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

"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). Preliminary tests of the box model were made using solar radiation and temperature to represent chemical transformations, as well as using wind speed and boundary layer height to represent dilution. However these attempts failed to reproduce the diurnal variation found in the measurements. 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."

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

"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."

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

193 6185 data in 2014 (70.6%). Most of the missing data were in the cold seasons. Data in the summer were
194 more complete than during the other seasons and we found that there was weak decrease of TGM in the
195 summer at the Nam Co Station.

196

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

200 **Response:** Thanks for your suggestion. We added the mean ± standard deviation for seasons as you
201 suggested in lines 30-33: "The seasonal variation of TGM was characterized by higher concentrations
202 during warm seasons and lower concentrations during cold seasons, decreasing in the following order:
203 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
204 ng m^{-3})."

205

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

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

214

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

220 **Response:** Thanks for your suggestion.

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

223 Sentences were modified in lines 71-77: "Atmospheric mercury concentrations in Guizhou, one of
224 the most important mercury producing and coal producing regions in China, was reported to be 6.2 - 9.7
225 ng m^{-3} of TGM in the capital city of Guiyang between 2001- 2009 (Feng et al., 2004; Liu et al., 2011; Fu
226 et al., 2011). Measurements of atmospheric mercury at background and remote sites in China include the
227 following sites Wuzhishan (2011-2012), Mt. Changbai (2008-2010), Mt. Waliguan (2007-2008), Mt.
228 Ailao (2011-2012), Shangeri-La (2009-2010) and Mt. Gongga (2005-2006) with concentrations ranging
229 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.,
230 2015; Fu et al., 2008).".

231

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

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

239

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

241 **Response:** Pacyna et al. (2016) stated that "A decrease in emissions in Europe and North America
242 during the time period has been offset by an increase in Asia. The largest increase in emissions is
243 generally due to an increase in coal burning for power and heat generation and for industrial purposes.
244 Increased use of air pollution controls, removing mercury as a co-benefit (and some mercury-specific
245 removing technologies), has slowed down or even reduced the emissions from the increased energy
246 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) 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)"

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

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

Sentence was added in lines 126-128: "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."

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

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)".

Lines 126-127: “A 45-mm diameter Teflon filter was placed in front of the inlet”. How often did 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?

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

407

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

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

415

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

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

419

420 Line 285: "Constant depletion existed in the spring". Use another word than "depletion".

421 **Response:** Thank you for pointing this out and "depletion" was changed to "reduction" in lines 317-
422 318: "Constant reductions existed in the spring, summer and autumn which would correspond to
423 reduction rates of around 1 to 2 ng m^{-2} h^{-1}."

424

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

428 **Response:** In the study at Beiluhe, it was stated that Hg(0) flux showed a diurnal pattern with
429 emission in the daytime and deposition in nighttime, and solar radiation had a great influence on Hg(0)
430 exchange between air and surface. Hg(0) flux started to increase when photosynthetically active radiation
431 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)?

561 Additionally, how many hourly values did you have to calculate the monthly mean in January 2013,
562 August 2013 and October 2014. It looks like you just have missing values.

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

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

566

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

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

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

572

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