng m^{-3} during the PISM period and 1.44±0.36 ng m^{-3} during the ISM period). Compared with the Hg concentration in Nam Co station (Yin et al., 2018) in the central Tibetan plateau (1.33±0.24 ng m^{-3}), the GEM concentration in QNNP was higher during the ISM period. Compared with Hg concentrations observed at China's background stations and rural regions (e.g., Waliguan Baseline Observatory (1.98±0.98 ng m^{-3}) (Fu et al., 2012a), Ailaoshan Mountain National Natural Reserve (2.09±0.63 ng m^{-3}) (Zhang et al., 2016a), and Shangri-La Baseline Observatory in Yunnan province (2.55±0.73 ng m^{-3}) (Zhang et al., 2015a)), the average GEM concentration in the QNNP was lower.

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). One possible explanation for the high GOM concentration is the strong subsidence in QNNP. The subsidence of the free troposphere would bring GOM-enriched air masses to the surface layer (Faïn et al., 2009), resulting in the observed high surface GOM levels (Weiss-Penzias et al., 2009). In QNNP, with the wide distribution of glaciers, glacier winds could bring the upper air masses to the land surface layer (Song et al., 2007), which could further strengthen the subsidence movement. Low wet deposition rate of GOM caused by the rare precipitation in QNNP (~270mm) (Chen et al., 2016c) could be another reason for the high GOM concentrations (Prestbo and Gay, 2009).

The increases of GEM concentrations during the ISM period could indicate the impacts of trans-boundary transport, which has been confirmed by previous studies (Fu et al., 2012a;Zhang et al., 2016a). The deposition of GEM from the atmosphere to the land surface is difficult, and GEM has a much longer residence time than the other Hg species (Horowitz et al., 2017;Travnikov et al., 2017;Selin, 2009). At Ailaoshan in Yunnan province (Zhang et al., 2016a), a higher TGM concentration during the ISM period (2.22±0.58 ng m^{-3}) than the PISM period (1.99±0.66 ng m^{-3}) was also observed. The TGM concentration during the ISM period (2.00±0.77 ng m^{-3}) was also higher than that during the PISM period (1.83±0.78 ng m^{-3}) at Waliguan station in the northeastern Tibetan Plateau (Fu et al., 2012a). In contrast to GEM, the GOM and PBM levels during the ISM period were lower than the monitored values during the PISM period (Figure S2 and Table 2). In previous studies, the PBM concentration in the Kathmandu Valley was lower during the monsoon period (with a value of 120.5±105.9 pg m^{-3}) than the pre-monsoon (with a value of 1855.4±780.8 pg m^{-3}) and post-monsoon period (with a value of 237.6±199.4 pg m^{-3}) (Guo et al., 2017). In India, PBM concentrations during the monsoon period (with a value of 158±34 pg m^{-3}) were lower than those in the non-monsoon season (with a value of 231±51 pg m^{-3}) (Das et al., 2016). This fact could be

possibly attributed to precipitation increases brought by the monsoon, which further causes wet depositions of PBM from atmosphere. During the ISM period, the precipitation could increase by up to 25% in the South Asia and Tibetan Plateau (Ji et al., 2011).

3.2 Diurnal variation of atmospheric Hg species in QNNP

During the PISM period, all the atmospheric Hg species showed clear diurnal patterns (Figure 3 and Figure S3). For GEM, the minimum concentrations usually occurred at ~12 p.m. (0.84±0.11 ng m^{-3}, UTC +6 time), while maximum values occurred before dawn (1.98±0.51 ng m^{-3} at ~5:30 a.m.). During the afternoon, GEM concentration increased consistently and reached a peak at sunrise (with a value of 1.98 ng m^{-3}). Unlike the daily GEM changes, GOM and PBM concentrations usually reached maximum concentrations from ~10:00 a.m. to ~4:00 p.m. in the day, and the concentrations remained relative stable for the rest of the day. During the ISM period, the diurnal variation of atmospheric Hg species was less pronounced compared to the values in the PISM period. At different stages of the ISM period, the diurnal pattern was also different. The GEM diurnal variation value (peak value minus lowest value in the same period) decreased over time, from 1.03 ng m^{-3} during the initial ISM period to 0.43 ng m^{-3} during the final ISM period. For GEM concentrations during the ISM period, the minimum values all occurred at ~2:00 p.m., and the maximum values were observed at ~6:00 a.m. After sunrise, GEM concentrations decreased continuously to lower values at noon.

Compared with diurnal profiles of GEM from previous studies, the diurnal tendency in QNNP is unique (shown in Table 2). For the sampling sites in other studies, the highest GEM concentrations were usually observed during the daytime (Nair et al., 2012;Fu et al., 2008;Mukherjee et al., 2009;Karthik et al., 2017;Jen et al., 2014). Kellerhals et al. (2003) reported that the majority of monitoring sites in CAMNet have a common pattern with the maximum concentrations around noon and minimum concentrations before sunrise. Compared to other observation stations and considering QNNP as a remote region with high altitude, sparse population and rare industries, the observed results here may indicate a simple mechanism of variation in GEM

concentration without the complex effect of human activities. Previous studies suggested that the planetary boundary layer (PBL) could have significant effects on the concentrations of atmospheric pollutants near the ground (Han et al., 2009;Tie et al., 2007;Quan et al., 2013). With a large glacier coverage (~2,710 km^2), the structure of the boundary layer over QNNP was significantly affected by glacier winds (Li et al., 2006). The local PBL may be subject to impacts from the glacier-covered environment and have a significant diurnal variation. The height of the atmospheric boundary layer could vary significantly from ~350 m above ground level to ~2000 m in one day (Li et al., 2006). Following sunrise, with the strengthening of the glacier wind, a strong convection current starts to grow in the troposphere, and the stock of GEM in the near-ground atmosphere is depleted quickly, leading to the quick decrease in concentrations. In contrast, after sunset, with the weakening of the glacier wind, the nocturnal stable boundary layer takes a dominate position controlling the surface layer, and its height is relatively low (Li et al., 2006), which could lead to increases in GEM concentrations.

