Response to referee comments

We would like to thank the referees and editor for the interest in our work and the helpful comments and suggestions to improve our manuscript. We have carefully considered all comments and the replies are listed below. The changes have been marked in the text using blue color.

Anonymous Referee #1

The behavior of Hg in the atmospheric is very important for the global Hg cycle. In this manuscript, Yin and colleagues determined TGM in Nam Co in the inland of Tibet Plateau and then used related models to address the transportation, transformation and source of TGM in the study region. This data reported in this paper is valuable because the study on the fate and transportation of TGM over the inland Tibet Plateau is almost blank. I suggest the manuscript to be accepted after minor revision.

Response: Thanks for your valuable advices and comments.

General comments:

1. Title: In the study, the authors measured the TGM concentration from January 2012 to October 2014 (< 3 years). Generally, the longer-term measurement should be over 5 years. I suggest the authors to modify the title for clarity.

Response: Thanks for your suggestion.

We modified the title to "Multi-year monitoring of atmospheric TGM at a remote high-altitude site (Nam Co, 4730 m a.s.l.) in the inland Tibetan Plateau" in lines 1-2.

2. Introduction. I suggest the authors to add some text to address the fate and transport of Hg in atmosphere, such as the redox chemistry of Hg, wet and dry deposition of atmospheric Hg and so on.

Response: Thanks for your suggestion. Changed as suggested in lines 54-56: "The global residence time of GEM is in the range of 0.5-2 years due to its high volatility, low solubility and chemical stability (Schroeder and Munthe, 1998; Shia et al., 1999). It is therefore transported globally over long distances

(tens of thousands of kilometers) far from pollution sources." and lines 63-65: "RGM and Hg-P are generally depicted as local and regional pollutants, and the dry and wet deposition of RGM and Hg-P are much faster than GEM (Schroeder and Munthe, 1998; Lin and Pehkonen, 1999; Lindberg and Stratton, 1998)."

3. Results and discussion. The present paper investigated the atmospheric TGM in a remote site. It should discuss more about the remote or rural sites, but not the urban sites.

Response: Thanks for your suggestion. Changed as suggested in lines 249-252: "The mean TGM concentration at the Nam Co Station is 1.33±0.24 ng m^{-3}, which is the lowest among all reported TGM concentrations at remote and rural sites in China (Liu et al., 2016; Fu et al., 2012b; Fu et al., 2012a; Fu et al. 2015; Ci et al., 2011; Dou et al., 2013; Zhang et al., 2015; Fu et al., 2010; Li et al., 2011; Zhang et al., 2013; Yu et al., 2015; Fu et al., 2008; Chen et al., 2013)."

4. Conclusion. I suggest the authors to shorten the conclusion.

Response: Thanks for your suggestion. We removed "The background TGM variation at the Nam Co Station was jointly regulated by surface-air flux and dilution in the planetary boundary layer in the diurnal cycle." as suggested.

Specific comments:

L67-72: The present paper focuses on the TGM in remote region, but the authors discussed a lot about atmospheric Hg in urban region. As mentioned above, I suggest he authors to address the fate and transport of Hg in the atmosphere. If the authors like to discuss the atmospheric Hg concentrations in different regions, it is more reasonable to discuss TGM in background or remote regions.

Response: Thanks for your suggestion. Changed as suggested in lines 63-65: "RGM and Hg-P are generally depicted as local and regional pollutants, and the dry and wet deposition of RGM and Hg-P are much faster than GEM (Schroeder and Munthe, 1998; Lin and Pehkonen, 1999; Lindberg and Stratton, 1998)." and lines 73-77: "Measurements of atmospheric mercury at background and remote sites in China

include the following sites: Wuzhishan (2011-2012), Mt. Changbai (2008-2010), Mt. Waliguan (2007-2008), Mt. Ailao (2011-2012), Shangeri-La (2009-2010) and Mt. Gongga (2005-2006) with concentrations ranging from 1.58 to 3.98 ng m^{-3} (Liu et al., 2016; Fu et al., 2012b; Fu et al., 2012a; Fu et al. 2015; Zhang et al., 2015; Fu et al., 2008)."

L107-109: This information is too general. I suggest to delete this paragraph in the revised manuscript.

Response: Thanks for your suggestion. We removed this paragraph.

L219-221: As mentioned above, it is not reasonable to compare to the urban and industrial regions. I suggest to compare the data to some rural or background sites in China.

Response: Thanks for your suggestion. Changed as suggested in lines 249-252: "The mean TGM concentration at the Nam Co Station is 1.33±0.24 ng m^{-3}, which is the lowest among all reported TGM concentrations at remote and rural sites in China (Liu et al., 2016; Fu et al., 2012b; Fu et al., 2012a; Fu et al. 2015; Ci et al., 2011; Dou et al., 2013; Zhang et al., 2015; Fu et al., 2010; Li et al., 2011; Zhang et al., 2013; Yu et al., 2015; Fu et al., 2008; Chen et al., 2013)."

Figure 1 and Table S1: I suggest the authors to remove all urban sites and use the global map to show the distribution of TGM or GEM concentrations in the remote and rural sites around the world. In the Table S1, most sites labelled as remote sites should be changed to the rural site. Don't forget the study in the ocean.

Response: Thanks for your suggestion. Table S1 was changed as suggested in supplementary material.

Figure 4: I suggest the authors to delete this figure. If they like to keep, try to merge all figures into one figure.

Response: Figure 4 was merged into a single figure as suggested in line 883.

Figure 8. Move to supplementary material.

Response: There was also some hesitation about the relevance of the box model from referee #3. However, referee #2 thought that there were important results that should be included in the conclusions. We have expanded the explanation for the use of the box model in Sec 2.4 to clarify the importance of the result. In brief, a model based on expected processes was not able to characterize the diurnal profile, but an alternative model with simple inputs was found that reproduced the measurements accurately. We believe that this is an important part of the model that will help in improving future models of reactions and processes affecting GEM and RGM.

Figure 10. Remove the line for no data. When there is no data, it should show blank.

Response: Thank you for pointing this out. Figure was modified as you suggested.

Anonymous Referee #2

General comments

Yin et al. present total gaseous mercury measurements from an extraordinary environment in "Long-term monitoring of atmospheric TGM at a remote high altitude site (Nam Co, 4730 m a.s.l.) in the inland Tibetan Plateau". The results are important as mercury data from a remote site are interpreted that is located between two major mercury emission regions the Indo-Gangetic plain in southern Asia and China in the west. With a box model and a relatively small set of parameters, the major seasonal and diurnal changes could be reproduced. The results fill an important gab in understanding atmospheric back ground concentrations of gaseous mercury in an especially important part of Asia. A slight drawback might be that only TGM as chemical parameter is measured during the study, despite others are mentioned (ozone, black carbon, RGM *: : :*). However, the data set is concisely analyzed and the findings are supported by box model simulations. Trajectories and potential source contribution functions were calculated and used for source allocation. I suggest publication in ACP after a few comments have been addressed.

Response: Thanks for your valuable advice and comments.

I agree with Referee #1 that the expression "Long-term" for the study is not appropriate. Moreover, the number of Figures should be restricted to about 10: Figure 2 could be moved to the supplementary material. Figures 11-14 have a large overlap regarding the displayed information. Either one should be selected or a combination of two of them might be displayed in the main text, the others can be moved to the supplementary material.

Response: We modified the title to "Multi-year monitoring of atmospheric TGM at a remote high-altitude site (Nam Co, 4730 m a.s.l.) in the inland Tibetan Plateau" in lines 1-2.

Thank you for the suggestion. We have moved Figs. 2, 12 and 13 to the supplementary material. Fig. 11 (new number 10) and Fig. 14 (new number 11) were kept in the paper as they provide two different viewpoints on the air mass transport. The first shows transport by cluster and the second by season.

Fig.2 was moved to the supplementary material (Fig. S4) and sentence was changed in line 260: "The frequency distribution of TGM at the Nam Co Station was normally distributed (Fig. S4)."

Fig. 12 was moved to supplementary material (Fig. S5) and sentences were changed in lines 378-

380: “Most HYSPLIT trajectories originated from the west of Nam Co including the western and central Tibetan Plateau, the southwestern part of the Xinjiang Uygur Autonomous Region, South Asia, Central Asia and Western Asia. Very few trajectories originated from eastern China (Fig. S5).”

Fig. 13 was moved to supplementary material (Fig. S6) and sentences were changed in lines 388-390: “Areas including IGP, the southern part of the Xinjiang Uygur Autonomous Region, the western part of Qinghai province and areas near the Nam Co Station in the Tibet Autonomous Region were identified as overall high potential sources regions and pathways (Fig. S6).”

Specific comments

Lines 79-81. The importance of background measurements between two major mercury source areas should be explained in more detail. Is the change in the background or the actual deviations from the background the important information, i.e., the episodic events?

Response: The decrease of TGM at the Nam Co Station was similar to many sites. This was stated in this study in Sec. 3.1 as: “The monthly average TGM at the Nam Co Station showed a weak decrease (slope = -0.006) during the entire monitoring period, and the decrease was more pronounced in the summer (slope = -0.013). Despite the short time span of the TGM time series with some missing data mostly in the winter, the slight decrease of TGM especially in the summer was in agreement with a recent study using plant biomonitoring which identified a decreasing atmospheric mercury since 2010 near Dangxiong county (Tong et al., 2016) as well as decreases of TGM at other sites (Slemr et al., 2011; Zhang et al., 2016)”. WRF-FLEXPART results revealed that the high concentrations of atmospheric mercury (above 2 ng m^{-3}) at the Nam Co station were associated with the air mass transported from the east and though Lhasa, which was also probably due to the further impact from eastern Indo-Gangetic Plain and the possibility of episodic transport events from China as stated in lines 371-373.

Lines 85-86. Similarly, as the previous comment. Be more explicit: Tibetan plateau is an important part of the global mercury cycle *: : :* . Is it an ideal place to monitor TGM or an important sink?

Response: Thanks for your suggestion. Changed as suggested in lines 89-91: “Notably, mercury

157 records from glaciers and lake sediments suggest that the Tibetan Plateau is an important part of the
158 global mercury cycle, acting as both a sink (mercury deposition to snow) and a source (release of mercury
159 from melting ice) (e.g., Kang et al., 2016; Yang et al., 2010; Sun et al., 2017, Sun et al., 2018)."

160

161 Line 180-185. This appears to be an important result and should be mentioned in the conclusions.

162 **Response:** Thanks for your suggestion. Changed as suggested in lines 440-441: "An exploratory
163 box model simulation shows that this diurnal profile can be accurately represented using TGM reductions
164 24 hours per day, TGM increases near sunrise and sunset, and dilution due to vertical mixing."

165

166 Line 184. "... constant TGM deposition" – how is this justified considering diurnal soil temperature
167 change and the major fraction of TGM being volatile GEM?

168 **Response:** We have changed "deposition" to "reduction," and we have also changed the explanation
169 of the box model as described above. The point of the box model was to illustrate that known processes
170 were not able to match the measured diurnal profile of TGM concentrations. We did indeed expect that
171 temperature would account for some of the variation, but it did not in the model. This suggests that future
172 modeling efforts will be required to explore the impact of soil emissions as well as snow and ice melting
173 on concentrations at Nam Co.

174

175 Line 281-282. As before: "... constant TGM deposition" – please support this by a mechanism, and
176 further on, "*: : :* TGM emissions in the early evening *: : :* ". - This statement seems to be in contradiction
177 to what is said before. To clarify the contributions a modified Figure 8 as stacked bar plot would help.

178 **Response:** The description was changed from "deposition" to "reduction" and from "emission" to
179 "increase" to highlight the fact that the box model is a hypothetical experiment to see which processes
180 would be required to accurately represent the measured diurnal profile. We would like to note that the
181 combined 5 inputs are hypothetical factors that were found to match the diurnal profile of TGM. As such,
182 they are not based on known mechanisms but rather are there to emphasize the need for further research
183 to identify the factors influencing the diurnal profile. Figure 8 cannot be made as a stacked bar plot – this

would necessitate a more detailed process analysis study which is outside the bounds of the current paper. The description of box model was modified in lines 201-208: "TGM at the Nam Co Station is expected to be well mixed and the site is not influenced by local sources. It is therefore expected that a box model should be able to reproduce the diurnal profile of concentrations. A box model that accurately simulates the diurnal profile of TGM would provide constraints on known processes affecting the concentrations. Comparisons with measured profiles would further identify missing processes in the model. This approach was used for reactive mercury at the same site, where it identified the role of the reduction of reactive mercury to gaseous elementary mercury mediated by sunlight (de Foy et al., 2016b). A box model was made that included free parameters to represent known chemical reactions and dispersion processes. An optimization algorithm was used to identify the parameters required to fit the model to the data, as was done in de Foy et al. (2016b)"; and 210-215 "A simplified exploratory model was therefore sought that would represent the measured diurnal variations as simply as possible, according to Occam's razor. Although this model does not yield direct information on known processes, it does identify the kinds of processes and their magnitude that would be required to accurately represent the measured diurnal profile. The final model combined the following 5 inputs: TGM increases at sunrise and in the early evening, constant TGM reductions 24 hours a day, a constant lifetime for TGM loss during daylight hours and TGM dilution due to vertical mixing".

Lines 294-300. Mixing versus oxidation: as TGM (GEM+ RGM) is measured this explanation is vague.

Response: Thanks for your suggestion. Changed as suggested in lines 325-329: "As the temperature and radiation increased, so did the boundary layer height which developed into a convective mixed boundary layer and generated greater vertical mixing between the surface and loft. At the same time, the surface wind speed also increased. With increased vertical and horizontal dispersion, TGM released from the surface was diluted during the daytime (Liu et al., 2011; Lee et al., 1998)."

Lines 346-347. This part also reflects an important finding which could be emphasized in the conclusions.

Response: Thanks for your suggestion. Changed as suggested in lines 437-438: "Peak concentrations of TGM at the Nam Co Station were associated with air masses from the eastern Indo-Gangetic Plain with the possibility of episodic transport events from China."

Line 370. What is the new finding compared to the Beiluhe site study {Ci, 2016 #16007},

Response: The box model in this study was found that reproduced the measurements of atmospheric mercury at site in the Tibetan Plateau accurately, and it provided constraints on known processes affecting the concentrations. Furthermore, Box model results in this study illustrated that surface-air fluxes of mercury were connected to atmospheric mercury with dilution effect.

Line 426. The average of 1.33 ng m-3 is almost in agreement with Ci et al. {, 2016 #16007}

Response: The concentrations of atmospheric mercury at both of these sites in the Tibetan Plateau were very low illustrating the pristine environments of atmosphere at both sites. Study of diurnal variation of atmospheric mercury in this study probably can supplement existing studies at Beiluhe.

Lines 430-431. What is stated? Currently it sounds self-evident.

Response: Compared with other high-altitude background sites, the low concentration of TGM at the Nam Co Station in the winter may be due to the reduction of mercury due to halogen, as discussed in Sec. 3.2. We explained these in lines 286-292: "Compared to other high-altitude background sites in the mid-latitudes in Europe (Fig. 4) (Denzler et al., 2017; Fu et al., 2016a; Ebinghaus et al., 2002) and sites in mid-latitudes in the US (Holmes et al., 2010; Weiss-Penzias et al., 2003; Sigler et al., 2009; Yatavelli et al., 2006), the lower concentration of TGM at the Nam Co Station in the winter might be indicative of atmospheric mercury removal in the winter caused by reactive halogens (Br and Br_2). The reaction rates for these reactions are a strong inverse function of temperature (de Foy et al., 2016b; Goodsite et al., 2004), and they are accompanied by lower surface ozone concentration (Yin et al., 2017), which is catalytically destroyed by halogens (Bottenheim et al., 1986; Obrist et al., 2011)."

Lines 436-437. It appears to be in contradiction to a previous interpretation (comment on statement in Lines 281-282).

Response: Thanks for pointing this out. Note that the contradiction is only apparent; we have adjusted the sentence in lines 439-441 to clarify the continuity in our results: "At the Nam Co Station, the diurnal TGM profile had a peak 2-3 hours after sunrise and reached its lowest concentration before sunset. An exploratory box model simulation shows that this diurnal profile can be accurately represented using TGM reductions 24 hours per day, TGM increases near sunrise and sunset, and dilution due to vertical mixing."

In lines 439-440, we stated that "At the Nam Co Station, the diurnal TGM profile had a peak 2-3 hours after sunrise and reached its lowest concentration before sunset.". In lines 312-315, we stated that "The best match in the box model was obtained by using variables including constant TGM reduction throughout the day, TGM increases at sunrise, TGM increases in the early evening, TGM dilution due to vertical mixing and a lifetime of TGM loss during daylight hours (Table 2)." The diurnal peak of TGM at the Nam Co Station was happened 2-3 hours after sunrise, and it was because the accumulation of TGM emissions. While the lowest concentration before sunset was due to the strong dilution in the afternoon and they were supported by the results of box model.

Line 442. The transformation GEM -> RGM was analyzed in detail by de Foy et al. {, 2016 #16006}, but it does not reflect an important fraction.

Response: Thank you for pointing this out. According to the previous study, RGM concentrations were much lower than GEM at the Nam Co Station (de Foy et al., 2016). But we want to make sure the potential impact was stated in this study.

Line 443. Due to insolubility of TGM *: : :* – less soluble GEM? At least the RGM part should be better soluble. Please, be more specific.

