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Interactive comment

Interactive comment on "Long-term monitoring of atmospheric TGM at a remote high altitude site (Nam Co, 4730 m a.s.l.) in the inland Tibetan Plateau" *by* Xiufeng Yin et al.

Anonymous Referee #3

Received and published: 17 April 2018

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:

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)

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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 . . .?

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.

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) air-surface 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.

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.

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.

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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).

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.

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

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.

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.

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

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

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 ïĆś standard deviation for both seasons. Is the difference between mean concentrations significantly different?

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.

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.

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/en-US/Default.aspx

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

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

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

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.

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.

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.

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

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”.

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

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

Line 158: Please define MLR.

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

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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.

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.

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).

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?

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.

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

Line 259: “Accompanied by lower surface ozone concentration”. Can you add ozone concentrations in Figure 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.

Section 3.3. Diurnal variations of TGM: I don’t really see the point of the box model.

See main comment #3.

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

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?

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

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

Line 321: “The middle panel”. There is no middle panel.

Line 324: “with very low TGM concentrations”. Please define “very low”.

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

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.

Line 365: Replace “old mercury” by “legacy mercury”.

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.

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

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.

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

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?

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.

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 (Jan 2012-Oct 2014)). See main comment #7.

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.

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.

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.

Figure 10: Please remove the line for missing data.

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

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.

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?

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