Comparing the diurnal variations between the PISM and ISM period, the atmospheric Hg concentrations have almost the same pattern of variations. However, the magnitude of the variation during the ISM period is lower relative to the PISM period, and the variation becomes even smaller in the later stages of the ISM (Figure 3). The GEM concentration usually peaked at ~5 a.m. - 6 a.m. in both PISM and ISM periods. While the peak GEM concentrations were almost at the same level in the whole period, the decreasing diurnal variations were mainly due to the increasing GEM concentrations in the afternoon. The increased GEM concentrations in the afternoon may indicate new GEM sources in the ISM period. One possible source of GEM in the afternoon might be Hg(0) reemission from the glaciers. Holmes et al. (2010) reported that snow-covered land could be a reservoir for the conversion of oxidized Hg to Hg(0) under sunlight, and approximately 60% of the Hg deposited to snow cover could eventually be reemitted to the air. A shorter reservoir lifetime for deposited Hg in snowpack was also reported when temperature rises (Faïn et al., 2007). With the increase of ambient temperature and radiation from April to August, the reemission of GEM from the glaciers could increase as well. As the snow coverage in the QNNP decreased

387 significantly from the PISM to the ISM period (Figure S4), some of the released Hg
388 may become a source of new GEM from the initial ISM to the final stage of the ISM
389 period. More Hg(0) could be released due to the higher temperature and stronger
390 radiation in the afternoon. However, some other factors such as changes in the PBL
391 heights and in wind directions could also be partly responsible for the diurnal variations
392 of GEM concentrations (Horowitz et al., 2017;Travnikov et al., 2017;Selin, 2009;Li et
393 al., 2006).

394 **3.3 Source identification for atmospheric Hg in the QNNP**

395 **3.3.1 Wind direction dependence of Hg concentrations**

396 Figure 4 shows the concentration roses of GEM, GOM and PBM at the sampling site
397 during the PISM and ISM period, respectively. All concentrations of the three species
398 have a strong dependence on the wind directions. During the PISM period, the
399 predominant wind directions with Hg masses are northeast and southwest. Wind from
400 the northeast of QNNP originates from and/or passes through other parts of China. The
401 southwest wind, which is the dominant direction and contains the largest amount of Hg,
402 potentially brought air masses from India and Nepal to QNNP. During the ISM period,
403 the predominant wind directions with Hg changed to the south and northeast.
404 Considering the transport rates of species Hg concentrations (length of sector) from
405 different directions, both directions may have greatly contributed to the Hg
406 concentration in QNNP, while the air masses from south brought relatively larger
407 amounts of GOM and PBM.

408 Relatively low GEM concentrations (<1.5 ng m^{-3}) were observed in most of the
409 samples (80.0%) of air masses in the predominant Hg-transport direction (from
410 southwest to west) during the PISM period, which is due to the control of westerlies.
411 With high wind speeds (Table 1) and coming from Central Asia, the westerlies are the
412 predominant wind containing low pollutant levels that spread in the QNNP during the
413 PISM period (Kotlia et al., 2015). Relatively high GEM concentrations (>1.5 ng m^{-3})
414 were found in 92.4% of the samples for the predominant Hg direction during the ISM
415 period under the control of the monsoon (Kotlia et al., 2015), which might indicate that
416 the transported air masses are coming from polluted regions. GOM and PBM had

similar patterns under the control of the westerlies and monsoon during the PISM and ISM period, respectively.

3.3.2 Air mass back trajectories analysis

To further quantify the contributions of different sources to GEM concentrations, an air mass back trajectory simulation and trajectory cluster analyses were applied in this study. Figure 5 provides the trajectory clusters of GEM during the PISM and ISM periods. According to the total spatial variation index, all the trajectories in different periods were grouped into 3-6 clusters. During the PISM period (Figure 5a), GEM concentration from cluster 1 (with a frequency of 12%) was the highest (1.32 ng m^{-3}), which originated from or passed through central Asia and northern India. Cluster 2 (30%) and cluster 4 (17%) represent air masses that pass through northern India and northwestern Nepal. According to the local Hg emission inventory (AMAP/UNEP, 2013), Hg in this air mass most likely originated from central Pakistan and northern India. Cluster 3 (41%) represents the air masses that originated from or passed through different cities in northern India. Based on the previous atmospheric Hg emission inventories (Simone et al., 2016;AMAP/UNEP, 2013), Hg emissions in west Asia and central Asia are not significant. Based on a combination of the pathway analysis, emission inventory and GEM concentrations during the PISM period, almost all the GEM transported by air masses to QNNP was from northern India and passed through Nepal.

During the ISM period (Figure 5b-5f), the transport pathways of atmospheric Hg changed significantly with the onset of the monsoon and differed strongly from the PISM period. During the ISM1 period (Figure 5b), the onset of the ISM was under development, leading to scattered clusters. 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 (Xiao et al., 2015;Chen et al., 2015;Rhode et al., 2007;Huang et al., 2016). Cluster 1 (17%) and cluster 3 (60%) represent the pollutant coming from Nepal, and the trajectory is relatively short. During the ISM2 period, all the clusters originated from or passed through central Asia, northern India and northwestern Nepal (Figure 5c).

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 ISM2 (Finley et al., 2009) (Figure S5). During the ISM3 period (Figure 5d), most of the clusters moved from west to south of QNNP. Cluster 4 (1.56 ng m^{-3}, 46%) represents the pollutant coming from Bangladesh and passing through southeastern Nepal. Cluster 3 (1.54 ng m^{-3}, 40%) originated from or passed through central Nepal. The share of air masses coming from central Asia, northern India and northwestern Nepal dropped to approximately 14%. During the ISM4 period (Figure 5e), the clusters moved further west to Bangladesh and eastern India. Except for cluster 1 (5%), the other clusters originated from or passed through Bangladesh, eastern India and northeastern Nepal. The condition during the ISM5 period was almost the same as the ISM4 period: most of the pollutants were coming from Bangladesh and eastern India and passed through southeastern Nepal.