Response: Thanks for your suggestion. The RGM was < 2% of TGM at the Nam Co Station, so the

TGM measured at the Nam Co Station was mainly GEM. Comparing to black carbon and hexachlorocyclohexanes discussed in this study, TGM was less soluble. Sentences were added in lines 145-147: "At the Nam Co Station, the TGM fraction consists mostly of GEM (more than 98%). The operationally defined RGM accounted for less than 2% of TGM (Figure S1 in supplementary material in de Foy et al., 2016b). We consider the Tekran data to represent TGM in line with previous studies (e.g. Kock et al., 2005; Slemr et al., 2008; Müller et al., 2012)".

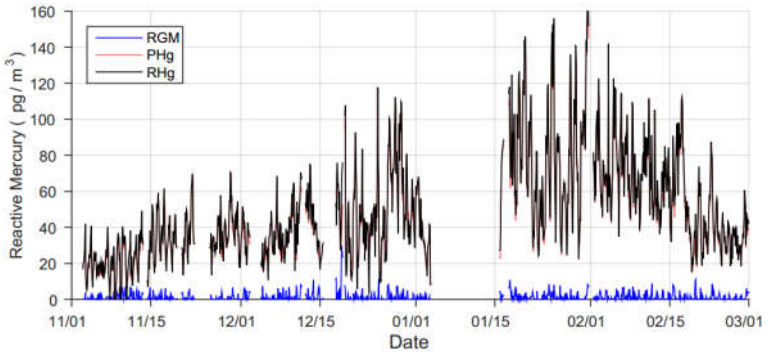

Figure S1: Time series of Reactive Mercury (RHg) and its components Reactive Gaseous Mercury (RGM) and Particle-bound Mercury (PHg) in supplementary material in de Foy et al., 2016b

Figure 9. Pink line, what does q (k kg-1), Please give more information in the figure caption.

Response: Thanks for pointing this out. q is the symbol used for specific humidity and we modified Fig.8 as you suggested (new number 8).

References referred to in the comments:

Ci, Z., Peng, F., Xue, X. and Zhang, X., 2016. Air–surface exchange of gaseous mercury over permafrost soil: an investigation at a high-altitude (4700 m a.s.l.) and remote site in the central Qinghai–Tibet Plateau. Atmos. Chem. Phys., 16(22): 14741-14754.

de Foy, B. et al., 2016. First field-based atmospheric observation of the reduction of reactive mercury driven by sunlight. Atmospheric Environment, 134: 27-39

Slemr, F., Brunke, E.-G., Labuschagne, C., and Ebinghaus, R.: Total gaseous mercury

concentrations at the Cape Point GAW station and their seasonality, Geophys. Res. Lett., 35, L11807, doi:10.1029/2008GL033741, 2008

Kock, H. H., Bieber, E., Ebinghaus, R., Spain, T. G., and Thees, B.: Comparison of long-term trends and seasonal variations of atmospheric mercury concentrations at the two European coastal monitoring stations Mace Head, Ireland, and Zingst, Germany, Atmos. Environ., 39, 7549–7556, doi:10.1016/j.atmosenv.2005.02.059, 2005

Müller, D., Wip, D., Warneke, T., Holmes, C. D., Dastoor, A., & Notholt, J. (2012). Sources of atmospheric mercury in the tropics: continuous observations at a coastal site in Suriname. Atmospheric Chemistry and Physics, 12(16), 7391-7397.

Anonymous Referee #3

General comments

This paper presents a multi-year record of gaseous mercury concentrations at Nam Co station on the Tibetan Plateau. It will make a valuable addition to the literature given the scarcity of multi-year measurements in that region of the world. Remote stations are very useful to constrain global atmospheric models and for long-term trend analysis. I recognize the author's efforts to interpret the data set and I think the paper will be suitable for publication in ACP after the authors address the following issues:

Response: Thanks for your valuable advices and comments.

Main comment #1: To me, "GEM" and "TGM" are not really interchangeable. The authors sometimes refer to GEM concentrations, sometimes to TGM (Fig.5 for instance) but there is no discussion on why and this is quite confusing. Do you assume that there is a difference depending on location? I suggest you refer to the first paragraph of page 11919 in Sprovieri et al. (2016). I think you can assume that you monitor GEM concentrations at Nam Co station. Additionally, rather than using "TGM" or "GEM", something useful would be to add the type of instrumentation used at each site in Table S1: "Tekran speciation unit" or "Tekran 2535 + PTFE filter at the entrance inlet" or *: : :*?

Response: Thanks for your suggestion. Sentences were added in lines 145-147: "At the Nam Co Station, the TGM fraction consists mostly of GEM (more than 98%). The operationally defined RGM accounted for less than 2% of TGM (Figure S1 in supplementary material in de Foy et al., 2016b). We consider the Tekran data to represent TGM in line with previous studies (e.g. Kock et al., 2005; Slemr et al., 2008; Müller et al., 2012)".

Table S1 was changed as suggested in supplementary material.

Main comment #2: The authors used various models to interpret the data set: HYSPLIT, FLEXPART, MLR model and a box model. It is however not always easy to understand why you needed that many models and how the models are complementary. For instance, why do you need both HYSPLIT and FLEXPART to perform the cluster analysis. It is not straightforward to me, and I would appreciate a

sentence or two in the Materials and Methods section to clarify that.

Response: Thanks for your suggestion. The box model was used to investigate the diurnal variation of atmospheric mercury at the Nam Co Station; the MLR model was used to investigate the contribution from parameters to the overall variation of atmospheric mercury at the Nam Co Station, and FLEXPART result was one of the parameters in MLR model indicating local winds; HYSPLIT was used to investigate the trajectories arriving at Nam Co and were also used to calculate the Potential Source Contribution Function.

Sentences were changed as suggested in lines 166-168: "HYSPLIT backward trajectories were used to calculate the Potential Source Contribution Function (section 2.6) which serves to investigate the potential sources contributing to atmospheric mercury at the Nam Co Station" and lines 171-174: "The use of two different trajectory models (HYSPLIT and WRF-FLEXPART) with different input meteorology can add robustness to the discussion as was done for the ozone study at Nam Co (Yin et al., 2017). Furthermore, the WRF-FLEXPART simulations were some of the parameters used in the multiple linear regression model (section 2.4)".

Main comment #3: The way it is presented and discussed, I don't really understand the usefulness of the box model to describe the diurnal cycle. As the initial model failed to reproduce the diurnal cycle, you added, among other things, TGM emissions at sunrise and in the early evening. To me, you are just tuning the model to reproduce observations, and these two "bursts" are not in line with the diurnal cycle of Hg(0) airsurface exchanges described by Ci et al. (2016). I therefore don't see why you can conclude that the box model provides "supporting evidence and estimates of diurnal TGM deposition and TGM bursts of (re)emissions". A reorganization of the manuscript (see main comment #6) might be useful to explain what you did and why more clearly.

Response: Thank you for your comment. The plot of the diurnal variation of TGM concentrations by seasons shows that there is a significant difference in the profile of TGM concentrations with other sites. Nam Co is a remote site on flat terrain, and is therefore expected to experience concentrations that are uniform over large distances (of the order of 50 km at least). We therefore sought to build a box model that would represent the diurnal variations of the concentrations based on what is known about TGM.

366 The box model thus constructed was not able to reproduce the measured profile. We therefore constructed
367 an alternative model that would be able to reproduce the profile. Although this is hypothetical, it points
368 the way to the type of processes that may need to be included in order to better simulate the measurements.
369 This is an integral part of the development of models and chemical mechanisms which closes the loop
370 between measurements and simulations. We have expanded the rationale for the box model in section
371 2.4 as follows (lines 201-215):

372 "TGM at the Nam Co Station is expected to be well mixed and the site is not influenced by local
373 sources. It is therefore expected that a box model should be able to reproduce the diurnal profile of
374 concentrations. A box model that accurately simulates the diurnal profile of TGM would provide
375 constraints on known processes affecting the concentrations. Comparisons with measured profiles would
376 further identify missing processes in the model. This approach was used for reactive mercury at the same
377 site, where it identified the role of the reduction of reactive mercury to gaseous elementary mercury
378 mediated by sunlight (de Foy et al., 2016b). A box model was made that included free parameters to
379 represent known chemical reactions and dispersion processes. An optimization algorithm was used to
380 identify the parameters required to fit the model to the data, as was done in de Foy et al. (2016b).
381 Preliminary tests of the box model were made using solar radiation and temperature to represent chemical
382 transformations, as well as using wind speed and boundary layer height to represent dilution. However
383 these attempts failed to reproduce the diurnal variation found in the measurements. A simplified
384 exploratory model was therefore sought that would represent the measured diurnal variations as simply
385 as possible, according to Occam's razor. Although this model does not yield direct information on known
386 processes, it does identify the kinds of processes and their magnitude that would be required to accurately
387 represent the measured diurnal profile. The final model combined the following 5 inputs: TGM increases
388 at sunrise and in the early evening, constant TGM reductions 24 hours a day, a constant lifetime for TGM
389 loss during daylight hours and TGM dilution due to vertical mixing."

390 We have also expanded the discussion of the results in section 3.3 as follows (lines 312-331):

391 "Fig. 7 showed the comparison of TGM concentrations with a box model simulation by seasons.
392 The best match in the box model was obtained by using variables including constant TGM reduction
393 throughout the day, TGM increases at sunrise, TGM increases in the early evening, TGM dilution due to
394 vertical mixing and a lifetime of TGM loss during daylight hours (Table 2). The R^2 of the model

simulation ranged from 0.91 to 0.99, suggesting that the simulations reproduced the diurnal variations accurately. As described above, both the measurements and the model have sharp bursts of TGM in the morning (7:00-9:00) and in the evening (18:00-22:00) during all seasons. Constant reductions existed in the spring, summer and autumn which would correspond to reduction rates of around 1 to 2 ng m^{-2} h^{-1}.

Fig. 8 showed the seasonal diurnal profiles of TGM and meteorological parameters. TGM concentrations were stable or slightly decreasing after midnight (0:00-6:00) under shallow nocturnal boundary layers. Notably, the morning increase of TGM happens immediately after sunrise, but before the increases of temperature, wind speed or humidity. The atmospheric mercury bursts in the morning (7:00-9:00) is probably due to prompt re-emission of nocturnal mercury deposition on the Earth's surface (Fu et al., 2016b; Howard et al., 2017; Kim 2010). The stable nocturnal boundary layer terminated at sunrise at which point mercury, including the mercury in the soil indigenously and/or deposited overnight, started to be reemitted into the shallow stable boundary layer before the increase of temperature which leads to an increase in the mixing height. As the temperature and radiation increased, so did the boundary layer height which developed into a convective mixed boundary layer and generated greater vertical mixing between the surface and loft. At the same time, the surface wind speed also increased. With increased vertical and horizontal dispersion, TGM released from the surface was diluted during the daytime (Liu et al., 2011; Lee et al., 1998). When the temperature decreased and the boundary layer converted back into a nocturnal boundary layer after sunset, depressed vertical mixing facilitated the build-up of TGM and such build-up was more significant in the warm seasons. In the evening, increases in TGM correspond to increases in specific humidity, especially in the summer."

Main comment #4: I agree with the other reviewers, I think that there are too many figures. Figures 2, 4 and 5 can be moved to SI. Figures 7-9 can be combined, Figures 11-14 as well.

Response: Thanks for your suggestion. Figure 2 was moved to SI. Figure 4 was combined into a single panel (new number 3). Figure 12 and 13 were moved to SI.

Main comment #5: I think that your time series is too short to do a trend analysis, especially given the number of missing values in 2013 and 2014.

Response: Thanks for your suggestion which was also raised by reviewer #1. We changed the title of the paper to say "multi-year" instead of trend and have modified the title to "Multi-year monitoring of atmospheric TGM at a remote high-altitude site (Nam Co, 4730 m a.s.l.) in the inland Tibetan Plateau" in lines 1-2.

We did our best to make sure instruments in a good condition under the harsh environment in the Tibetan Plateau. For the valid TGM data at the Nam Co Station, there were 6276 hourly data in 2012 (71.44%), 3561 hourly data in 2013 (40.65%) and 6185 data in 2014 (70.6%). Most of the missing data were in the cold seasons. The data in the summer were more complete than the other seasons and we found that there was a weak decrease of TGM in the summer at the Nam Co Station.

Main comment #6: The discussion is a bit messy and difficult to follow (see comments #2 and #3). I suggest a reorganization of the manuscript. Here is an idea:

1. Introduction Move section 3.5 ("Anthropogenic and natural sources of TGM") here as there is no discussion of the results in it and it provides useful information regarding emissions sources in the region (especially natural sources).

Response: Thanks for your suggestion. Section 3.5 was moved to introduction and introduction was rewritten in lines 49-116, please refer to the context in the revised manuscript.

2. Measurements and Methods (unchanged)

3. Results and Discussion

3.1. GEM concentrations

Here you first add your current section 3.1 (TGM concentrations). Then, you can present results from the MLR model in order to emphasize which parameters explain the observed GEM variations. Then discussion on seasonal variations. I suggest you move your current section 3.7 here. Finally, you discuss the diurnal cycle.

Response: For section 3, we first present the datasets, then the analysis of the results of the models

and finally the implications based on our studies of TGM and other relevant pollutants in the region. In section 3.7 we suggested that the Indian summer monsoon has an important impact on the seasonal variation of TGM. In addition, we highlighted that mercury, which is a passive tracer representative of gaseous pollutants with low reactivity, differs in seasonal variation from particulate pollutants. Therefore, we suggested that additional measurements of multiple pollutants and comparative studies are required to achieve a more comprehensive understanding and assessment of transboundary air pollution to the Tibetan Plateau. We would prefer to keep section 3.7 at the end so that our study can be more informative to a wider range of readers.

3.2. Cluster analysis

Here you combine results from FLEXPART and HYSPLIT to discuss long-range transport to Nam Co station.

Response: Thanks for your suggestion. WRF-FLEXPART is used as an input to the multiple linear regression. HYSPLIT is used to calculate the Potential Source Contribution Function. Due to the different function of these two sections, we prefer to discuss them separately.

4. Conclusion

Main comment #7: The authors make good use of the literature and compare results at Nam Co stations with other stations around the world, especially in China. Given the large inter-annual variability and significant decreasing trends observed in China (e.g., Tang et al., 2018), I suggest you add the date (year) at which monitoring was performed when you refer to another study.

Response: Thanks for your suggestion. Changed as suggested in lines 71-73: "Atmospheric mercury concentrations in Guizhou, one of the most important mercury producing and coal producing regions in China, was reported to be 6.2 - 9.7 ng m^{-3} of TGM in the capital city of Guiyang between 2001- 2009 (Feng et al., 2004; Liu et al., 2011; Fu et al., 2011)." and lines 73-77: "Measurements of atmospheric mercury at background and remote sites in China include the following sites Wuzhishan (2011-2012), Mt. Changbai (2008-2010), Mt. Waliguan (2007-2008), Mt. Ailao (2011-2012), Shangeri-

La (2009-2010) and Mt. Gongga (2005-2006) with concentrations ranging from 1.58 to 3.98 ng m^{-3} (Liu et al., 2016; Fu et al., 2012b; Fu et al., 2012a; Fu et al. 2015; Zhang et al., 2015; Fu et al., 2008)."

The following line by line comments should be useful to fully comprehend and address the various "main comments". Line by line comments

Line 1: "Long-term monitoring of atmospheric TGM". I agree with the other reviewers, "multi-year monitoring" would perhaps be more appropriate here.

Response: Thanks for your suggestion. We modified the title to "Multi-year monitoring of atmospheric TGM at a remote high-altitude site (Nam Co, 4730 m a.s.l.) in the inland Tibetan Plateau" in lines 1-2.

Line 25: "Total gaseous mercury concentrations". See main comment #1.

Response: Sentences were added in lines 145-147: "At the Nam Co Station, the TGM fraction consists mostly of GEM (more than 98%). The operationally defined RGM accounted for less than 2% of TGM (Figure S1 in supplementary material in de Foy et al., 2016b). We consider the Tekran data to represent TGM in line with previous studies (e.g. Kock et al., 2005; Slemr et al., 2008; Müller et al., 2012)".

Line 30: "TGM at the Nam Co Station exhibited a slight decreasing trend especially for summer seasons". See main comment #5.

Response: Thank you for your suggestion. We did our best to make sure that the instruments were in a good condition under the harsh environment in the Tibetan Plateau. For the valid TGM data at the Nam Co Station, there were 6276 hourly data in 2012 (71.44%), 3561 hourly data in 2013 (40.65%) and 6185 data in 2014 (70.6%). Most of the missing data were in the cold seasons. Data in the summer were more complete than during the other seasons and we found that there was weak decrease of TGM in the summer at the Nam Co Station.

Lines 30-31: “The seasonal variation of TGM was characterized by high levels during warm seasons and low levels during cold seasons”. Please, define “high” and “low”. Perhaps give mean ïC´ s standard deviation for both seasons. Is the difference between mean concentrations significantly different?

Response: Thanks for your suggestion. We added the mean ± standard deviation for seasons as you suggested in lines 30-33: “The seasonal variation of TGM was characterized by higher concentrations during warm seasons and lower concentrations during cold seasons, decreasing in the following order: summer (1.50±0.20 ng m^{-3}) > spring (1.28±0.20 ng m^{-3}) > autumn (1.22±0.17 ng m^{-3}) > winter (1.14±0.18 ng m^{-3}).”

Lines 54-55: “The global residence time of GEM is in the range of 0.5-2 years due to its high volatility, low solubility and chemical stability (Schroeder and Munthe, 1998; Shia et al., 1999)”. I suggest you add Horowitz et al. (2017). Using a new mechanism for atmospheric Hg redox chemistry in GEOS-Chem, the authors found that the chemical lifetime of tropospheric GEM against oxidation is 2.7 months, shorter than previous estimates.