PSCF models were also applied to identify potential sources by combining the backward trajectory simulations and Hg monitoring concentrations. Figure 6 shows the regional contributions of GEM emission sources during the PISM period and ISM period (ISM1-5). During the PISM period (Figure 6a), most of the Hg sources were in Pakistan, northern India and central Nepal (Zhang et al., 2015a). The QNNP was most likely impacted by the Hg emissions in Karachi, Lahore (Pakistan), New Delhi, Uttar Pradesh (India), Katmandu and Pokhara (Nepal), all of which are large urban regions with intensive industrial activities. With the development of the ISM, the potential sources gradually shifted from western Nepal to eastern Nepal and Bangladesh (Figure 6b-f). The PSCF analysis indicated that the air masses could have transboundary transport events from Pakistan, India, Nepal and Bangladesh to QNNP.

Atmospheric Hg clusters during both the PISM and ISM periods indicated that the air masses, which originated from or passed through northern India and Nepal, would make great contributions to the Hg concentration in the QNNP. Northern India and Nepal were also identified as potential source regions for QNNP. Clusters 2-4 of the PISM period represent the air masses from outside China, and they show that over 88%

of the GEM in QNNP was transported from outside China during the PISM period. During ISM2-5 the period, over 95% of the GEM was transported to QNNP from outside China. Meanwhile, the GEM concentration increased by 10% from the PISM to ISM period according to the site monitoring data, indicating the increasing amount of transported GEM. According to the UNEP Hg emission inventory (AMAP/UNEP, 2013), northern India is an important Hg source which might be responsible for the trans-boundary transportation of Hg to China (Figure 5), and the growing emissions in India are related to the rapidly growing economy and increasing usage of fossil fuels (Sharma, 2003). Considering the heavy air pollutions in Nepal (Rupakheti et al., 2017;Forouzanfar et al., 2015) and in Bangladesh (Islam et al., 2015;Rahman et al., 2018;Rana et al., 2016;Mondol et al., 2014), Nepal and Bangladesh might be underestimated Hg source regions in the modeling and should be taken into consideration in further study.

Under the control of the ISM during the ISM2 period, the high PBM concentration may be related to the biomass burning in the source region. According to the PSCF analysis, northern India and Nepal are the potential source regions during the ISM2 period. The source identification by back trajectory simulation and trajectory cluster analyses also indicated that northern India and Nepal are in the air mass transport trajectory that would transport Hg to QNNP. Finley et al. (2009) reported that PBM concentrations could be associated with Hg emissions from wildfire events. One possible cause of the observed high PBM concentration is the frequent fire events that occurred during the ISM2 period in the air masses trajectory. Figure S4 shows the fire hotspots observed by MODIS from April to August 2016. During the ISM2 period, frequent fire hotspots were identified in the source region, and large amounts of PBM may have been released into the atmosphere from biomass burning (Finley et al., 2009). The transport of those air masses with enriched PBM was controlled by the ISM and intensified by glacier winds. The transport of polluted air to QNNP resulted in the outburst of PBM concentration during the ISM2 period. During the PISM period, although the number of fire hotspots was much higher, most of the fire hotspots locations were not in the potential source region (Figure 6a and Figure S4), resulting in

the low PBM concentration observed.

3.4 Implications from this study

At a high altitude and located in the deep southern Tibetan Plateau, QNNP is isolated from anthropogenic perturbations and industrial activities, and this area was thought to be shielded from pollutant inputs from South Asia. However, our results show that the Hg concentration in this region is not as low as previously expected. 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 to 1.7 ng m^{-3}) (Lindberg et al., 2007;Slemr et al., 2015;Venter et al., 2015). The average GEM concentration in the middle stage of the ISM was 1.56 ng m^{-3}, which is inside the average range of observed Northern Hemisphere GEM concentrations. 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.

We now recognize that trans-boundary transportation is an important mechanism that can influence Hg distribution in this region. In particular, the air masses transported to QNNP might be primary under the control of mesoscale ISM drivers and intensified by regional glacier winds (Figure 7). From the PISM to ISM periods, the warm center gradually shifts northwestward from low latitudes to the QNNP (Wang et al., 2001;Ge et al., 2017), and the South Asian High moves onto the Tibetan Plateau and maintains a strong upper-level divergence and upward motion. The upward motion makes the air masses cross the high-altitude Himalayan Mountains and move to mainland China (Xu et al., 2009; Bonasoni et al., 2010). During the ISM period, the transboundary transport of atmospheric Hg is strengthened by both monsoon and glacial winds. However, this effect seems to be weaker during the PISM period. The transboundary-transported air masses can be pumped down right after crossing Mt. Qomolangma due to the control of the regionally unique wind transportation mode, the glacier wind. Hence, in addition to the monsoon, the trans-boundary transport of Hg could also be intensified by regional glacier winds, leading to the increases of atmospheric Hg in this region. 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.

In 2013, the Minamata Convention on Mercury was developed to control global Hg pollution. Atmospheric Hg 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). Under the Convention, a National Implementation Plan on Mercury Control has been developed in China to fulfill the commitment to control and reduce Hg emissions (World Bank, 2016). Average GEM concentrations in East China decreased from 2.68±1.07 ng m^{-3} in 2014 to 1.60±0.56 ng m^{-3} in 2016 (Tang et al., 2018). 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 in the concentration of plant Hg from a sampling site near QNNP, which recorded the decrease of atmospheric Hg concentrations in Tibet since the year of 2010 (Tong et al., 2016). However, the source identity analysis in QNNP indicates that foreign regions of China were the main contributor responsible for the observed pollutants (accounting for 95% of the whole trajectory during the main ISM period). This result indicates that the Hg concentration in QNNP could hardly benefit from China's efforts toward Hg reductions. South Asian developing countries (e.g., India, Nepal, and Bangladesh) (Streets et al., 2011; Zhang et al., 2015b; Yang et al., 2018) should be the key to controlling atmospheric Hg concentrations in QNNP. Hg emissions in India were estimated to be approximately 310 tonnes in 2010 and are predicted to rise to 540 tonnes in 2020 (Burger Chakraborty et al., 2013). India, Nepal and Bangladesh all have signed the Minamata Convention, however, only the Indian government has ratified the convention so far. It is urgent for those countries to take immediate actions to reduce Hg emissions, which is crucial to reducing atmospheric Hg concentrations in QNNP.

4. Conclusions

A comprehensive investigation of the concentrations, origin and transport of GEM, GOM and PBM was made in QNNP, a remote, high-altitude station located at the boundary between the Indian subcontinent and the Tibetan Plateau and in the transport pathway of the Indian Summer Monsoon from South Asia to the Tibetan Plateau. The average GEM concentration (1.31±0.42 ng m^{-3}) during the PISM period was lower than that during the ISM period (1.44±0.36 ng m^{-3}). The average GOM and PBM concentrations during the PISM period were higher than those during the ISM period, which might be related to the increasing wet depositions during the ISM period. The average GOM concentration was higher than in most rural areas in the US and China. The GEM concentration had a significant diurnal variation pattern in QNNP, with the maximum GEM concentration observed before sunrise and a sharp decrease after sunrise until noon. The magnitude of the diurnal variation declined from April to August, which could be related to the re-emission of Hg from snow cover and change of planetary boundary layer.

According to the backward trajectory analysis and cluster analysis, most of the air masses with high GEM concentrations in QNNP originated from or passed through Bangladesh, northern India and central Nepal. With the PSCF analysis, we found that Pakistan, northern India and Nepal are potential source regions during the PISM period, and Bangladesh, north India, Nepal were identified as outbound potential sources during the ISM period. During the ISM period, the air masses were able to cross the high-altitude Himalayan Mountains with the help of the ISM. Once the air masses passed over the Himalayas, they could be brought into the surface layer and transported to QNNP by the all-day-long downslope glacier wind. Because Hg is easily transported long distances via the atmosphere, the nations in South Asia must work together to develop and apply appropriate pollutant-reduction strategies to reduce atmospheric Hg emissions.

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Figure captions

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.

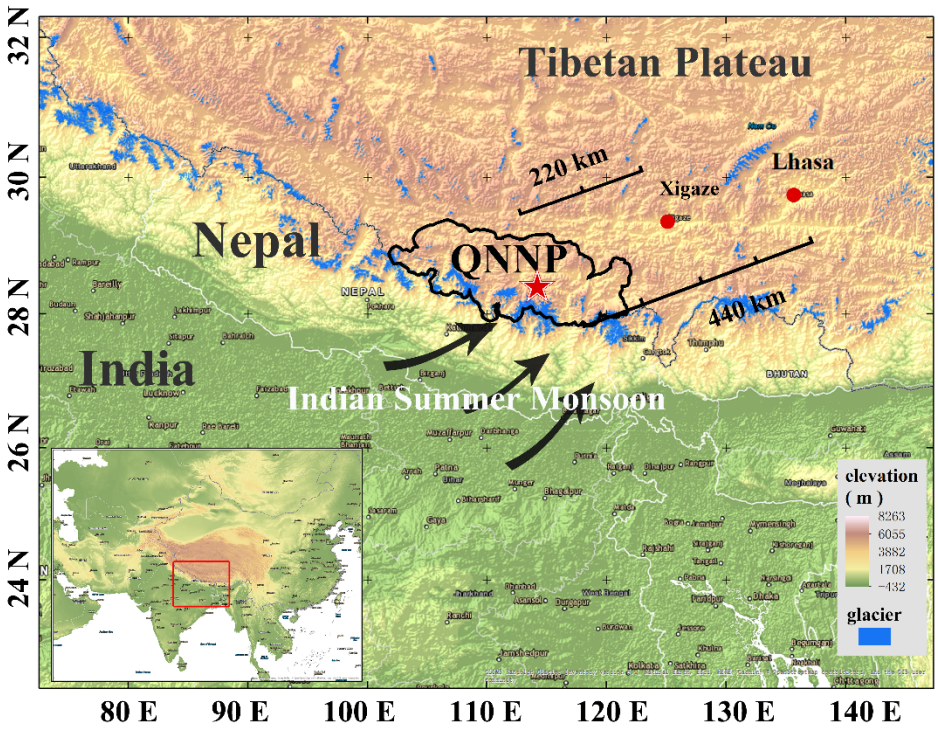
Figure 4. Concentration roses of GEM, GOM and PBM from different wind directions. The length of each spoke describes the frequency of flow from the corresponding direction.

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 (AMAP/UNEP, 2013).