Response: Thanks for your suggestion. Sentence was added as you suggested in lines 56-58: “Horowitz et al. (2017) recently reported that the chemical lifetime of tropospheric GEM against oxidation may be much shorter than previously reported: it could be as short as 2.7 months.”

Lines 67-69: “For example, atmospheric mercury concentrations in Guizhou, one of the most important mercury producing and coal producing regions in China, was reported to be 6.2-9.7 ng/m3 of TGM in the capital city of Guiyang”. When was the monitoring performed (which year)? See main comment #7. Lines 71-72: “With levels ranging from 4.8 to 18.4 ng/m3”. Same as above, see main comment #7.

Response: Thanks for your suggestion.

Table S1 was changed as you suggested. The sentence about urban sites was removed as suggested by another reviewer.

Sentences were modified in lines 71-77: “Atmospheric mercury concentrations in Guizhou, one of the most important mercury producing and coal producing regions in China, was reported to be 6.2 - 9.7 ng m^{-3} of TGM in the capital city of Guiyang between 2001- 2009 (Feng et al., 2004; Liu et al., 2011; Fu et al., 2011). Measurements of atmospheric mercury at background and remote sites in China include the following sites Wuzhishan (2011-2012), Mt. Changbai (2008-2010), Mt. Waliguan (2007-2008), Mt. Ailao (2011-2012), Shangeri-La (2009-2010) and Mt. Gongga (2005-2006) with concentrations ranging from 1.58 to 3.98 ng m^{-3} (Liu et al., 2016; Fu et al., 2012b; Fu et al., 2012a; Fu et al. 2015; Zhang et al., 2015; Fu et al., 2008).”.

Line 75: “In recent years, China and India signed the Minamata Convention and have started to control Hg emissions more strictly”. Note that China signed to Convention in 2013 and ratified it in August 2016 while India signed it in 2014 but hasn’t ratified it yet. http://mercuryconvention.org/Countries/Parties/tabid/3428/language/enUS/Default.aspx

Response: Thanks for your suggestion. Changed as suggested in lines 99-100: “In recent years, China and India signed the Minamata Convention and will probably control mercury emissions more strictly (Selin, 2014).”

Lines 76-79: Is that in line with latest emissions scenarios by Pacyna et al. (2016)?

Response: Pacyna et al. (2016) stated that “A decrease in emissions in Europe and North America during the time period has been offset by an increase in Asia. The largest increase in emissions is generally due to an increase in coal burning for power and heat generation and for industrial purposes. Increased use of air pollution controls, removing mercury as a co-benefit (and some mercury-specific removing technologies), has slowed down or even reduced the emissions from the increased energy demand. This is especially the case for Europe and North America, but it is also reflected in new coal-fired power plants with state-of-art pollution controls implemented in China (AMAP/UNEP, 2013a).”; “According to the “New Policy” scenario (NP 2035) a moderate decrease in mercury deposition (20–30 %) is predicted over the whole of the globe except for South Asia (India), where an increase in deposition (10–15 %) is expected due to the growth of regional anthropogenic emissions (Fig. 8c and d)”.

But in Pacyna et al. (2016), the details of mercury emissions in China and India were not provided.

Lines 85-86: "(*: : :*) suggested that the Tibetan Plateau is an important part of global Hg cycle". What do you mean?

Response: Sentences was changed in lines 89-91: "Notably, mercury records from glaciers and lake sediments suggest that the Tibetan Plateau is an important part of the global mercury cycle, acting as both a sink (mercury deposition to snow) and a source (release of mercury from melting ice) (e.g., Kang et al., 2016; Yang et al., 2010; Sun et al., 2017, Sun et al., 2018)."

Lines 90-91: "(*: : :*) was found at high concentrations in Lhasa". Please, define "high".

Response: Thanks for your suggestion. Changed as suggested in lines 94-97: "Studies of mercury in precipitation and water vapor evidenced that the Tibetan Plateau is likely sensitive to pollutant input including mercury (Huang et al., 2012; Huang et al., 2013), and the particulate-bound mercury in total suspended particulates was found at high concentrations in Lhasa with an average of 224 pg m^{-3} which was comparable to other cities in China (Huang et al., 2016)."

Lines 99-101: "HYSPLIT, WRF-FLEXPART and PSCF were used to identify potential sources and impacts from long-range transport". What kind on information do they each provide? Are the methods complementary? See main comment #2.

Response: The box model was used to investigate the diurnal variation of atmospheric mercury at the Nam Co Station; The MLR model was used to investigate the contribution from parameters to the overall variation of atmospheric mercury at the Nam Co Station, and FLEXPART result was one of the parameters in MLR model indicating local winds; HYSPLIT was used to investigate the trajectories arrived at Nam Co and also be used to calculated in Potential Source Contribution Function.

For clarity, the following sentences were changed as suggested in lines 166-168: "HYSPLIT backward trajectories were used to calculate the Potential Source Contribution Function (section 2.6)

583 which serves to investigate the potential sources contributing to atmospheric mercury at the Nam Co
584 Station"; and lines 171-174:"The use of two different trajectory models (HYSPLIT and WRF-
585 FLEXPART) with different input meteorology can add robustness to the discussion as was done for the
586 ozone study at Nam Co (Yin et al., 2017). Furthermore, the WRF-FLEXPART simulations were some of
587 the parameters used in the multiple linear regression model (section 2.4)".

588

589 Section 2.1. Measurement site: Is there snow at the station? If so, at which period? I am just
590 wondering whether you could have Hg re-emissions from the snowpack.

591 **Response:** There was snow at the Nam Co Station discontinuously from October to March. But due
592 to the strong wind at this period and flat terrain at station, the snow did not remain on the ground for
593 more than a few days at a time. We did not measure the Hg re-emissions from the snowpack at the Nam
594 Co Station, but probably we will seek to do the field work for that in the future.

595 Sentence was added in lines 126-128: "There was snow at the Nam Co Station discontinuously from
596 October to March. But due to the strong wind at this period and the flat terrain surrounding the station,
597 the snow did not remain on the ground for more than a few days at a time."

598

599 Section 2.2. Measurements: TGM, surface ozone and meteorology. Shouldn't you say that you
600 measure GEM instead of TGM, according to Sprovieri et al. (2016)? See main comment #1.

601 **Response:** Thanks for your suggestion. Sentences were added in lines 145-147: "At the Nam Co
602 Station, the TGM fraction consists mostly of GEM (more than 98%). The operationally defined RGM
603 accounted for less than 2% of TGM (Figure S1 in supplementary material in de Foy et al., 2016b). We
604 consider the Tekran data to represent TGM in line with previous studies (e.g. Kock et al., 2005; Slemr et
605 al., 2008; Müller et al., 2012)".

606

607 Lines 126-127: "A 45-mm diameter Teflon filter was placed in front of the inlet". How often did
608 you change the filter?

Response: It was changed every two weeks. Sentence was modified in lines 137-138: "A 45-mm diameter Teflon filter (pore size 0.2 μm) was placed in front of the inlet and it was changed every two weeks."

Line 148: "The backward trajectories arrival height in HYSPLIT was set at 500 m above the surface". I suggest you add here (and delete there) what's described in lines 373-375: "Results of air masses at different heights (500 m, 1000 m and 1500 m) showed similar patterns, hence, we selected trajectories released at a height of 500 m as representative since 500 m is suitable for considerations of both the long-range transport and transport in the planetary boundary layer".

Response: Thanks for your suggestion. Changed as suggested in lines 163-165: "Results of air masses at different heights (500m, 1000m and 1500m) showed similar patterns, hence, we selected trajectories released at a height of 500 m as representative since 500 m is suitable for considerations of both the long-range transport and transport in the planetary boundary layer.".

Line 151: "In addition to HYSPLIT, WRF-FLEXPART was used". Could you briefly explain why? See main comment #2.

Response: MLR model was used to investigate the contribution from parameters to the overall variation of atmospheric mercury at the Nam Co Station, and FLEXPART result was one of the parameters in MLR model indicating local winds. While HYSPLIT was used to investigate the trajectories arrived at Nam Co and also be used to calculated in Potential Source Contribution Function.

For better understanding, sentences were changed as suggested in lines 166-168: "HYSPLIT backward trajectories were used to calculate the Potential Source Contribution Function (section 2.6) which serves to investigate the potential sources contributing to atmospheric mercury at the Nam Co Station" and lines 171-174:"The use of two different trajectory models (HYSPLIT and WRF-FLEXPART) with different input meteorology can add robustness to the discussion as was done for the ozone study at Nam Co (Yin et al., 2017). Furthermore, the WRF-FLEXPART simulations were some of the parameters used in the multiple linear regression model (section 2.4)".

Line 153: Out of curiosity, why is HYSPLIT ran for 5 days vs. 4 days for FLEXPART?

Response: Air mass transport times from areas surrounding the Tibetan Plateau are usually around 1 to 2 days. Using trajectories of 4 or 5 days guarantees that we account for events with longer residence times. The two sets of simulations were made independently which is why there are difference in the configurations in addition to differences in the input meteorological data and models. They are intended to show that despite the differences, the conclusions from the two models are in agreement.

Line 158: Please define MLR.

Response: Changed as suggested in lines 178-179: "A Multiple Linear Regression (MLR) model was used to quantify the main factors affecting the hourly concentrations of TGM."

Line 161: Could you briefly describe what kind of inter-annual, seasonal and diurnal factors you are referring to?

Response: We apologize for the short cut, the details are in Yin et al. (2017). Text added in lines 183-185: "Briefly, the inter-annual factors are separate scaling factors for each year of the measurements, the seasonal factors are 12-month and 6-month harmonic terms (sine and cosine), and the diurnal factors are scaling factors for each hour of the day."

Lines 183-184: "TGM emissions at sunrise and in the early evening". At this point of the manuscript, I don't really understand why you would have Hg emissions at sunrise and in the early evening. See main comment #3.

Response: Thank you for pointing this out. For better understanding, we have modified "Hg emissions at sunrise and in the early evening" to "increase of Hg at sunrise and in the early evening". Sentence was modified in lines 213-215: "The final model combined the following 5 inputs: TGM increases at sunrise and in the early evening, constant TGM reductions 24 hours a day, a constant lifetime

for TGM loss during daylight hours and TGM dilution due to vertical mixing."

Lines 233-234: I'm skeptical given the number of missing data in 2013 and 2014 vs. 2012. I don't think the time series is long enough to perform a trend analysis. See main comment #5.

Response: Thanks for your suggestion. We did our best to make sure instruments in a good condition under the harsh environment in the Tibetan Plateau. For the valid TGM data at the Nam Co Station, there were 6276 hourly data in 2012 (71.44%), 3561 hourly data in 2013 (40.65%) and 6185 data in 2014 (70.6%). Most of the missing data were in the cold seasons. Data in the summer were relatively more than other seasons and we found that there was weak decrease of TGM in the summer at the Nam Co Station.

Line 236-237: "(*: : :*) as well as a worldwide downward trend of TGM". There is no "worldwide downward trend". For instance, while a downward trend has been observed at Cape Point station in South Africa from 1996 to 2005, there is an upward one since 2007 (Martin et al., 2017; Slemr et al., 2015).

Response: Thanks for your suggestion. Changed as suggested in lines 265-268: "Despite the short time span of the TGM time series with some missing data mostly in the winter, the slight decrease of TGM especially in the summer was in agreement with a recent study using plant biomonitoring which identified a decreasing atmospheric mercury since 2010 near Dangxiong county (Tong et al., 2016) as well as decreases of TGM at other sites (Slemr et al., 2011; Zhang et al., 2016)."

Lines 241-242: "TGM at the Nam Co station shows a seasonal variation with a maximum in the summer and a minimum in the winter". Is there a statistically significant difference?

Response: Yes, the sig. in Independent-Samples T-test was <0.01.

Lines 257-258: "the lower concentration of TGM at the Nam Co station in the winter might be indicative of atmospheric mercury depletion". The word "depletion" is rather connoted. It usually refers

to concentrations reaching near-zero values.

Response: Thank you for pointing this out. Sentence was changed in lines 286-289: "Compared to other high-altitude background sites in the mid-latitudes in Europe (Fig. 4) (Denzler et al., 2017; Fu et al., 2016a; Ebinghaus et al., 2002) and sites in mid-latitudes in the US (Holmes et al., 2010; Weiss-Penzias et al., 2003; Sigler et al., 2009; Yatavelli et al., 2006), the lower concentration of TGM at the Nam Co Station in the winter might be indicative of atmospheric mercury removal in the winter caused by reactive halogens (Br and Br_2)."

Lines 258: "The reaction rates for these reactions". Which specific reactions are you referring to?

Response: The reactions referred to the reactions between GEM and Br_2:

GEM + $Br_2 \rightarrow HgBr_2$, $k_3 = 0.9 \times 10^{-17} p \left(\frac{T}{298}\right)^{-2.86} cm^3 molec^{-1} s^{-1}$

and between GEM and Br:

GEM + $Br \rightarrow HgBr$, $k_3 = 3.6 \times 10^{-13} p \left(\frac{T}{298}\right)^{-2.86} cm^3 molec^{-1} s^{-1}$

Sentences were changed in lines 286-292: "Compared to other high-altitude background sites in the mid-latitudes in Europe (Fig. 4) (Denzler et al., 2017; Fu et al., 2016a; Ebinghaus et al., 2002) and sites in mid-latitudes in the US (Holmes et al., 2010; Weiss-Penzias et al., 2003; Sigler et al., 2009; Yatavelli et al., 2006), the lower concentration of TGM at the Nam Co Station in the winter might be indicative of atmospheric mercury removal in the winter caused by reactive halogens (Br and Br_2). The reaction rates for these reactions are a strong inverse function of temperature (de Foy et al., 2016b; Goodsite et al., 2004), and they are accompanied by lower surface ozone concentration (Yin et al., 2017), which is catalytically destroyed by halogens (Bottenheim et al., 1986; Obrist et al., 2011). ".

Line 259: "Accompanied by lower surface ozone concentration". Can you add ozone concentrations in Figure 4?

Response: Thanks for your suggestion. Figure was changed as you suggested in line 883 (Fig. 4).

Lines 263-265: “Higher temperature in the warm seasons might lead to remobilization of soil Hg re-emission, which has been evidenced by a recent study on surface-air Hg exchange in the northern Tibetan Plateau”. I would expect higher Hg re-emissions around midday. Similarly, Ci et al. (2016) showed that Hg(0) fluxes were higher in the daytime. See main comment #3.

Response: Yes, the study at Beiluhe found that Hg(0) fluxes were higher in the daytime. During the daytime, boundary layer was high and wind was strong. Then Hg(0) re-emissions from surface were diluted in the boundary layer generating low concentrations of atmospheric mercury.

Section 3.3. Diurnal variations of TGM: I don’t really see the point of the box model. See main comment #3.

Response: Please see the discussion above and the new text in Sec. 2.4 and Sec 3.3.

Line 285: “Constant depletion existed in the spring”. Use another word than “depletion”.

Response: Thank you for pointing this out and “depletion” was changed to “reduction” in lines 317-318: “Constant reductions existed in the spring, summer and autumn which would correspond to reduction rates of around 1 to 2 ng m^{-2} h^{-1}.”

Lines 290: “burst in the morning is probably due to prompt re-emission of nocturnal Hg deposition:”. Is this consistent with Hg(0) fluxes reported by Ci et al. (2016)? Additionally, can the low decrease at night really explain the high morning increase?

Response: In the study at Beiluhe, it was stated that Hg(0) flux showed a diurnal pattern with emission in the daytime and deposition in nighttime, and solar radiation had a great influence on Hg(0) exchange between air and surface. Hg(0) flux started to increase when photosynthetically active radiation observed. In addition with the measurements of wind speed and boundary layer height, indicating the condition of dilution of pollutants, we stated that “The atmospheric mercury bursts in the morning (7:00-

9:00) is probably due to prompt re-emission of nocturnal mercury deposition on the Earth's surface (Fu et al., 2016b; Howard et al., 2017; Kim 2010). The stable nocturnal boundary layer terminated at sunrise at which point mercury, including the mercury in the soil indigenously and/or deposited overnight, started to be reemitted into the shallow stable boundary layer before the increase of temperature which leads to an increase in the mixing height."

Line 297: "The higher surface ozone concentration and SWD during daytime (Fig.9)". Can you please add ozone concentrations in Figure 9?

Response: Figure was changed as suggested in line 957 (new number 8).

Line 299: "depletion of atmospheric mercury". Use another word than depletion.

Response: Thanks for pointing this out. "Depletion" was changed to "reduction."

Line 321: "The middle panel". There is no middle panel.

Response: Sentence was modified in lines 348-350: "Fig. 9 showed that a number of the high outliers are associated with specific peak events, indicating that occasional plumes of high TGM are not associated with recurring emissions or periodically occurring conditions."

Line 324: "with very low TGM concentrations". Please define "very low".

Response: Sentence was modified as suggested in lines 350-351: "Additionally, a few events with very low TGM concentrations were not simulated. They have an average concentration of 0.9 ng m^{-3}.".

Line 344: "the highest concentrations are very clearly associated with". The mean is about the same. You have more extreme values.

Response: Thank you for pointing this out. This is exactly what we mean: "TGM concentrations

above 2 ng m^{-3} are very clearly associated with cluster 4 which has transport from the east and through Lhasa, which was also probably due to the further impact from eastern Indo-Gangetic Plain and the possibility of episodic transport events from China.", and it was changed in lines 371-373.