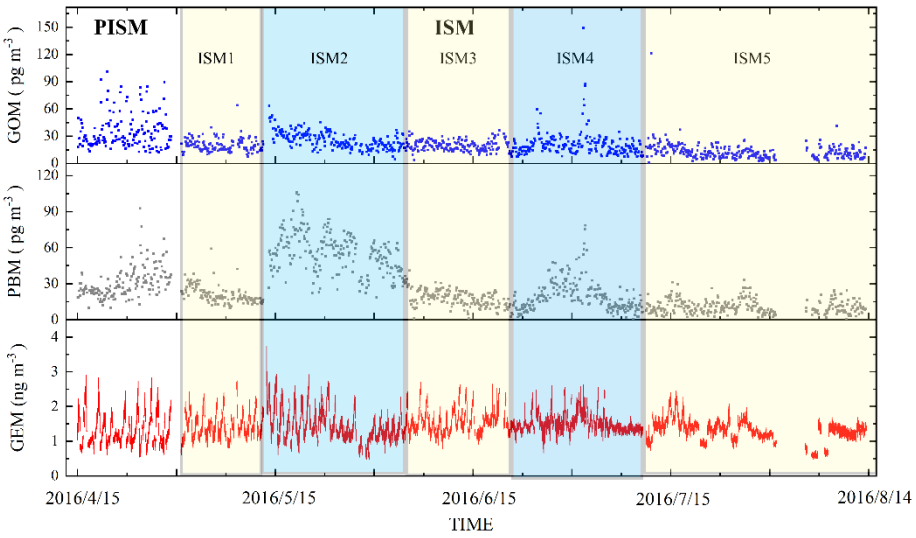
Figure 6. Potential source regions and pathways of GEM using the Potential Source Contribution Function (PSCF) method before and during the Indian Summer Monsoon (ISM). PSCF values represent the probability that a grid cell is a source of Hg.

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.

Figure 1


Tibetan Plateau
220 km
Xigaze
Lhasa
Nepal
QNNP
NEPAL
440 km
India
BHUTAN
Indian Summer Monsoon
elevation
(m)
8263
6055
3882
1708
−432
glacier
32 N
30 N
28 N
26 N
24 N
80 E
90 E
100 E
110 E
120 E
130 E
140 E

913 **Figure 2**



914

915

Figure 3


PISM period
Ism1 period
Ism2 period
Ism3 period
Ism4 period
Ism5 period
Solar Radiation (W m-2)
Wind Speed (m s-1)
Relative Humidity
Wind Direction
GOM (pg m-3)
PBM (pg m-3)
GEM (ng m-3)

Figure 4

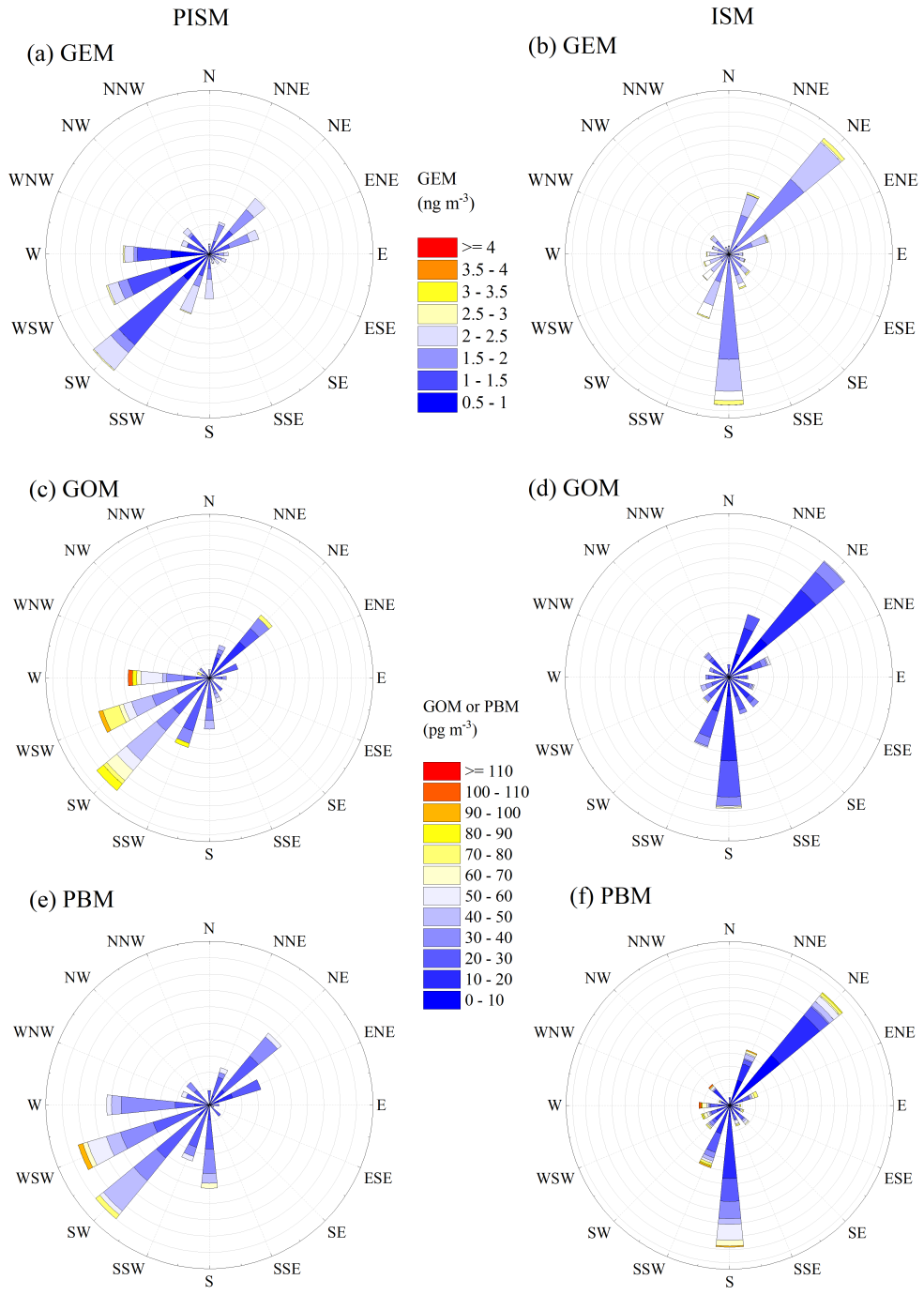

PISM
ISM
(a) GEM
(b) GEM
(c) GOM
(d) GOM
(e) PBM
(f) PBM
N
NNE
NE
ENE
E
ESE
SE
SSE
S
SSW
SW
WSW
W
WNW
NW
NNW
GEM (ng m-3)
>= 4
3.5 - 4
3 - 3.5
2.5 - 3
2 - 2.5
1.5 - 2
1 - 1.5
0.5 - 1
GOM or PBM (pg m-3)
>= 110
100 - 110
90 - 100
80 - 90
70 - 80
60 - 70
50 - 60
40 - 50
30 - 40
20 - 30
10 - 20
0 - 10

Figure 5

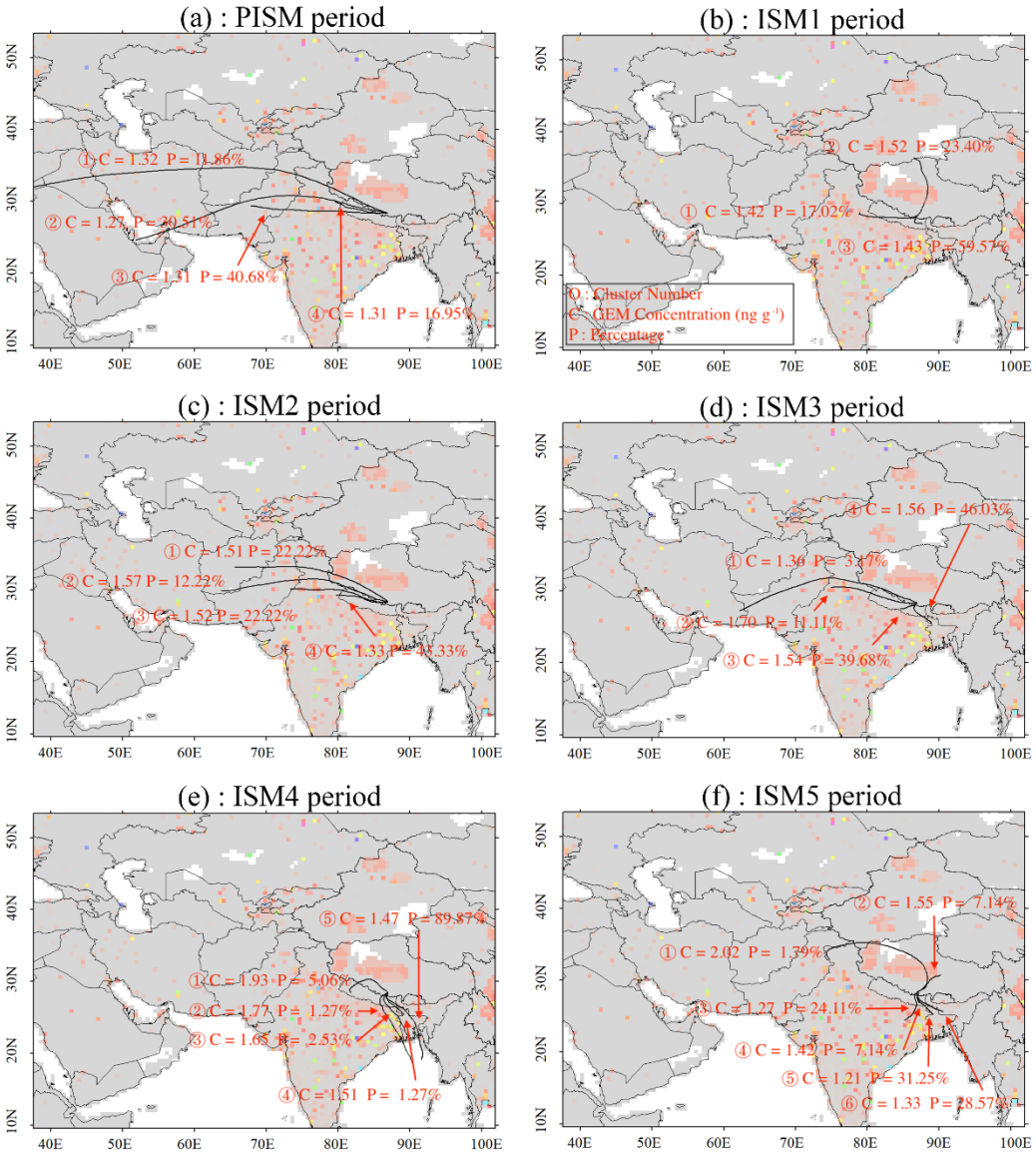

(a) : PISM period
① C = 1.32 P = 11.86%
② C = 1.27 P = 30.51%
③ C = 1.31 P = 40.68%
④ C = 1.31 P = 16.95%
(b) : ISM1 period
② C = 1.52 P = 23.40%
① C = 1.42 P = 17.02%
③ C = 1.43 P = 59.57%
O : Cluster Number
C : GEM Concentration (ng g-1)
P : Percentage
(c) : ISM2 period
① C = 1.51 P = 22.22%
② C = 1.57 P = 12.22%
③ C = 1.52 P = 22.22%
④ C = 1.33 P = 43.33%
(d) : ISM3 period
④ C = 1.56 P = 46.03%
① C = 1.36 P = 3.17%
② C = 1.70 P = 11.11%
③ C = 1.54 P = 39.68%
(e) : ISM4 period
⑤ C = 1.47 P = 89.87%
① C = 1.93 P = 5.06%
② C = 1.77 P = 1.27%
③ C = 1.65 P = 2.53%
④ C = 1.51 P = 1.27%
(f) : ISM5 period
② C = 1.55 P = 7.14%
① C = 2.02 P = 1.59%
③ C = 1.27 P = 24.11%
④ C = 1.42 P = 7.14%
⑤ C = 1.21 P = 31.25%
⑥ C = 1.33 P = 28.57%
50N
40N
30N
20N
10N
40E
50E
60E
70E
80E
90E
100E