Lines 355-363: Were you able to identify biomass burning plumes at Nam Co with high Hg(0) concentrations? The seasonality of biomass burning is not in line with TGM seasonality.

Response: Thanks for pointing out this. Currently, we are not attempt to identify biomass burning plumes at Nam Co with high Hg(0) concentrations in this study, and probably we will do that in the future. The impact of biomass burning through long-range transport was one of the potential influence factors to the seasonal variation of TGM at the Nam Co Station. The impact strength of biomass burning was variable due to the effect such as transport path. Re-emission and air masses mixing could also affect the seasonal variation of TGM at the Nam Co Station. It is a result of synthetic effect. PSCF results in this study proved that the high potential source areas of TGM at Nam Co in line with biomass burning in seasons.

Line 365: Replace "old mercury" by "legacy mercury".

Response: Thanks for your suggestion. This part was moved to the introduction as you suggested, and this sentence was removed.

Lines 366-369: This arrives too late in the manuscript. I suggest you move this section to the intro since you do not discuss any results here. See main comment #6.

Response: Thanks for your suggestion. This section was moved to the introduction as suggested, and this part was removed.

Line 369: "net sinks at night". Why do you parameterize Hg emissions from soils in the early evening?

Response: This section was moved to the introduction as suggested, and this part was removed. And we modified the description of box model in section 3.3.

Section 3.7. Implications for transboundary air pollution to the Tibetan Plateau: You don't really talk about implications, rather about the influence of the Indian summer monsoon on TGM seasonality. I suggest you move this to the section on TGM seasonality. See main comment #6.

Response: In section 3.7, we suggested that the Indian summer monsoon has important impact on the seasonal variation of TGM, more importantly, we highlighted that mercury, a representativeness of gaseous pollutants, differs in seasonal variation from other particulate pollutants. Therefore, we suggested that additional measurements of multiple pollutants and comparative studies are required to achieve a more comprehensive understanding and assessment of transboundary air pollution to the Tibetan Plateau. We would keep section 3.7 in the last so that our study can be more informative to wider readers.

Line 427: "extremely low TGM level". "extremely" is maybe too much here.

Response: Thanks for your suggestion. "extremely" was removed as you suggested and sentence was modified in lines 427-428: "The mean TGM concentration was 1.33 ± 0.24 ng m^{-3} during the whole measurement period and the low TGM level at the Nam Co Station indicated that the environment is pristine in the inland Tibetan Plateau."

Line 430-431: "the low concentration of TGM at the Nam Co station in the winter may be due to the depletion of mercury". Again, please use another word than depletion. Additionally, I am not really convinced by this explanation. Can't it just be explained by the back trajectories? According to Fig. 14, wintertime air masses are more "stagnant" over the Tibetan Plateau, with little long-range transport from polluted regions. The way I see it, you have background concentrations in wintertime, and higher concentrations in other seasons due to local re-emissions and long-range transport of pollution plumes. Do you have more frequent high outliers in summer vs. winter?

Response:

Thanks for your suggestion. For "depletion", sentence was modified as you suggested in lines 431-432: "Compared with other high-altitude background sites, the low concentration of TGM at the Nam Co Station in the winter may be due to the removal of mercury due to halogen."

Fig. 14 shows PSCF areas, not residence times. The residence times are actually lower in the winter as there are strong westerly winds impacting the measurement sites. Residence times increase in the summer as winds become more variable and there is slower transport from the south and east.

In this study, 91% of high outliers were in the summer.

Lines 437-438: "The box model provided supporting evidence and estimates of diurnal TGM deposition and TGM bursts of (re)emissions at the Nam Co Station in addition to dilution due to vertical mixing". I don't really see why. See main comment #3.

Response: Please see the new explanation of the rationale for the box model in Sec. 2.4 and Sec. 3.3 as well as the comment from reviewer #2 (line 180-185) that asks for this to be added to the conclusions.

Figure 1: I like this figure. However, can you add: - Standard deviation at each site -Date (year) at which monitoring was performed at each site (e.g., Nam Co station (Jan2012-Oct 2014)). See main comment #7.

Response: Thanks for your suggestion. Changed as suggested not in figure1 but in Table S1, due to the limited space in figure. Figure 1 was changed as the other reviewers suggested.

Figure 4: Similarly, can you add monthly standard deviation + date (year) at which monitoring was performed at each site? See main comment #7. Since you have too many figures, you can perhaps describe a little bit more the results in the manuscript and move this figure to SI.

Response: Thanks for your suggestion and figure was modified as suggested.

Because we were only able to obtain monthly mean concentrations of TGM at the other 3 sites, we are unable to add monthly standard deviation.

Figure 5: Why GEM or TGM? See main comment #1. Additionally, you can perhaps describe a little bit more the results in the manuscript and move this figure to SI.

Response: Thanks for your suggestion. Due to the different definitions of the measurements in their studies, GEM and TGM were used for different sites. At the Nam Co Station, the TGM fraction consists mostly of GEM (more than 98%). The operationally defined RGM accounted for less than 2% of TGM (Figure S1 in supplementary material in de Foy et al., 2016b). We consider the Tekran data to represent TGM in line with previous studies (e.g. Kock et al., 2005; Slemr et al., 2008; Müller et al., 2012).

Figure 9: This figure is rather difficult to read (too small). What is parameter q? A figure and its caption should form a self-contained element.

Response: Thanks for your suggestion. q is specific humidity and figure was modified in line 957 (new number 8).

Figure 10: Please remove the line for missing data.

Response: Changed as suggested.

Table S1: Please add: - Standard deviation - Year at which monitoring was performed - Instrumentation used (speciation unit or Tekran + PTFE filter). See main comment #1.

Response: Changed as suggested in Table S1.

Figure S1: Can you add the standard deviation for monthly mean concentrations (black squares)? Additionally, how many hourly values did you have to calculate the monthly mean in January 2013,

August 2013 and October 2014. It looks like you just have missing values.

Response: Thanks for your suggestion. Figure was changed as suggested.

There were 144 hourly values in January 2013, 452 hourly values in August 2013 and 90 values in October 2014. And due to the limited valid data, we removed January 2013 and October 2014 from figure.

Figure S2: Can you please add Nam Co station and Lhasa city? Additionally, can you add in the caption which emissions inventory you used and for which year?

Response: Thanks for your suggestion. Figure was change as suggested.

The information of anthropogenic mercury emissions inventory was stated in section 2.5: "These inventories were for the year 2010 and had a horizontal resolution of 0.5°×0.5°".

References

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Multi-year monitoring of atmospheric TGM at a remote high-altitude site (Nam Co, 4730 m a.s.l.) in the inland Tibetan Plateau

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Abstract

Total gaseous mercury (TGM) concentrations were continuously measured at the Nam Co Station, a remote high-altitude site (4730 m a.s.l.), in the inland Tibetan Plateau, China from January 2012 to October 2014 using a Tekran 2537B instrument. The mean concentration of TGM during the entire monitoring period was 1.33 ± 0.24 ng m^{-3} (mean ± standard deviation (SD)), ranking the lowest value among all continuous TGM measurements reported all over China, and was lower than most of sites in the Northern Hemisphere. This indicated the pristine atmospheric environment in the inland Tibetan Plateau. Long-term TGM at the Nam Co Station exhibited a slight decrease especially for summer seasons. The seasonal variation of TGM was characterized by higher concentrations during warm seasons and lower concentrations during cold seasons, decreasing in the following order: summer (1.50±0.20 ng m^{-3}) > spring (1.28±0.20 ng m^{-3}) > autumn (1.22±0.17 ng m^{-3}) > winter (1.14±0.18 ng m^{-3}). Diurnal variations of TGM exhibited uniform patterns in different seasons: the daily maximum was reached in the morning (around 2-4 hours after sunrise), followed by a decrease until sunset and a subsequent build-up at night, especially in the summer and the spring. Regional surface re-emission and vertical mixing were two major contributors to the temporal variations of TGM while long-range transported atmospheric mercury promoted elevated TGM during warm seasons. Results of multiple linear regression (MLR) revealed that humidity and temperature were the principal covariates of TGM. Potential source contribution function (PSCF) and FLEXible PARTicle dispersion model (WRF-FLEXPART) results indicated that the likely high potential source regions of TGM to the Nam Co are central and eastern Indo-Gangetic Plain (IGP) during the measurement period with high biomass burning and anthropogenic emissions. The seasonality of TGM at Nam Co was in phase with the Indian Monsoon Index, implying Indian Summer Monsoon as an important driver for transboundary transport of air pollution into the inland Tibetan Plateau. Our results provided atmospheric mercury baseline in the remote inland Tibetan Plateau and serve as new constraint for assessment of Asian mercury emission and pollution.

1 Introduction

Mercury (Hg) is one of the most toxic environmental pollutant because of the easy uptake of its organic forms by biota and the neurological and cardiovascular damage to humans resulting from bioaccumulation (Schroeder and Munthe, 1998). The majority of the mercury released to the environment is emitted into the atmosphere and can be transported from emission sources to deposition sites around the globe. Unlike other metals in the atmosphere, the majority of atmospheric mercury largely exists in the elemental form (Gaseous Elemental Mercury, GEM). The global residence time of GEM is in the range of 0.5-2 years due to its high volatility, low solubility and chemical stability (Schroeder and Munthe, 1998; Shia et al., 1999). It is therefore transported globally over long distances (tens of thousands of kilometers) far from pollution sources. Horowitz et al. (2017) recently reported that the chemical lifetime of tropospheric GEM against oxidation may be much shorter than previously reported: it could be as short as 2.7 months. GEM accounts for more than 95% of TGM (TGM, Total Gaseous Mercury. RGM, Reactive Gaseous Mercury. TGM= GEM + RGM). RGM and Hg-P (particle-bound mercury) compounds make up the remaining fraction of mercury in the atmosphere, and these two compounds have an estimated lifetime ranging from several days to a few weeks. RGM can be expected to be removed near a few tens to a few hundreds of kilometers from their source while Hg-P is likely to be deposited at intermediate distances of hundreds to thousands of kilometers (Schroeder and Munthe, 1998). RGM and Hg-P are generally depicted as local and regional pollutants, and the dry and wet deposition of RGM and Hg-P are much faster than GEM (Schroeder and Munthe, 1998; Lin and Pehkonen, 1999; Lindberg and Stratton, 1998).

East Asia and South Asia are two of the areas in the world with the fastest economic growth and the highest population density. These two areas are known for their heavily polluted air (Nair et al., 2007; Mukherjee et al., 2009), and anthropogenic mercury emissions in these areas are among the world's highest (Pirrone et al., 2010). China is the largest anthropogenic emitter of mercury worldwide with most of the emissions originating from coal combustion and non-ferrous smelting production (AMAP/UNEP, 2013; Pacyna et al, 2008). Geographically, most of China's mercury emissions are located in eastern and central China (Streets et al., 2005; Wu et al., 2016) (Fig. S1). Atmospheric mercury concentrations in Guizhou, one of the most important mercury producing and coal producing regions in China, was reported to be 6.2 - 9.7 ng m^{-3} of TGM in the capital city of Guiyang between 2001- 2009 (Feng et al., 2004; Liu et al., 2011; Fu et al., 2011). Measurements of

atmospheric mercury at background and remote sites in China include the following sites: Wuzhishan (2011-2012), Mt. Changbai (2008-2010), Mt. Waliguan (2007-2008), Mt. Ailao (2011-2012), Shangeri-La (2009-2010) and Mt. Gongga (2005-2006) with concentrations ranging from 1.58 to 3.98 ng m^{-3} (Liu et al., 2016; Fu et al., 2012b; Fu et al., 2012a; Fu et al. 2015; Zhang et al., 2015; Fu et al., 2008). Similarly, South Asia has serious problems of environmental pollution due to elevated mercury emissions (UNEP, 2013), resulting in hazardous mercury levels reported in water, lake sediment and fish samples (Karunasagar et al., 2006; Parvathi et al., 2010; Subramanian, 2004). Anthropogenic mercury emissions in South Asia were mostly in the Indo-Gangetic Plain (IGP) including most of northern and eastern India, the eastern parts of Pakistan, and all of Bangladesh (Fig. S1) all of which have high population density and many industrial centers. Biomass burning is another important source of atmospheric mercury, especially for TGM/GEM (Pirrone et al., 2010), and can lead to high TGM concentration events at sites far from the emissions (de Foy et al., 2012). Plenty of fire hot spots were observed in South Asia and East Asia including the IGP, the Indo-China Peninsula and southeastern China indicating the biomass burning at these areas (Fig. S2), while few biomass burning events were detected in the Tibetan Plateau (Fig. S2).

Located between South Asia and East Asia, the Tibetan Plateau is a vast high-altitude landform featured by remote and pristine environments. There are limited local anthropogenic activities in the Tibetan Plateau and previous studies reported that the atmospheric environment of the Tibetan Plateau remains global background levels (Fu et al., 2012a; Sheng et al., 2013; Xiao et al., 2012). Notably, mercury records from glaciers and lake sediments suggest that the Tibetan Plateau is an important part of the global mercury cycle, acting as both a sink (mercury deposition to snow) and a source (release of mercury from melting ice) (e.g., Kang et al., 2016; Yang et al., 2010; Sun et al., 2017, Sun et al., 2018). Further, it has been increasingly perceived that the inland of Tibetan Plateau can be influenced by trans-boundary air pollution such as black carbon originating from biomass burning in South Asia by crossing the Himalayas (Xia et al., 2011; Cong et al., 2015; Wan et al., 2015; Li et al., 2016). Studies of mercury in precipitation and water vapor evidenced that the Tibetan Plateau is likely sensitive to pollutant input including mercury (Huang et al., 2012; Huang et al., 2013), and the particulate-bound mercury in total suspended particulates was found at high concentrations in Lhasa with an average of 224 pg m^{-3} which was comparable to other cities in China (Huang et al., 2016). A few measurements of atmospheric mercury at sites on the fringes of the Tibetan Plateau reported TGM concentrations in the range of 1.98-3.98 ng m^{-3} (Fu et al., 2012a; Fu et al., 2008; Zhang et al., 2015), which were slightly

higher than the northern hemispherical background level, implying possible impact of anthropogenic emissions. In recent years, China and India signed the Minamata Convention and will probably control mercury emissions more strictly (Selin, 2014). Wu et al. (2017) stated that atmospheric mercury emissions from iron and steel production decreased from 35.6 Mg in 2013 to 32.7 Mg in 2015, and Pacyna et al. (2010) estimated that total mercury emissions in China would decrease from 635 Mg in 2005 to 290–380 Mg in 2020. Burger et al. (2013) estimated that total mercury emissions in India would increase from 310 Mg in 2010 to 540 Mg in 2020. In the context of serious mercury pollution and fast changes of regional mercury emission, atmospheric mercury observations at background sites neighboring regions of higher mercury pollution can provide a scientific basis for evaluating the extent of mercury pollution, for determining potential sources of atmospheric mercury and for informing public policy. The Nam Co Station, an inland site in the Tibetan Plateau, is an ideal site to determine the TGM of the inland Tibetan Plateau because it is rarely affected by locally anthropogenic emission of mercury.

In this study, high-time resolution TGM was measured at the Nam Co Station from January 2012 to October 2014 and the temporal characteristics of atmospheric mercury were studied. Comparison with meteorological data, Multiple Linear Regression (MLR) and a box model were used to investigate the temporal mercury variations at the Nam Co Station. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory), WRF-FLEXPART (FLEXible PARTicle dispersion model) and Potential Source Contribution Function (PSCF) were used to identify potential sources and impacts from long-range transport. The objective of this study is to (1) summarize the levels and temporal characteristics of TGM at a remote site in the inland Tibetan Plateau in a long-term measurement, (2) identify potential source regions of TGM at the Nam Co Station and (3) provide in-situ observational constraint that may contribute to understand changes in Asian mercury pollution.

2 Measurements and Methods

2.1 Measurement site

The Nam Co comprehensive observation and research station (namely the Nam Co Station, 30°46.44′ N, 90°59.31′ E, and 4730 m a.s.l.) is a remote site between Nam Co Lake and the Nyainqêntanglha mountain range (Fig. 1). The Nam Co Station has been established since 2005 for maintaining a long-term record of the meteorological, ecological, and atmospheric measurements in the Tibetan Plateau (Cong et al., 2007; Li et al., 2007; Kang et al., 2011; Huang et al., 2012; Liu et al., 2015;

de Foy et al., 2016b). There are restricted point sources of anthropogenic mercury emissions nearby the Nam Co Station. Dangxiong County is the nearest town on the southern slopes of the Nyainqêntanglha mountain range approximately 60 km south from Nam Co and Dangxiong is about 500 m lower than the Nam Co Station. Nomadism and tourism are the only human activities mostly during summer. Lhasa, the largest city in Tibet, is ~125 km south of the Nam Co Station. There was snow at the Nam Co Station discontinuously from October to March. But due to the strong wind at this period and the flat terrain surrounding the station, the snow did not remain on the ground for more than a few days at a time.

TGM measurements were conducted at the Nam Co Station starting on January 15, 2012 until October 4, 2014 (Fig. S3). Field operators checked the instruments and created a monitoring log file each day at the Nam Co Station. Measurements were intermittently interrupted because of equipment maintenance and unstable power supply due to damage from strong winds to the electrical wires at the Nam Co Station. All data displayed in this study are in UTC+8 and solar noon at the Nam Co Station is at 13:56 in UTC+8 (China Standard Time, Beijing Time).

2.2 Measurements: TGM, surface ozone and meteorology

Measurements of TGM concentrations were performed with a Tekran model 2537 B instrument (Tekran Instruments Corp., Toronto, Ontario, Canada). The Tekran 2537 B was installed in the monitoring house at the Nam Co Station and ambient air was introduced from the inlet which was 1.5 m above the roof and 4 m above the ground. A 45-mm diameter Teflon filter (pore size 0.2 μm) was placed in front of the inlet and it was changed every two weeks. The Tekran 2537 B measurements are based on the amalgamation of mercury onto a pure gold surface. By using a dual cartridge design, continuous measurements of mercury in the air can be made. The amalgamated mercury was thermally desorbed into an argon carrier gas stream and analyzed using an internal detector which was designed by cold vapor atomic fluorescence spectrophotometry (λ=253.7nm) (Landis et al., 2002) providing TGM analysis at sub-ng m^{-3} levels. The sampling interval of the Tekran 2537 B was 5 min and the sampling flow rate was 0.8 L min^{-1} (at standard temperature and pressure). The Tekran 2537 B was calibrated automatically every 25 hours using the internal mercury permeation source and was calibrated manually using a Tekran 2505 randomly 1-2 times a year. At the Nam Co Station, the TGM fraction consists mostly of GEM (more than 98%). The operationally defined RGM accounted for less than 2% of TGM (Figure S1 in supplementary material in de Foy et al., 2016b). We consider the

Tekran data to represent TGM in line with previous studies (e.g. Kock et al., 2005; Slemr et al., 2008; Müller et al., 2012).

Surface ozone was measured as a surrogate measure of oxidizing potential of the atmosphere (Stamenkovic et al., 2007) at the Nam Co Station using a UV photometric instrument (Thermo Environmental Instruments, USA, Model 49i) which uses absorption of radiation at 254 nm and has a dual cell design. The monitor was calibrated using a 49i-PS calibrator (Thermo Environmental Instruments, USA) before measurements and using aperiodic calibration during the monitoring periods. Details and analysis of the surface ozone measurements at the Nam Co Station were reported in Yin et al. (2017).

Measurements of temperature (T), relative humidity (RH), wind speed (WS), wind direction (WD) and downward shortwave radiation (SWD) were conducted at the Nam Co Station by a local weather station system (Milos 520, Vaisala Co., Finland) and a radiation measurement system (CNR1, Kipp & Zonen Co., US), respectively (Ma et al., 2008).

2.3 Meteorological simulations

Gridded meteorological data for backward trajectories were obtained from the Global Data Assimilation System (GDAS-1) of the U.S. National Oceanic and Atmospheric Administration (NOAA) with 1°×1° latitude and longitude horizontal resolution and vertical levels of 23 from 1000 hPa to 20 hPa (http: // www. arl. noaa. gov/ gdas1. php).

Backward trajectories and clusters were calculated using the NOAA-HYSPLIT model (Draxler and Rolph, 2003, http://ready.arl.noaa.gov/HYSPLIT.php) using TrajStat (Wang et al., 2009), which is a free software plugin of MeteoInfo (Wang, 2014). The backward trajectories arrival height in HYSPLIT was set at 500 m above the surface and the total run times was 120 hours for each backward trajectory. Results of air masses at different heights (500m, 1000m and 1500m) showed similar patterns, hence, we selected trajectories released at a height of 500 m as representative since 500 m is suitable for considerations of both the long-range transport and transport in the planetary boundary layer. Trajectory positions were stored at time intervals of 3 hours. Angular distance was chosen to calculate clusters in HYSPLIT calculation. HYSPLIT backward trajectories were used to calculate the Potential Source Contribution Function (section 2.6) which serves to investigate the potential sources contributing to atmospheric mercury at the Nam Co Station.

In addition to HYSPLIT, WRF-FLEXPART (Brioude et al., 2013) was used to obtain clusters of particle trajectories

reaching the Nam Co Station. 1000 particles were released per hour in the bottom 100 m surface layer above the Nam Co Station and were tracked in backward mode for 4 days (de Foy et al., 2016b). The use of two different trajectory models (HYSPLIT and WRF-FLEXPART) with different input meteorology can add robustness to the discussion as was done for the ozone study at Nam Co (Yin et al., 2017). Furthermore, the WRF-FLEXPART simulations were some of the parameters used in the multiple linear regression model (section 2.4). Residence Time Analysis (RTA) (Ashbaugh et al., 1985) was utilized to show the dominant transport paths of air masses impacting the samples (Wang et al., 2016; Wang et al., 2017). Six clusters were found to represent the prevailing flow patterns to the Nam Co Station simulated using WRF-FLEXPART.

2.4 Multiple linear regression model and box model

A Multiple Linear Regression (MLR) model was used to quantify the main factors affecting the hourly concentrations of TGM. The method follows the description provided in de Foy et al. (2016a; 2016c) and de Foy (2017) and was used to analyze surface ozone concentrations at the Nam Co Station (Yin et al., 2017). The inputs to the MLR model include meteorological parameters (wind speed, temperature, solar radiation and humidity), surface ozone, inter-annual variation factors, seasonal factors, diurnal factors, WRF boundary layer heights, WRF-FLEXPART trajectory clusters and a CAMx stratospheric ozone tracer (see Yin et al. (2017) for more details). Briefly, the inter-annual factors are separate scaling factors for each year of the measurements, the seasonal factors are 12-month and 6-month harmonic terms (sine and cosine), and the diurnal factors are scaling factors for each hour of the day. The inputs to the model were normalized linearly. An Iteratively Reweighted Least Squares (IRLS) procedure was used to screen for outliers. Measurement times when the model residual was greater than two standard deviations of all the residuals were excluded from the analysis. This was repeated iteratively until the method converged on a stable set of outliers. The variables to be included in the regression were obtained iteratively. At each iteration, the variable leading to the greatest increase in the square of Pearson's correlation coefficient was added to the inputs as long as the increase was greater than 0.005.

The distribution of TGM concentrations is approximately normal (see details in section 3.1), and so a linear model was used. TGM was scaled linearly to have a mean of 0 and a standard deviation of 1 in the regression model. A Kolmogorov-Zurbenko filter (Rao et al., 1997) was used to separate the time series of specific humidity and temperature into a synoptic

194 scale signal (> 3-5 days) and a diurnal scale signal using 5 passes of a 13-point moving average. Only the synoptic scale signal
195 was included in the final regression results, as the diurnal variation was characterized by the other variables in the analysis.
196 The other meteorological parameters used were the 24-hour average boundary layer height from WRF and the 8-hour local
197 measured wind speeds (4 directions, 5 wind speed segments for a total of 20 factors corresponding to different wind speeds
198 from different wind directions). The 24-hour average of ozone measurements (log-transformed) contributed to the model. In
199 addition, a seasonal K-Z filtered time series of a CAMx tracer for transport from the free troposphere (above 300 hPa) to the
200 surface contributed to the model.

201 TGM at the Nam Co Station is expected to be well mixed and the site is not influenced by local sources. It is therefore
202 expected that a box model should be able to reproduce the diurnal profile of concentrations. A box model that accurately
203 simulates the diurnal profile of TGM would provide constraints on known processes affecting the concentrations. Comparisons
204 with measured profiles would further identify missing processes in the model. This approach was used for reactive mercury at
205 the same site, where it identified the role of the reduction of reactive mercury to gaseous elementary mercury mediated by
206 sunlight (de Foy et al., 2016b). A box model was made that included free parameters to represent known chemical reactions
207 and dispersion processes. An optimization algorithm was used to identify the parameters required to fit the model to the data,
208 as was done in de Foy et al. (2016b). Preliminary tests of the box model were made using solar radiation and temperature to
209 represent chemical transformations, as well as using wind speed and boundary layer height to represent dilution. However
210 these attempts failed to reproduce the diurnal variation found in the measurements. A simplified exploratory model was
211 therefore sought that would represent the measured diurnal variations as simply as possible, according to Occam's razor
212 (Larsen et al., 2014). Although this model does not yield direct information on known processes, it does identify the kinds of
213 processes and their magnitude that would be required to accurately represent the measured diurnal profile. The final model
214 combined the following 5 inputs: TGM increases at sunrise and in the early evening, constant TGM reductions 24 hours a day,
215 a constant lifetime for TGM loss during daylight hours and TGM dilution due to vertical mixing.

216 **2.5 Anthropogenic mercury emissions and fire hot spots distribution**

217 The mercury emission inventory of China was obtained from Wu et al. (2016), which used a technology-based approach
218 to compile a comprehensive estimate of Chinese provincial emissions for all primary anthropogenic sources. The emissions

over other Asian countries were from UNEP global anthropogenic emission inventory (AMAP/UNEP, 2013). These inventories were for the year 2010 and had a horizontal resolution of 0.5°×0.5°.

MODIS fire spots were obtained from 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).

2.6 Potential Source Contribution Function (PSCF)

PSCF assumes that back-trajectories arriving at times of higher mixing ratios likely point to the more significant source directions (Ashbaugh et al., 1985). PSCF has been applied in previous studies to locate sources of TGM for different sites (Fu et al., 2012a; Fu et al., 2012b; Zhang et al., 2015). The PSCF values for the grid cells in the study domain are based on a count of the trajectory segment (hourly trajectory positions) that terminate within each cell (Ashbaugh et al., 1985). Let n_{ij} be the total number of endpoints that fall in the *ij*th cell during whole simulation period. Let m_{ij} represents the number of points in the same cell that have arrival times at the sampling site corresponding to TGM concentrations higher than a set criterion. In this study, we calculate the PSCF based on trajectories corresponding to concentrations that exceed the mean level (1.33 ng m^{-3}) of TGM. The PSCF value for the *ij*th cell is then defined as:

$$PSCF_{ij} = m_{ij}/n_{ij}$$

The PSCF value can be interpreted as the conditional probability that the TGM concentration at measurement site is greater than the mean mixing ratios if the air parcel passes though the *ij*th cell before arriving at the measurement site. In cells with high PSCF values are associated with the arrival of air parcels at the receptor site that have TGM concentrations that exceed the criterion value. These cells are indicative of areas of 'high potential' contributions for the chemical constituent.

Identical $PSCF_{ij}$ values can be obtained from cells with very different counts of back-trajectory points (e.g. grid cell A with m*ij*=5000 and n*ij*=10000 and grid cell B with m*ij* = 5 and n*ij* = 10). In this extreme situation grid cell A has 1000 times more air parcels passing through than grid cell B. Because of the sparse particle count in grid cell B, the PSCF values are more uncertain and the contribution from B is limited. To account for the uncertainty due to low values of n*ij*, the PSCF values were scaled by a weighting function W_{ij} (Polissar et al., 1999). The weighting function reduced the PSCF values when the total number of the endpoints in a cell was less than about three times the average value of the end points per each cell. In this case,

W_{ij} was set as follows:

$$W_{ij} = \begin{cases} 1.00 & n_{ij} > 3N_{ave} \\ 0.70 & 3N_{ave} > n_{ij} > 1.5N_{ave} \\ 0.42 & 1.5N_{ave} > n_{ij} > N_{ave} \\ 0.05 & N_{ave} > n_{ij} \end{cases} \quad (1)$$

where N_{ave} represents the mean n_{ij} of all grid cells. The weighted PSCF values obtained by multiplying the original PSCF values by the weighting factor: weighted PSCF result=W_{ij}×PSCF.

3 Results and discussion

3.1 TGM concentrations

The mean TGM concentration at the Nam Co Station is 1.33±0.24 ng m^{-3}, which is the lowest among all reported TGM concentrations at remote and rural sites in China (Liu et al., 2016; Fu et al., 2012b; Fu et al., 2012a; Fu et al. 2015; Ci et al., 2011; Dou et al., 2013; Zhang et al., 2015; Fu et al., 2010; Li et al., 2011; Zhang et al., 2013; Yu et al., 2015; Fu et al., 2008; Chen et al., 2013). The mean concentration of TGM is slightly lower than the annual mean concentration at background sites in the Northern Hemisphere (1.55 ng m^{-3} in 2013 and 1.51 ng m^{-3} in 2014), and higher than those in the Southern Hemisphere (0.93 ng m^{-3} in 2013 and 0.97 ng m^{-3} in 2014) (Sprovieri et al., 2016). Comparable results were reported from EvK2CNR on the south slope of the Himalayas (1.2 ng m^{-3}, Gratz et al., 2013), and from tropical sites in the Global Mercury Observation System in the Northern Hemisphere (1.23 ng m^{-3} in 2013 and 1.22 ng m^{-3} in 2014) (Sprovieri et al., 2016). Comparing to the three sites at the edge of the Tibetan Plateau (Mt. Waliguan, Shangri-La and Mt. Gongga, Table S1), the mean TGM concentration at the Nam Co Station was substantially lower, indicating that the inland Tibetan Plateau has a more pristine environment than the edges of the plateau.

The frequency distribution of TGM at the Nam Co Station was normally distributed (Fig. S4). 81% of hourly average TGM concentrations were in the range of 1.0-1.6 ng m^{-3} with few episodically elevated TGM and low TGM concentrations. 1.6% (n=236) out of all hourly mean TGM data (n=14408) were greater than 1.81 ng m^{-3} (overall mean TGM + 2×SD, namely 1.33+2×0.24=1.81), and 1.5% (n=213) were lower than 0.85 ng m^{-3} (overall mean TGM - 2×SD, namely 1.33-2×0.24=0.85).

The monthly average TGM at the Nam Co Station showed a weak decrease (slope = -0.006) during the entire monitoring period, and the decrease was more pronounced in the summer (slope = -0.013). Despite the short time span of the TGM time

series with some missing data mostly in the winter, the slight decrease of TGM especially in the summer was in agreement with a recent study using plant biomonitoring which identified a decreasing atmospheric mercury since 2010 near Dangxiong county (Tong et al., 2016) as well as decreases of TGM at other sites (Slemr et al., 2011; Zhang et al., 2016).

3.2 Seasonal variations of TGM

In contrast with many previous observations in China (Zhang et al., 2015; Fu et al., 2008b; Fu et al., 2009; Fu et al., 2010; Fu et al., 2011; Fu et al., 2012b; Feng et al., 2004; Xiu et al., 2009; Xu et al., 2015; Wan et al., 2009) and most AMNet (Atmospheric Mercury Network) sites (Lan et al., 2012) , TGM at the Nam Co Station showed a seasonal variation with a maximum in the summer (June, July and August) and a minimum in the winter (December, January and February) (Fig. 2). The seasonal mean TGM values decreased in the following order: summer (1.50±0.20 ng m^{-3}) > spring (1.28±0.20 ng m^{-3}) > autumn (1.22±0.17 ng m^{-3}) > winter (1.14±0.18 ng m^{-3}) (Table 1). The highest monthly mean TGM concentration of 1.54 ng m^{-3} in July was 0.43 ng m^{-3} higher than the lowest of 1.11 ng m^{-3} in November.

Measurements of TGM in other sites in the Tibetan Plateau also reported diverse seasonal patterns (Fig. 3). For example, Fu et al. (2012a) found that at Waliguan the maximum TGM concentration was in January 2008, resulting from long-range transport of pollutions from Northern India. Aside from January, monthly mean TGM concentrations at Waliguan had a clear trend with high levels in warm seasons, and lower levels in cold seasons. The TGM variation at Mt. Gongga (Fu et al., 2008) had a minimum in the summer, possibly due to the accelerated oxidation followed by dry deposition and wet scavenging processes in the summer. The winter maximum of TGM at Mt. Gongga (Fu et al., 2008) implied the impact from anthropogenic mercury emissions in the cold months. The seasonal variation of TGM at Shangri-La (Zhang et al., 2015) had high levels in the spring and autumn, and low levels in the summer and winter which was different from all the other sites in the Tibetan Plateau.

Compared to other high-altitude background sites in the mid-latitudes in Europe (Fig. 4) (Denzler et al., 2017; Fu et al., 2016a; Ebinghaus et al., 2002) and sites in mid-latitudes in the US (Holmes et al., 2010; Weiss-Penzias et al., 2003; Sigler et al., 2009; Yatavelli et al., 2006), the lower concentration of TGM at the Nam Co Station in the winter might be indicative of atmospheric mercury removal in the winter caused by reactive halogens (Br and Br_2). The reaction rates for these reactions are a strong inverse function of temperature (de Foy et al., 2016b; Goodsite et al., 2004), and they are accompanied by lower

surface ozone concentration (Yin et al., 2017), which is catalytically destroyed by halogens (Bottenheim et al., 1986; Obrist et al., 2011).

The summer peak of TGM at the Nam Co Station may be related to both the local re-emission of mercury from the earth's surface, and the long-range transport of mercury from South Asia (see details in section 3.5). At the Nam Co Station, daily mean TGM had a correlation coefficient with daily mean temperature reaching 0.56. Higher temperature in the warm seasons (Fig. 5) might lead to remobilization of soil mercury re-emission, which has been evidenced by a recent study on surface-air mercury exchange in the northern Tibetan Plateau (Ci et al., 2016). It is also possible that weaker wind speeds during the warm season (Fig. 5) suppressed the dilution of TGM with fresh air aloft in a low boundary layer. Furthermore, most precipitation happens in the summer at the Nam Co Station (You et al., 2007) and can increase emission of mercury from the Earth's surface by physical displacement of interstitial soil air by the infiltrating water (Ci et al., 2016) and by additional input of mercury from wet deposition (Huang et al., 2012). Besides local emissions, the summer monsoon can facilitate the transport of air masses with higher TGM concentrations from South Asia, and hence may also contribute to the summer peak of TGM.

The month of April in both 2012 and 2013 had higher monthly TGM levels than the months before and after (Fig. S3), possibly resulting from mercury emission from Nam Co Lake as the lake started to thaw in April (Gou et al., 2015).