Figure 6

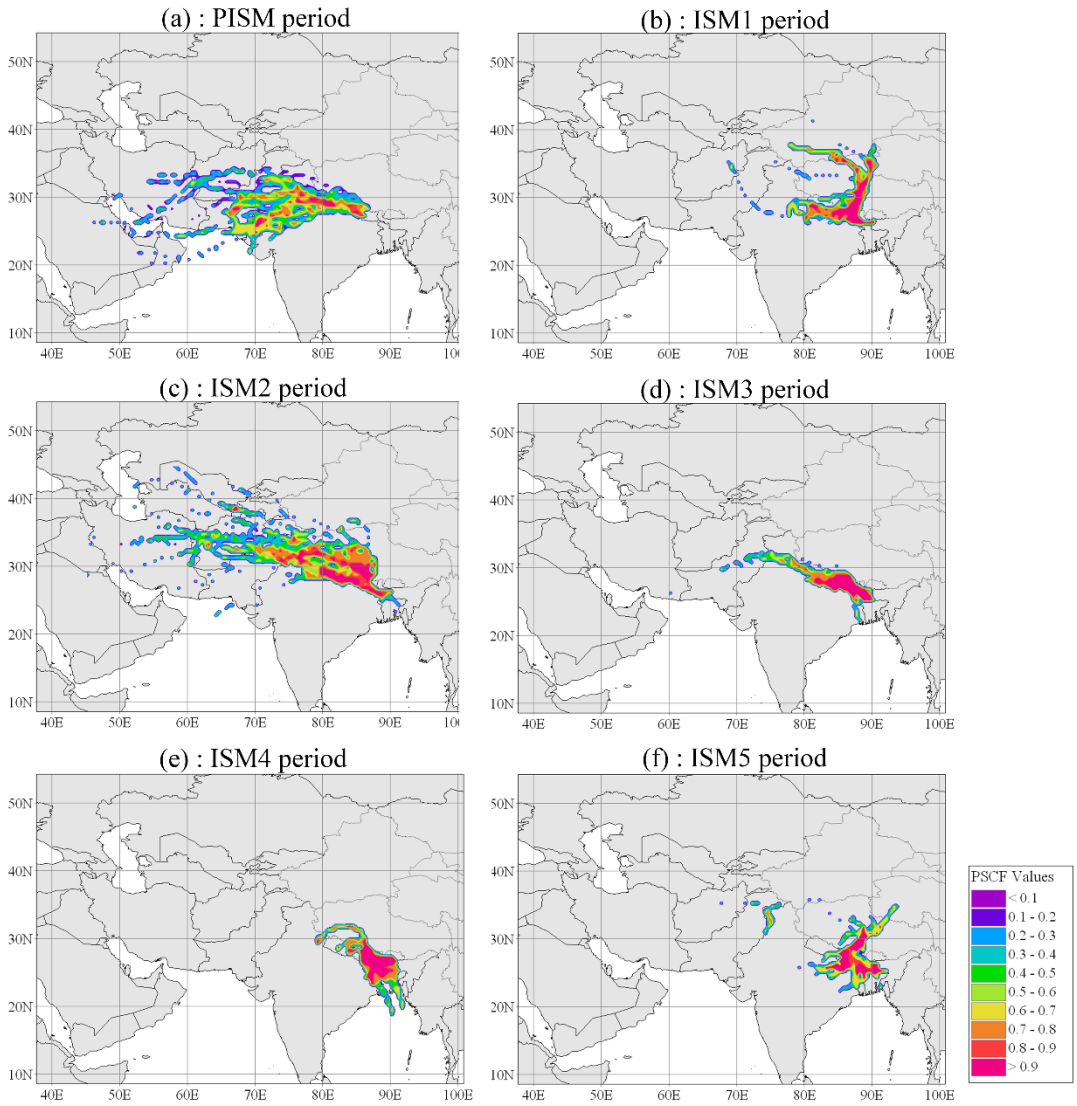
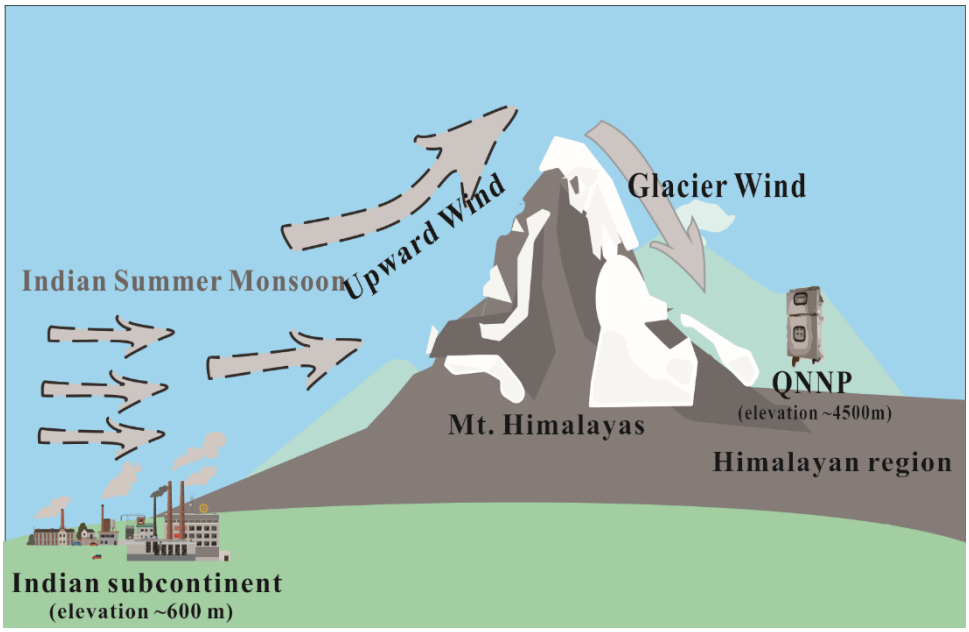