3.3 Diurnal variations of TGM

Diurnal variations of TGM in different seasons exhibited a regular pattern, characterized by a sharp rise shortly after sunrise and a fairly steady decrease from the morning peak until sunset (Fig. 6). After sunset, TGM increased until midnight in the summer, the spring and the autumn. The diurnal variation of TGM at the Nam Co Station was similar to those of Mt. Gongga (Fu et al., 2009), Mt. Leigong (Fu et al., 2010), Mt. Changbai (Fu et al., 2012b), Mt. Waliguan (Fu et al., 2012a) and Reno (Peterson et al., 2009) except that the morning increase occurs earlier and is shorter compared during other sites that have a gradual increase throughout the morning.

Fig. 7 showed the comparison of TGM concentrations with a box model simulation by seasons. The best match in the box model was obtained by using variables including constant TGM reduction throughout the day, TGM increases at sunrise, TGM increases in the early evening, TGM dilution due to vertical mixing and a lifetime of TGM loss during daylight hours (Table 2). The R^2 of the model simulation ranged from 0.91 to 0.99, suggesting that the simulations reproduced the diurnal variations

accurately. As described above, both the measurements and the model have sharp bursts of TGM in the morning (7:00-9:00) and in the evening (18:00-22:00) during all seasons. Constant reductions existed in the spring, summer and autumn which would correspond to reduction rates of around 1 to 2 ng $m^{-2} h^{-1}$.

Fig. 8 showed the seasonal diurnal profiles of TGM and meteorological parameters. TGM concentrations were stable or slightly decreasing after midnight (0:00-6:00) under shallow nocturnal boundary layers. Notably, the morning increase of TGM happens immediately after sunrise, but before the increases of temperature, wind speed or humidity. The atmospheric mercury bursts in the morning (7:00-9:00) is probably due to prompt re-emission of nocturnal mercury deposition on the Earth's surface (Fu et al., 2016b; Howard et al., 2017; Kim 2010). The stable nocturnal boundary layer terminated at sunrise at which point mercury, including the mercury in the soil indigenously and/or deposited overnight, started to be reemitted into the shallow stable boundary layer before the increase of temperature which leads to an increase in the mixing height. As the temperature and radiation increased, so did the boundary layer height which developed into a convective mixed boundary layer and generated greater vertical mixing between the surface and loft. At the same time, the surface wind speed also increased. With increased vertical and horizontal dispersion, TGM released from the surface was diluted during the daytime (Liu et al., 2011; Lee et al., 1998). When the temperature decreased and the boundary layer converted back into a nocturnal boundary layer after sunset, depressed vertical mixing facilitated the build-up of TGM and such build-up was more significant in the warm seasons. In the evening, increases in TGM correspond to increases in specific humidity, especially in the summer.

3.4 Multiple linear regression and WRF-FLEXPART clusters results

Results of the MLR simulations for the entire measurement period (2012-2014) had a close correlation with the measurements: the correlation coefficient was 0.77 for all 12649 data points and 0.84 excluding the 383 outliers (Fig. 9). The primary contributor to the variance of the simulated time series was the seasonal signal, including the 12-month and 6-month harmonics as well as the smoothed specific humidity and temperature time series (Table 3). These were grouped together when presenting the results because they were not orthogonal to each other, and they contributed 84% of the variance of TGM in MLR simulation. The diurnal factors accounted for 4% of the variance, the WRF boundary layer heights accounted for 4% of the variance, and the local winds were associated with 1% of the variance. These factors showed that there was an impact from horizontal and vertical dispersion as well as daily cycling patterns due to either transport or chemistry, but that these factors

were considerably smaller than the seasonal variation at the site. Only 1% of the variance was associated with the annual signal, showing that the decrease in the concentrations reported in Sec. 3.1 was a small contributor to variations in TGM at Nam Co. The time series of surface ozone concentration contributed 3% to the variance and the stratospheric ozone tracer contributed 3%. We hypothesized that this was because ozone concentrations acted as an indicator of the oxidative potential of the air mass, although in the case of surface ozone concentration it could also be because they were a tracer of aged polluted air masses.

The regression analysis screens for high and low outliers. In particular, high outliers were significant in terms of TGM concentrations: they had an average concentration of 1.91 ng m^{-3} which is 0.58 ng m^{-3} higher than the average of the measurements retained in the simulations (Fig. 9). Fig. 9 showed that a number of the high outliers are associated with specific peak events, indicating that occasional plumes of high TGM are not associated with recurring emissions or periodically occurring conditions. A significant amount of TGM not accounted-for in the model was due to the high outliers. Additionally, a few events with very low TGM concentrations were not simulated. They have an average concentration of 0.9 ng m^{-3}. Fig. 10a showed the 6 wind transport clusters based on the hourly WRF-FLEXPART simulations. The figure showed the average residence time analysis for all the hours in each cluster, which characterizes the path of the air masses arriving at the measurement site for each cluster. The most frequent clusters were clusters 1 and 2 which accounted for 30% and 34% of measurement hours respectively. For measurement times during these clusters, the air masses clearly came from the west with a slight southern component for cluster 1 and a slight northern component in the case of cluster 2. Cluster 3 represented hours influenced by transport from the north which occurred during 15% of the measurement period. These were associated with the passage of storms at Nam Co: as the low pressure system moved to the east, the winds shifted from northwesterly to northeasterly. Clusters 4, 5 and 6 occurred less frequently and all represented different types of wind transport across the Himalayas from the south. Cluster 4 was the least frequent cluster, occurring 5% of the time. It included transport from the southeast including the northeastern corner of the Indo-Gangetic plain and occasional transport from southwestern China. This cluster also included transport from the direction of Lhasa. Cluster 5 occurred 7% of the time and represents transport from the south including Bangladesh. Cluster 6 occurred 9% of the time and included transport from Nepal and northern India.

The WRF-FLEXPART clusters were included in the MLR analysis and helped to improve the simulations for several tests. However, they did not increase the correlation coefficient of the final regression time series and consequently were not

included in the final MLR results. This could be because transport was already characterized by the other variables in the model such as temperature and humidity (which can serve as tracers of different air masses) and local wind speed and direction. Nevertheless, the importance of air mass transport can be seen from the probability density function of the TGM concentrations by cluster shown in Fig. 10b. Clusters 1 and 2, which had transport from the west, clearly had the lowest TGM concentrations. Next in terms of increasing TGM concentrations were clusters 3 and 6 which had transport from the north and from the southwest. TGM concentrations above 2 ng m^{-3} are very clearly associated with cluster 4 which has transport from the east and through Lhasa, which was also probably due to the further impact from eastern Indo-Gangetic Plain and the possibility of episodic transport events from China. Of the 87 hours with concentrations higher than 2 ng m^{-3}, 59% occurred during cluster 4 and 17% during cluster 5 with less than 8% for each of the other clusters. This demonstrated clearly that in addition to having the highest average levels, clusters 4 and 5 accounted for most of the peak concentrations.

3.5 HYSPLIT and PSCF results

Backward trajectories were calculated using HYSPLIT to identify the origins of air masses and associated TGM concentrations to the Nam Co Station. Most HYSPLIT trajectories originated from the west of Nam Co including the western and central Tibetan Plateau, the southwestern part of the Xinjiang Uygur Autonomous Region, South Asia, Central Asia and Western Asia. Very few trajectories originated from eastern China (Fig. S5). The backward trajectories were grouped into 6 clusters. Cluster 3 indicated the air mass from the south, originating from Bhutan and Bangladesh. This cluster had the lowest starting heights as well as traveling heights, but the highest mean TGM concentration (1.48 ng m^{-3}) (Table S2) in agreement with the FLEXPART results (Sec. 3.4). Clusters 1, 2, 4, 5 and 6 originated in the west, including air masses originating from northern India, Pakistan, Afghanistan and Iran passed over the Himalayas before arriving at the Nam Co Station. They had longer pathways through the Tibetan Plateau than Cluster 3. Cluster 4 had the longest transport route from the west, suggestive of faster wind speeds, and also the lowest TGM mean concentration (1.12 ng m^{-3}) with relatively high transport height.

PSCF calculations were based on concurrent TGM measurements and HYSPLIT backward trajectories, and thus can further constrain the potential source regions. Areas including IGP, the southern part of the Xinjiang Uygur Autonomous Region, the western part of Qinghai province and areas near the Nam Co Station in the Tibet Autonomous Region were identified as overall high potential sources regions and pathways (Fig. S6). Except for the areas near the Nam Co Station, these

potential sources regions correspond well with the atmospheric mercury emissions and biomass burning. The Bay of Bengal was identified as a potential source region probably due to high emissions from its surroundings associated with frequent occurrence of trajectories passing through this area in the summer.

Seasonal PSCFs were calculated in 2012 to investigate the potential sources by seasons (Fig. 11). In the spring, the autumn and the winter, the Nam Co Station was dominated by the Westerlies. Pollutants from South Asia might be diluted by the clean air during the transport within the Tibetan Plateau before they arrived at the Nam Co Station (Fig. S7). A zonal region in the central IGP (Fig. 11) with elevated pollution represents a constant potential source (Gautam et al., 2011; Mallik and Lal, 2014). The significant impact of long-range transport pollution from northwestern India on the Tibetan Plateau was also evidenced by TGM measurements at Waliguan (Fu et al., 2012a). In the summer, the Indian Monsoon prevails and air masses arrived at the Nam Co Station that had shorter pathway after entering the Tibetan Plateau than those in other seasons (Fig. S7). The central IGP was again found to have higher PSCF values than other regions, even though these were much lower than the PSCF values of other seasons. The highest PSCF values in the summer were in the eastern IGP (Fig. 11). For all seasons, the region near the Nam Co Station, especially its south and west, was high in PSCF values all through the year, indicating that air masses with high TGM concentrations predominantly came from the south-southwest.

3.6 Implications for transboundary air pollution to the Tibetan Plateau

The seasonal atmospheric circulation pattern in the Tibetan Plateau was characterized by the Indian monsoon in the summer and the Westerlies in the winter. Such a climate regime exerted a profound impact on the seasonal atmospheric environment by affecting the air transport dynamic and associated climate conditions. Pollutants like black carbon and hexachlorocyclohexanes peaked in pre-monsoon season and declined during monsoon season at Nam Co and Lulang, resulting from seasonal rainfall variations that can scavenge aerosols during their transport from source regions to the Tibetan Plateau (Zhang et al., 2017; Wan et al., 2015; Sheng et al., 2013). In contrast, gaseous pollutants showed different seasonal patterns: TGM at Nam Co in this study and persistent organic pollutants (dichlorodiphenyltrichloroethane and polychlorinated biphenyls) at Lulang showed higher concentrations during the monsoon season compared to the pre-monsoon season (Sheng et al., 2013). TGM at Nam Co showed strong covariance with temperature and specific humidity, all of which are in phase with the Indian Monsoon Index (IMI) (Wang and Fan, 1999; Wang et al., 2001) (Fig. 12), indicating the importance of Indian Summer

Monsoon as a major driver delivering of transboundary transport of air pollution into the inland Tibetan Plateau. We suggested that gaseous pollutants were not readily deposited and/or washed out by precipitation during their transport and were more likely associated with the transport dynamics driven by the Indian Summer Monsoon, hence they showed high values when the Indian Summer Monsoon prevails. Transboundary air pollution was not the sole factor contributing to elevated TGM during summer: temperature-dependent processes such as gas-particle fractionation and surface reemission can also contribute to such seasonal patterns. Nonetheless, the close relationship between TGM and the Indian Summer Monsoon and the clear difference in seasonal patterns between gaseous and particulate pollutants together indicated that additional measurements of multiple pollutants and comparative studies are required to achieve a more comprehensive understanding and assessment of transboundary air pollution to the Tibetan Plateau.

4 Conclusions

We conducted three-years of TGM measurements at the Nam Co Station in the inland area of the Tibetan Plateau, China, from January 2012 to October 2014. The mean TGM concentration was 1.33 ± 0.24 ng m^{-3} during the whole measurement period and the low TGM level at the Nam Co Station indicated that the environment is pristine in the inland Tibetan Plateau. A weak decrease of TGM was identified over the course of the measurements.

In contrast to many other sites in China, TGM at the Nam Co Station showed high concentrations in warm seasons and low concentrations in cold seasons. Compared with other high-altitude background sites, the low concentration of TGM at the Nam Co Station in the winter may be due to the removal of mercury due to halogen. Seasonal variation of TGM at the Nam Co Station was influenced by factors such as re-emission processes of deposited mercury over the Earth's surfaces, vertical mixing and long-range transport. Multiple linear regression, backward trajectories and PSCF were investigated at the Nam Co Station and results indicated that long-range transports from the central and eastern Indo-Gangetic Plain were potentially the main sources for seasonally elevated TGM at the Nam Co Station due to the alternate impact of the Westerlies and of the Indian monsoon. Peak concentrations of TGM at the Nam Co Station were associated with air masses from the eastern Indo-Gangetic Plain with the possibility of episodic transport events from China.

At the Nam Co Station, the diurnal TGM profile had a peak 2-3 hours after sunrise and reached its lowest concentration before sunset. An exploratory box model simulation shows that this diurnal profile can be accurately represented using TGM

reductions 24 hours per day, TGM increases near sunrise and sunset, and dilution due to vertical mixing. Daily meteorology conditions, such as high temperature, high solar radiation and more precipitation facilitated the Earth's surface mercury emission. The decline of TGM concentrations in the daytime was likely due to vertical dilution from increased vertical mixing, as well as due to the conversion of GEM to oxidized species that are easily deposited.

Due to the insolubility of TGM, which is different from particulate pollutant, TGM was less affected by the precipitation during the transport in monsoon season and measurement of TGM at the Nam Co Station can continually reflect the transboundary air pollution from the South Asia to the inland Tibetan Plateau.

The measurements of TGM at the Nam Co Station will be useful in providing atmospheric mercury baseline in the remote inland Tibetan Plateau, improving the accuracy of modeled concentrations of TGM in the inland Tibetan Plateau, and serving as new constraint for assessment of Asian mercury emission and pollution.

Data availability. All the data presented in this paper can be made available for scientific purposes upon request to the corresponding authors (Qianggong Zhang (qianggong.zhang@itpcas.ac.cn) or Shichang Kang (shichang.kang@lzb.ac.cn)).

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762
763 **Table 1. The statistics of TGM and meteorological variables in different seasons at the Nam Co Station during the measurement**
764 **period (2012-2014).**

Period	Statistical	TGM (ng m^{-3})	T (℃)	RH (%)	WS (m s^{-1})
Total	Mean	1.33	-0.29	50.67	3.32
	Median	1.34	0.30	50.00	2.80
	Standard Deviation	0.24	8.98	22.37	2.22
	Minimum	0.23	-28.90	5.30	0.00
	Maximum	3.14	19.00	98.00	15.60
	Count	14408	20695	20695	20695
Spring (MAM)	Mean	1.28	-0.90	51.58	3.21
	Median	1.30	-0.60	50.30	2.80
	Standard Deviation	0.20	6.48	24.38	2.11
	Minimum	0.42	-21.20	5.30	0.00
	Maximum	2.41	17.90	98.00	12.80
	Count	4506	4980	4980	4980
Summer (JJA)	Mean	1.50	8.80	63.32	2.94
	Median	1.50	8.60	65.30	2.60
	Standard Deviation	0.20	3.59	18.25	1.74
	Minimum	0.23	-4.10	11.00	0.00
	Maximum	3.14	19.00	97.00	11.10
	Count	5243	5805	5805	5805
Autumn (SON)	Mean	1.22	-0.78	47.06	3.36
	Median	1.20	-0.40	46.00	2.90
	Standard Deviation	0.17	7.23	20.55	2.07
	Minimum	0.87	-24.80	8.00	0.00
	Maximum	2.68	14.60	97.00	12.90
	Count	2267	4800	4800	4800
Winter (DJF)	Mean	1.14	-9.57	38.81	3.83
	Median	1.13	-9.00	36.00	3.00
	Standard Deviation	0.18	6.40	18.36	2.78
	Minimum	0.45	-28.90	7.00	0.00
	Maximum	2.08	5.20	91.70	15.60
	Count	2392	5110	5110	5110

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Table 2. Statistics of free parameters in the box model of TGM at the Nam Co Station by seasons.

	Initial TGM (ng m^{-3})	Morning TGM (7-9) burst (ng $m^{-2}h^{-1}$)	Evening TGM (18-22) burst (ng $m^{-2}h^{-1}$)	Constant TGM deposition (ng $m^{-2}h^{-1}$)	Free tropospheric TGM (ng m^{-3})	TGM lifetime during daylight (day)	root-mean-square error (RMSE)	R^2
Spring	1.288	58.29	37.66	-1.658	1.228	3.183	0.00983	0.96
Summer	1.521	14.2	25.65	-1.775	1.553	5.991	0.00796	0.91
Autumn	1.211	53.34	9.144	-1.061	1.036	Inf	0.0086	0.93
Winter	1.115	52.92	2.468	0	1.168	2.984	0.00368	0.99

Table 3. Contribution from the different groups to the total variance of the model. The standard deviation of each group gives a sense of the contribution of each group to the variance in units of ng m^{-3}. The variance contribution shows the percentage that each group contributes to the total variance of the model.

Group name	No. Variables	Std (ng m^{-3})	Variance Contribution (%)
Seasonal Signal	6	0.161	83.70
Diurnal Signal	24	0.036	4.08
WRF PBLH	5	0.034	3.81
Surface O_3 Conc	1	0.032	3.20
Strat. O_3 Tracer	1	0.031	3.04
Local Winds	20	0.020	1.34
Annual Signal	43	0.016	0.86

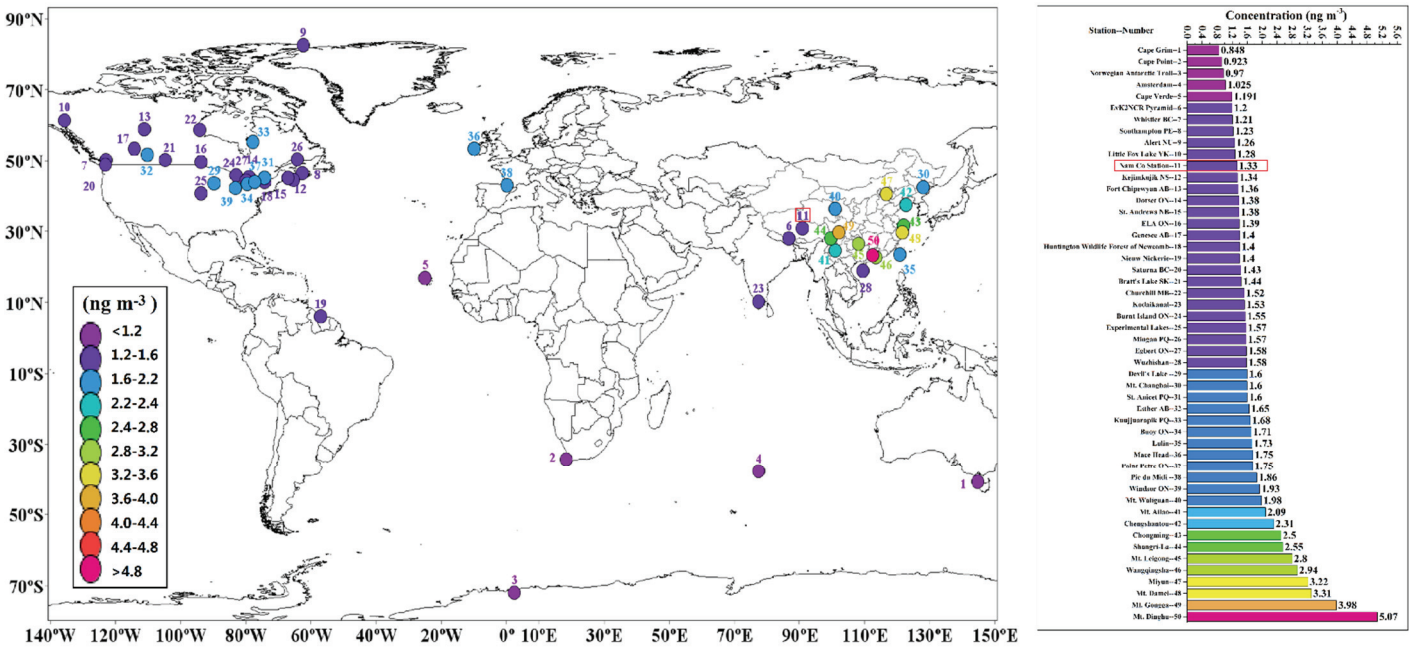

Fig. 1. Geographical location of the remote and rural sites with atmospheric mercury measurements.

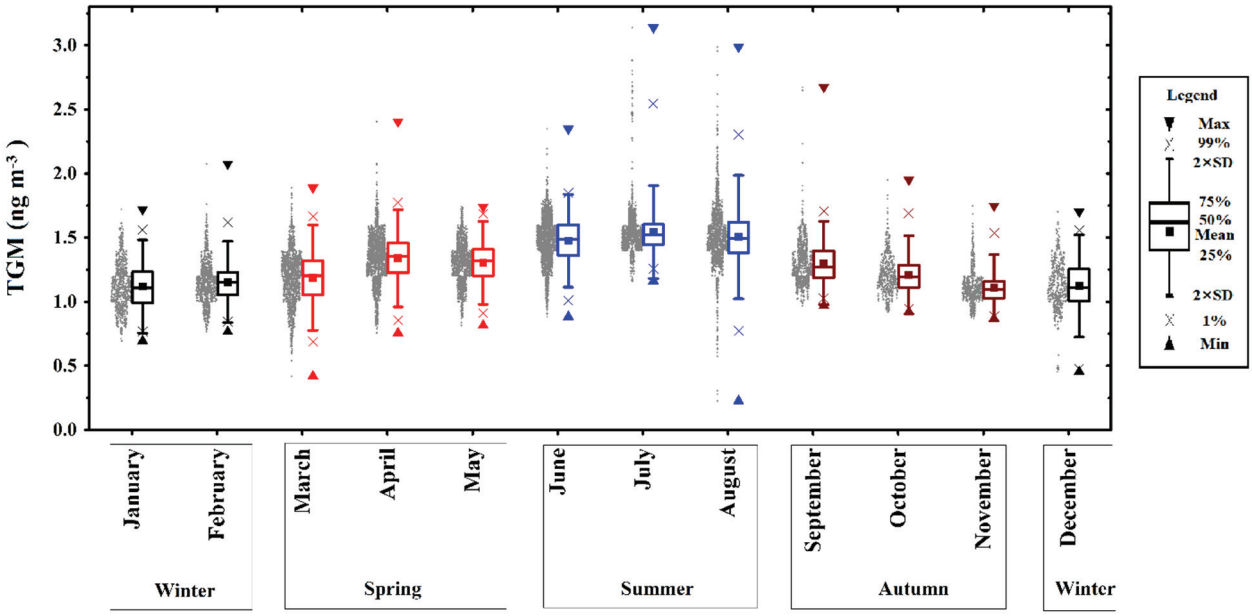

Fig. 2. Monthly average and statistical parameters of TGM at the Nam Co Station during the whole measurement period (spring (MAM) in red; summer (JJA) in blue; autumn (SON) in dark red; winter (DJF) in black).

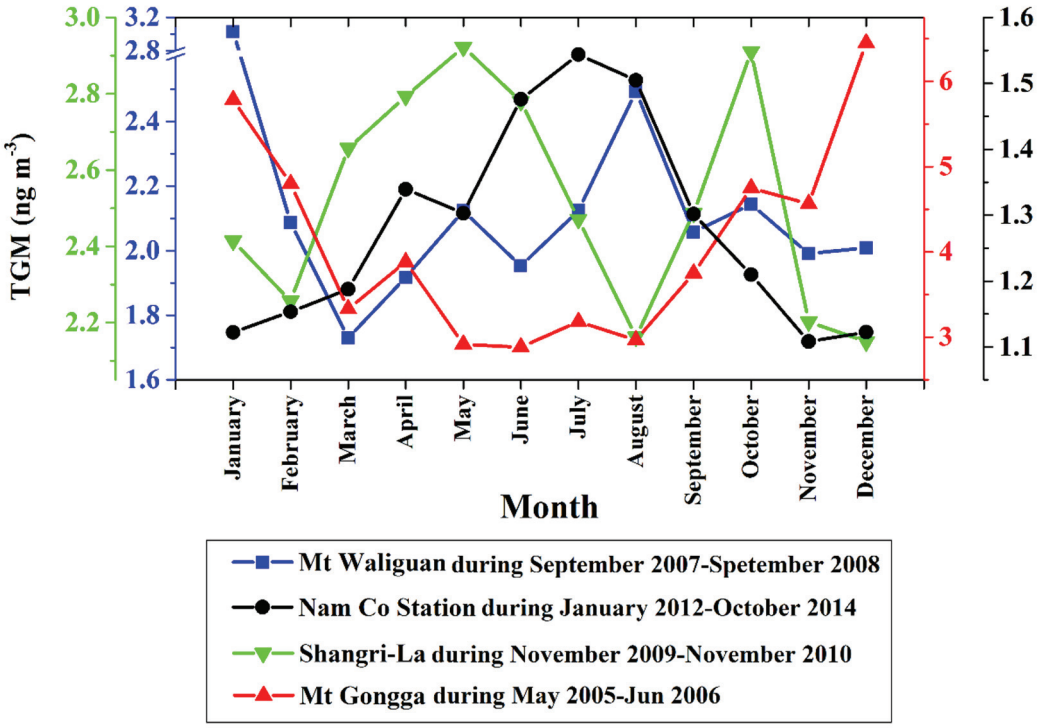

Fig. 3. Variations of monthly mean TGM at four sites (Mt. Waliguan (Fu et al., 2012a), Nam Co, Mt. Gongga (Fu et al., 2008) and Shangri-La (Zhang et al., 2015)) in the Tibetan Plateau.

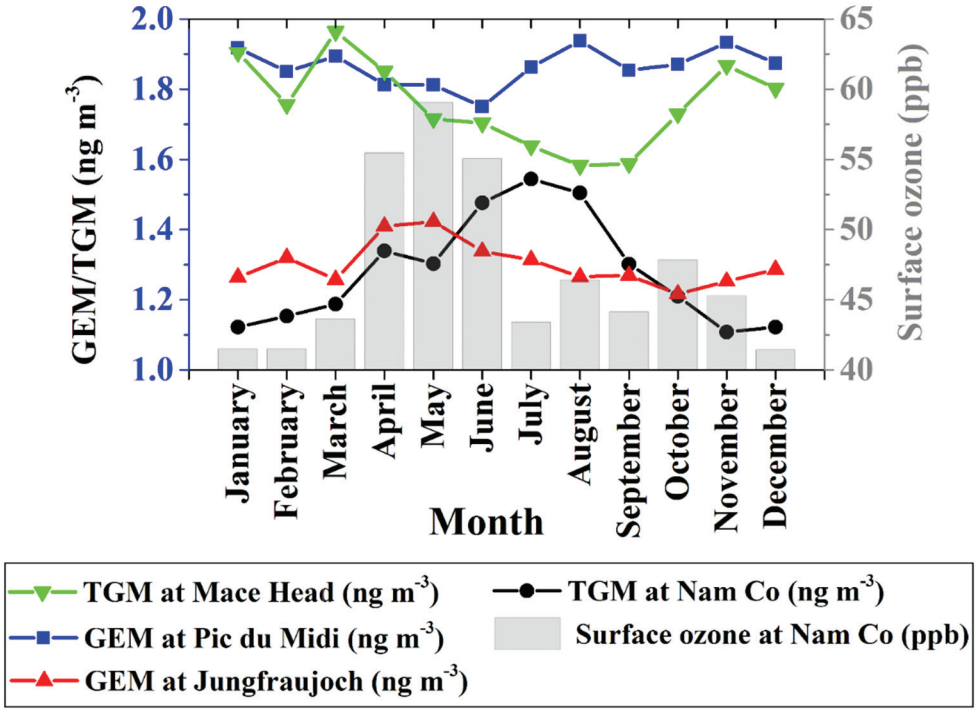

Fig. 4. Monthly average GEM/TGM at the Nam Co Station and three high-altitude background stations in the Northern Hemisphere (Denzler et al., 2017; Fu et al., 2016a; Ebinghaus et al., 2002) (average TGM at Mace Head in green; average GEM at Pic du Midi in blue; median GEM at Jungfraujoch in red; average TGM at the Nam Co Station in black); and monthly average surface ozone at Nam Co in column.

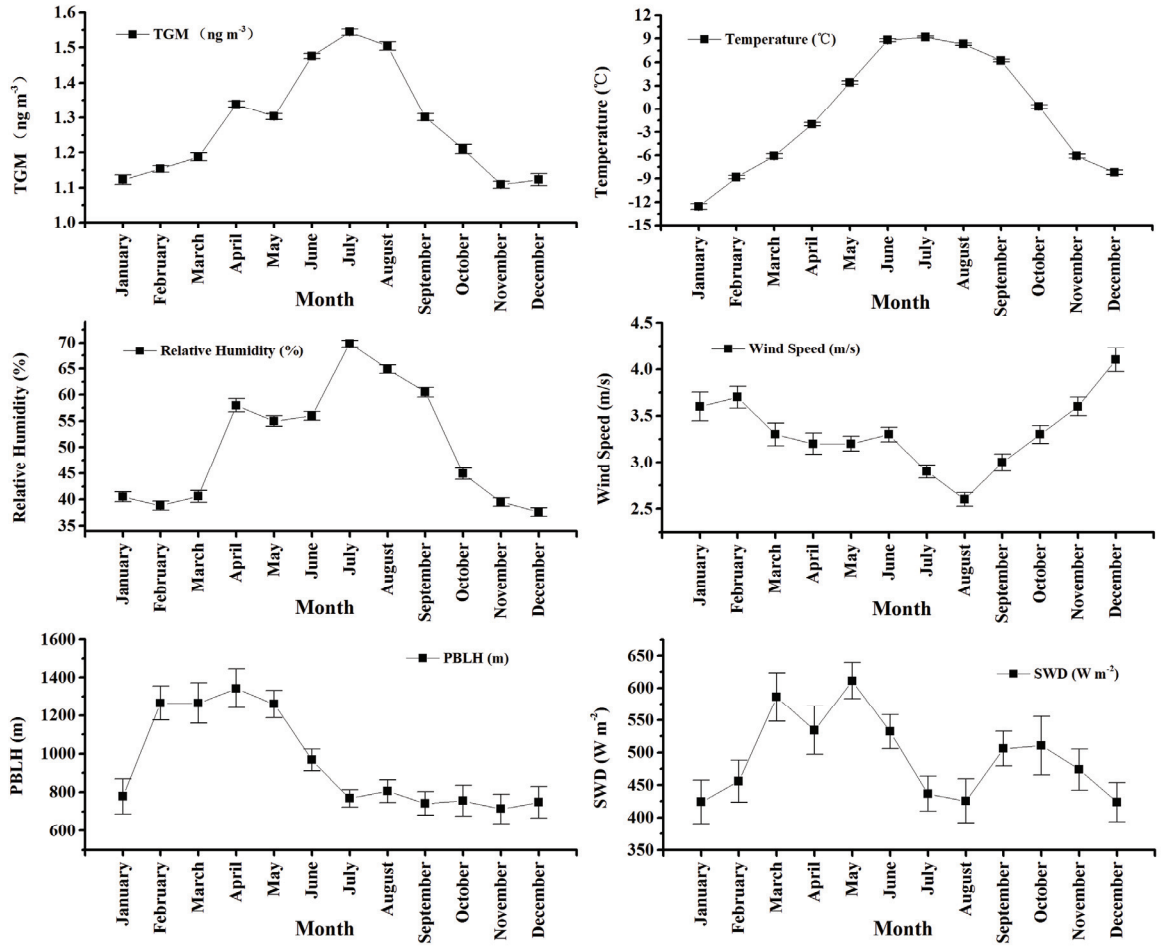

Fig. 5. Monthly variations of TGM, relative humidity, temperature, SWD (downward shortwave radiation), wind speed and PBLH (planetary boundary layer height) during the whole measurement period at the Nam Co Station. Error bars are 95% confidence levels.

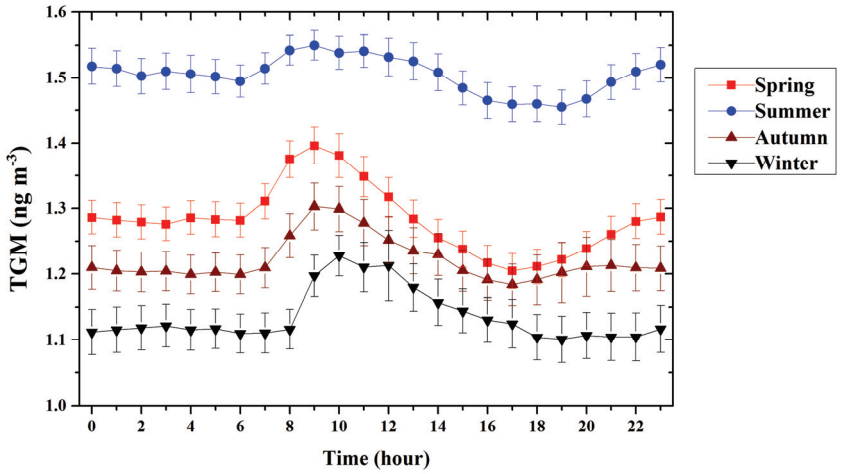

Fig. 6. Diurnal profiles of average hourly TGM at the Nam Co Station by seasons during the measurement period. Error bars are 95% confidence levels.