(a) : PISM period
(b) : ISM1 period
(c) : ISM2 period
(d) : ISM3 period
(e) : ISM4 period
(f) : ISM5 period
50N
40N
30N
20N
10N
40E
50E
60E
70E
80E
90E
100E
PSCF Values
< 0.1
0.1 - 0.2
0.2 - 0.3
0.3 - 0.4
0.4 - 0.5
0.5 - 0.6
0.6 - 0.7
0.7 - 0.8
0.8 - 0.9
> 0.9

Figure 7


Upward Wind
Glacier Wind
Indian Summer Monsoon
QNNP
(elevation ~4500m)
Mt. Himalayas
Himalayan region
Indian subcontinent
(elevation ~600 m)

933

934 **Table 1. Statistical metrics of GEM, GOM, PBM and meteorological variables at the**

935 **Qomolangma National Nature Preserve**

Period	Statistical	T (℃)	RH(%)	WS(m s^{-1})	GEM (ng m^{-3})	GOM (pg m^{-3})	PBM (pg m^{-3})
PISM	Minimum	-5.6	1	0	0.54	11.9	9.8
	1st Qu.	1.6	11	1.8	0.99	21.7	22.3
	Median	6.4	25	3.6	1.19	29.5	26.8
	Mean	6.1	33	4.1	1.31	35.2	30.4
	3rd Qu.	11.2	53	6.3	1.58	42.8	36.0
	Maximum	16.3	89	13.9	2.91	101.3	92.6
ISM1	Min	-3.8	9	0	0.15	7.5	9.5
	1st Qu.	1.6	33	1.3	1.20	15.2	17.0
	Median	5.6	49	2.2	1.38	19.2	19.2
	Mean	5.6	50	2.7	1.44	20.3	21.2
	3rd Qu.	9.8	65	3.6	1.63	24.1	24.5
	Max	15.7	91	10.3	2.74	64.0	59.1
ISM2	Min	-1.3	3	0	0.47	4.4	12.7
	1st Qu.	4.1	30	1.3	1.14	18.6	40.4
	Median	8.5	48	2.2	1.35	23.9	54.8
	Mean	8.8	46	3.0	1.45	25.5	53.4
	3rd Qu.	13.7	64	4	1.68	31.3	64.9
	Max	19.6	87	11.2	3.74	63.4	106.3
ISM3	Min	2.6	26	0	0.78	3.6	1.1
	1st Qu.	8.1	44	1.3	1.33	14.7	12.7
	Median	11.8	58	2.7	1.51	19.0	17.2
	Mean	12.0	58	2.9	1.56	19.3	16.9
	3rd Qu.	15.6	73	4	1.72	23.3	21.3
	Max	21.8	92	9.9	2.70	36.6	31.3
ISM4	Min	6.0	25	0	0.66	7.1	0.5
	1st Qu.	9.3	43	1.3	1.35	13.2	10.9
	Median	12.1	61	2.7	1.46	18.1	17.4
	Mean	13.0	58	2.9	1.51	21.1	20.0
	3rd Qu.	16.6	72	3.6	1.61	24.9	26.1
	Max	22.7	90	9.9	2.62	149.1	78.6
ISM5	Min	2.2	18	0	0.48	1.1	0.3
	1st Qu.	8.3	59	0.9	1.17	7.6	6.6
	Median	10.7	75	2.2	1.35	11.0	9.8
	Mean	11.4	72	2.3	1.32	12.6	11.0
	3rd Qu.	14.1	86	3.1	1.49	16.2	14.3
	Max	22.9	96	9.4	2.45	121.3	33.2

936

Table 2. Comparison of atmospheric Hg concentrations and diurnal variation of GEM at QNNP with measurements from previous studies

Location	Elevation	Classification	Time period	GEM/(*TGM*)	GOM	PBM	GEM diurnal variation (Local time/GEM Conc.)			reference
				(ng m^{-3})	(pg m^{-3})	(pg m^{-3})	peak	valley	variation value	
Mt. Waliguan, China	3816	remote	Sep 2007-Sep 2008	*(1.98±0.98)*	7.4±4.8	19.4±18.1	6/2.3	14/1.94	0.36	(Fu et al., 2012a)
Mt. Leigong, China	2178	remote	May 2008-May 2009	2.80±1.51	-	-	14/2.99	5/2.52	0.47	(Fu et al., 2010)
Mt. Gongga, China	1640	remote	May 2005-July 2006	*(3.98)*	-	-	11/4.45	2/3.55	0.90	(Fu et al., 2008)
Kodaikanal, India	2343	rural	Nov 2012-Sep 2013	*(1.53±0.21)*	-	-	16/1.66	7/1.43	0.23	(Karthik et al., 2017)
EvK2CNR, Nepal	5050	remote	Nov 2011-Apr 2012	*(1.2±0.2)*			18/1.3	6/1.1	0.1	(Gratz et al., 2013)
Shangri-La, China	3580	remote	Nov 2009-Nov 2010	*(2.51±0.73)*	8.22±7.9	38.32±31.26	17/2.48	6/1.71	0.77	(Zhang et al., 2015a)
Miyun, China	220	rural	Dec 2008-Nov 2009	3.22±1.74	10.1±18.8	98.2±112.7	20/3.40	10/3.00	0.40	(Zhang et al., 2013)
Penghu Islands, China	25	coastal	Mar 2011-Jan 2012	*(3.17±1.17)*	-	-	11/3.48	1/2.87	0.61	(Jen et al., 2014)
Shanghai, China	17	Urban	Jun 2014–Dec 2014	4.19±9.13	21±100	197±877				(Duan et al., 2017)
Namco, China	5300	remote	Nov 2014-Mar 2015	1.33±0.24	-	-				(Yin et al., 2018)
ALS, China	2450	remote	May 2011-May 2012	*(2.09±0.63)*	2.3±2.3	31.3±28.4	-	-	-	(Feng and Fu, 2016)
QNNP, China (this study)	4267	remote	Apr 2016-Aug 2016	1.42±0.37	21.4±13.4	25.6±19.1	6/2.04	13/1.11	0.93	This study