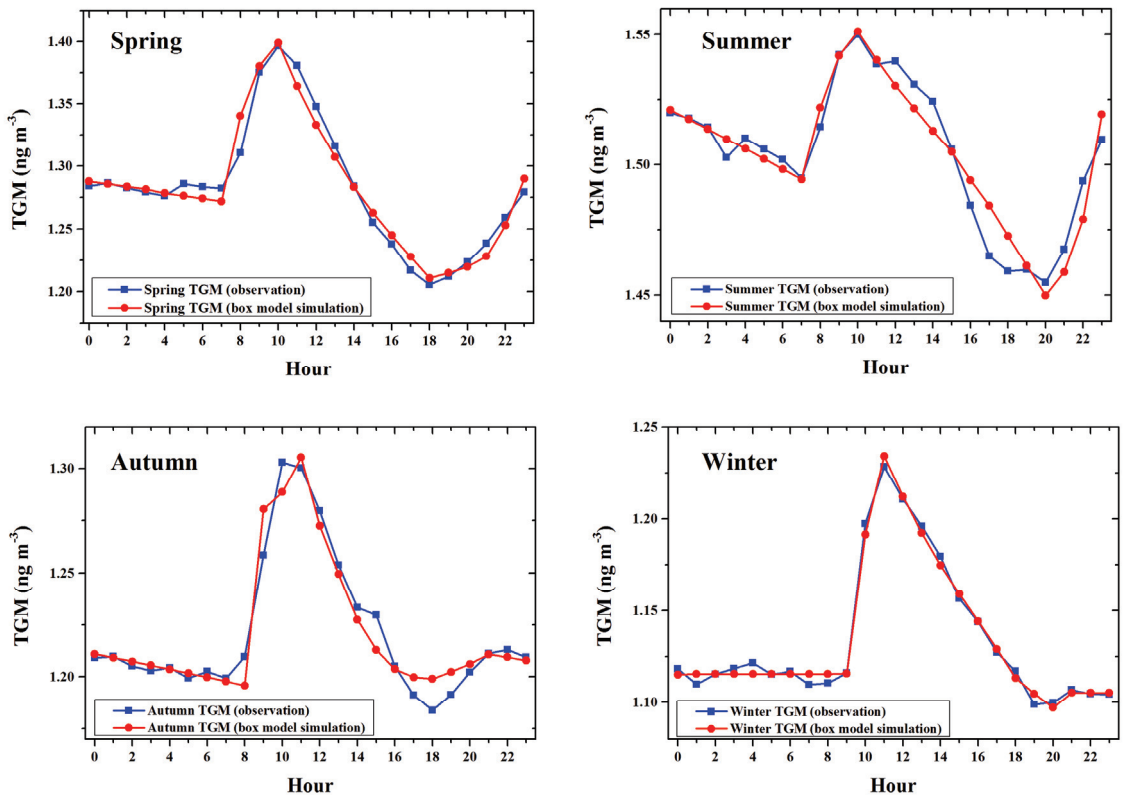

Fig. 7. Diurnal profiles of average hourly TGM at the Nam Co Station by seasons during the measurement period compared with box model simulation.

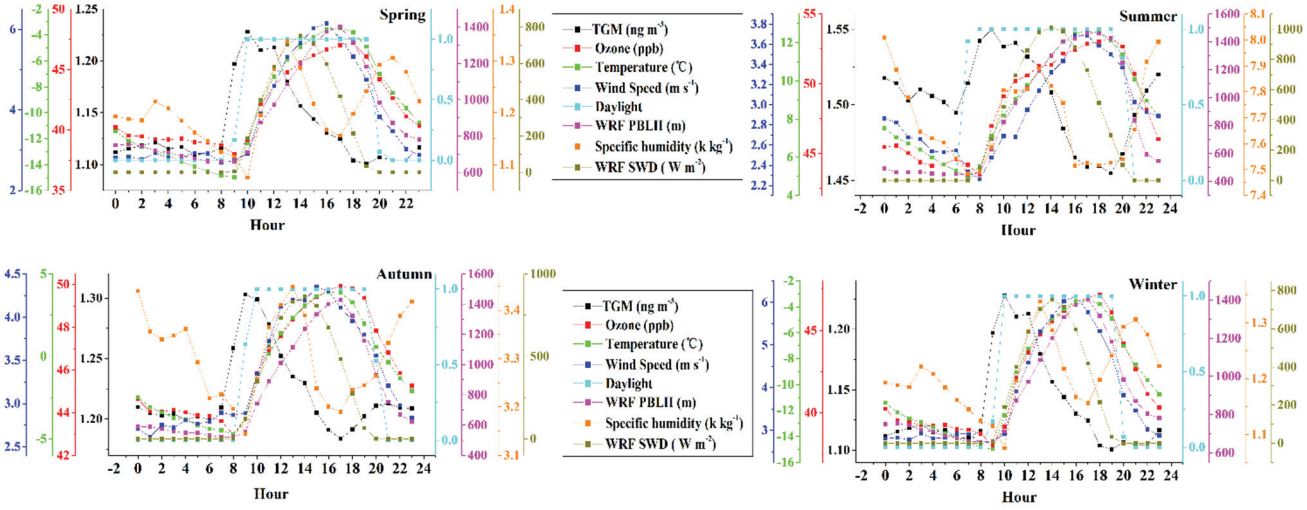

Fig. 8. Diurnal profiles of TGM, ozone and meteorological parameters (temperature, wind speed, daylight, WRF PBLH (planetary boundary layer height), specific humidity and WRF SWD (downward shortwave radiation)) at the Nam Co Station by seasons for the measurement period.

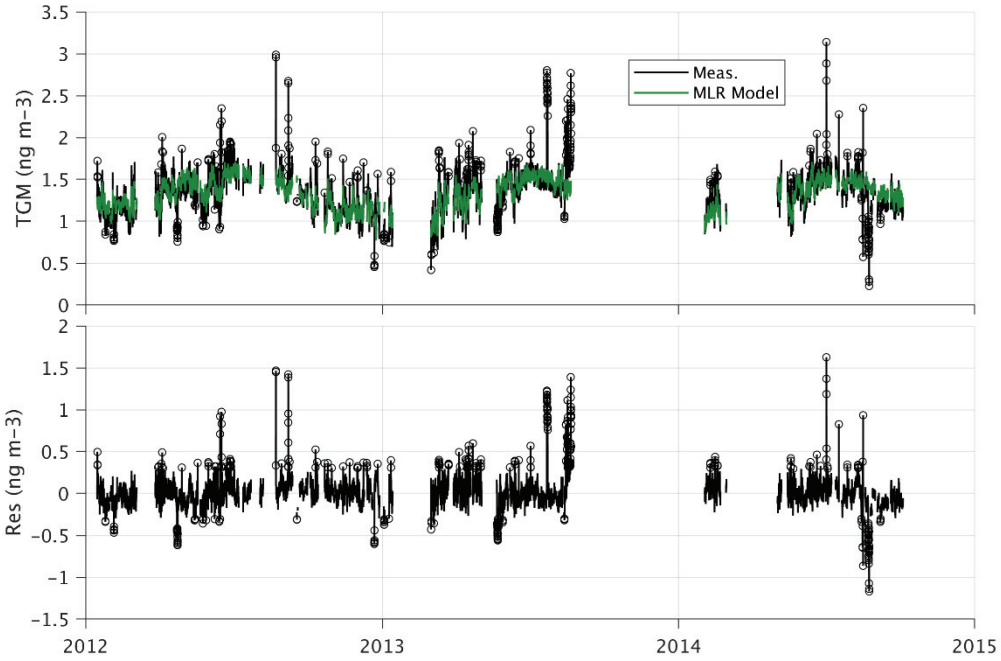

Fig. 9. The measurements and Multi Linear Regression (MLR) model of TGM (top) and model residual (bottom) (residual = measurement – simulation). The outliers are shown as circles.

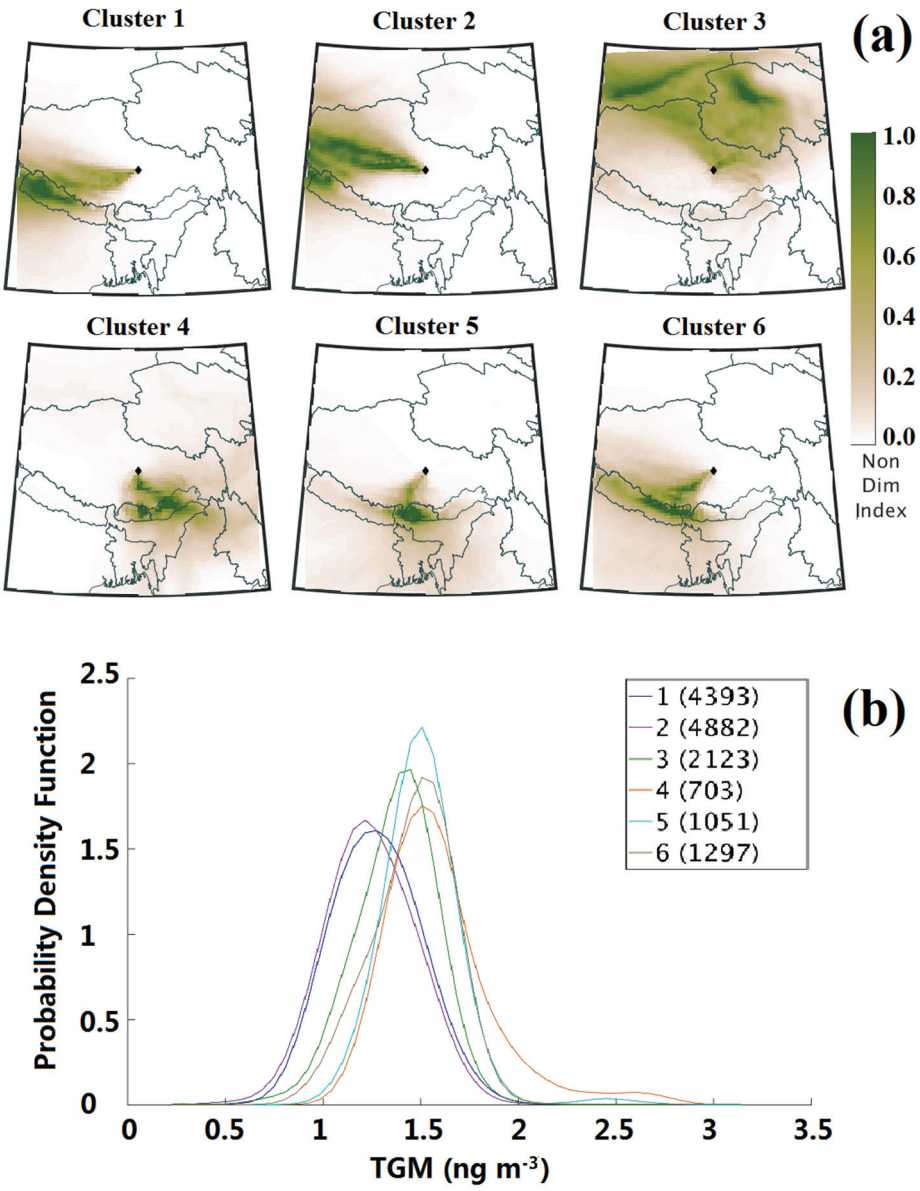

Fig. 10. Clusters of air mass transport to Nam Co using WRF-FLEXPART back-trajectories (a) and probability density function of TGM concentrations for each cluster, with number of data points in each cluster in parentheses (b).

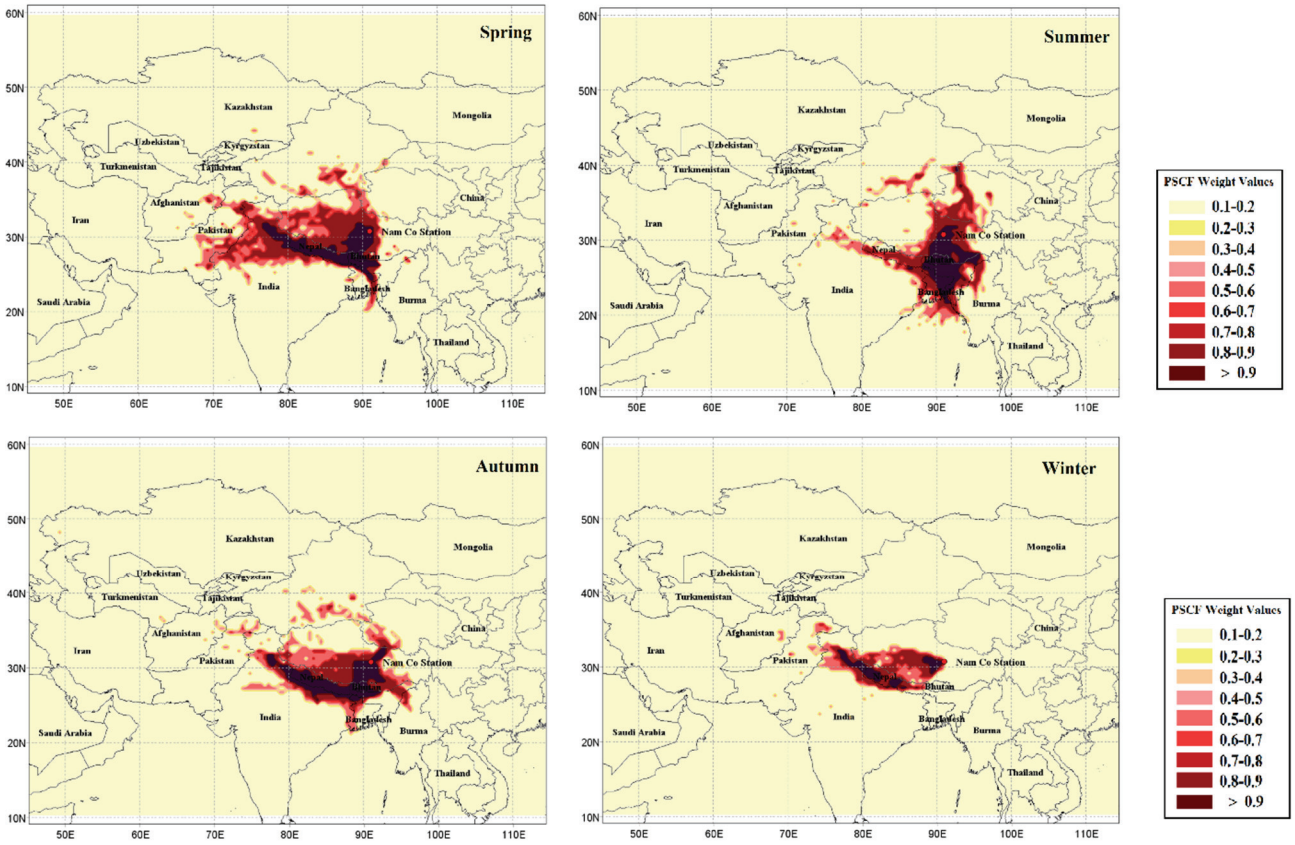

Fig. 11. Potential Source Contribution Function showing areas with possible emissions or air mass transport associated with higher TGM concentrations at the Nam Co Station by seasons in 2012.

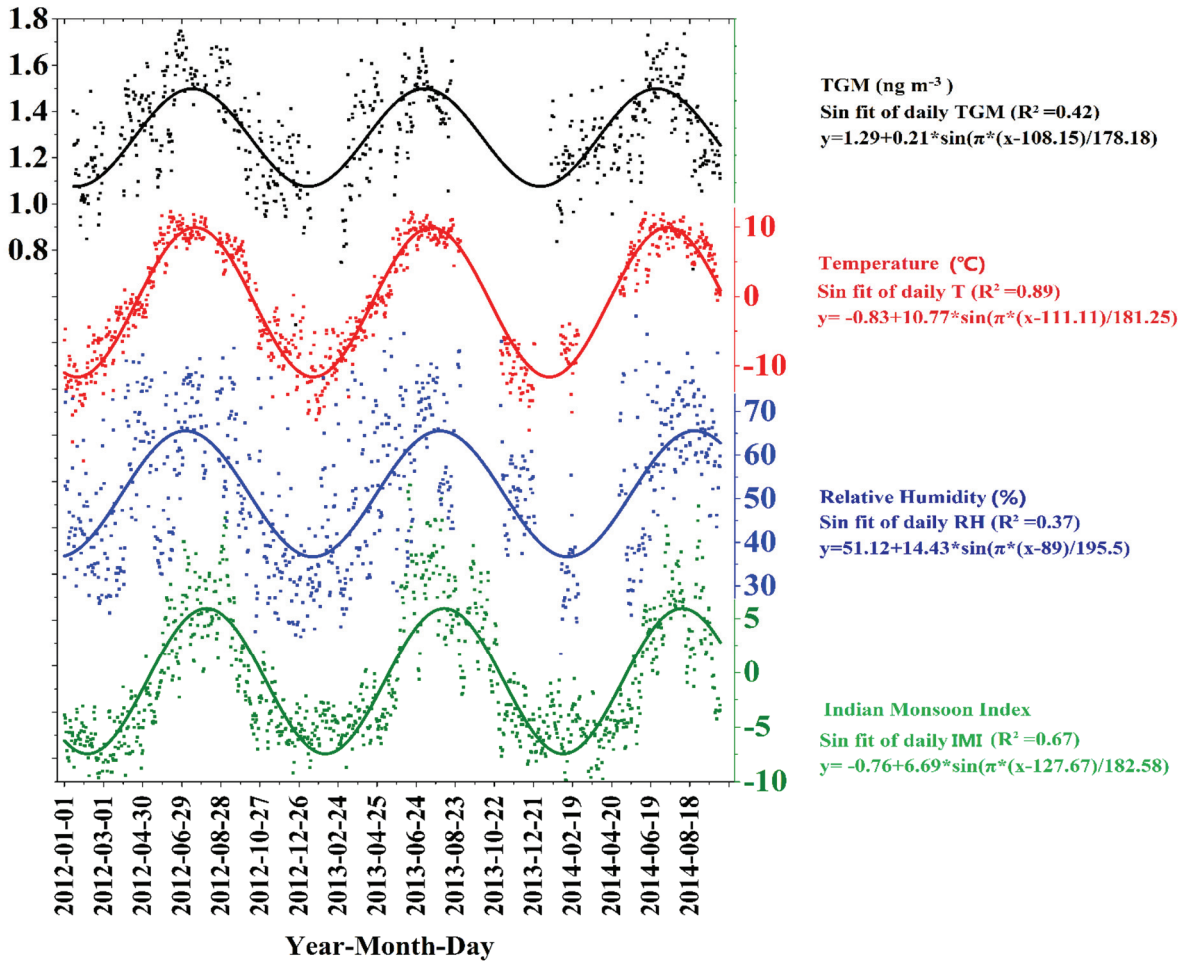

Fig. 12. Series of daily mean TGM, temperature, relative humidity and Indian Monsoon Index and their sinusoidal curve fits.