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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The variability in Gaseous Elemental Mercury at Villum Research Station, Station Nord in North Greenland from 1999 to 2017" *by* Henrik Skov et al.

Anonymous Referee #2

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This manuscript by Skov et al. presents a time-series of gaseous elemental mercury (GEM) concentrations at Villum Research Station (Station Nord), Greenland from 1999 to 2002 and 2009 to 2017. Alert (Canada), Station Nord, and Ny-Ålesund (Spitzbergen) are presently the only three long-term monitoring stations for GEM in the Arctic. As such, the data presented here are extremely valuable and I would like to acknowledge the authors for their work and dedication. That being said, I do not find the interpretation of the data convincing, mostly due to a confusing Material & Methods section (see suggestions below). The Results and Discussion section is also difficult to follow; I wasn't always sure whether the authors were referring to the annual or seasonal trend. Reorganizing the discussion per season could help. Finally, I think there is a lack of

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Discussion paper

sufficient references, especially recent ones. Below are some detailed comments and suggestions that will hopefully help the authors to improve their manuscript.

Measurements section

1. Line 89: “several generations of the instrument have been used (A, B, and X version)”. Could you add somewhere (in the text and/or on Figure 3) the dates at which the Tekran instrument was changed? Given the 20% intercomparison uncertainty between two instruments (Slemr et al., 2015), that should I think be taken into account when performing a trend analysis. The winter trend seems driven by the high value in 2000 and the low value in 2017. Does it coincide with a different instrument being used? According to Angot et al. (2016), you used a Tekran 2537A at least from 2011 to 2015. According to Kamp et al. (2018), you used a Tekran 2537X in spring 2016. When did you switch? Did you measure GEM concentrations with the two instruments for a certain period of time in order to evaluate the intercomparison uncertainty? The lack of information casts doubts on the trend analysis. GEM trend analysis is of utmost importance for the effectiveness evaluation of the Minamata Convention. However, potential implication of the use of multiple instruments for GEM trend analysis is somewhat overlooked by the community. A discussion on the matter could strengthen the conclusions of the manuscript.

2. What is the time resolution of the GEM measurements? 5 or 15 minutes? Did you use the 5/15 min data for the trend analysis or hourly means/medians, or annual averages?

Trend analysis

How did you perform the trend analysis? Please describe the method in the Material and Methods Section. It seems that you are simply using the regression line. It is of common practice to use the Sen’s slope and Mann-Kendall test for trend analysis (e.g., Berg et al., 2013; Cole and Steffen, 2010; Martin et al., 2017). Again, the lack of information casts doubts on the trend analysis.

Modeling section

1. Can you please provide more information regarding the Hg chemistry in the model (e.g., main oxidation and reduction pathways)?

2. Lines 126-130: I do not understand which Hg emissions were used in the simulations. You mention using “global historical AMAP Hg emissions inventories 1999 to 2017” which seems to suggest different emissions every year from 1999 to 2017. You later say that “the anthropogenic emissions are variable up to 2010 where after they are constant”. Please clarify: Does this mean you used variable emissions from 1999 to 2010, then constant emissions (equal to 2010 emissions?) for 2010-2017? Please also clarify what is the reason for doing so. Are you trying to investigate the influence of changing anthropogenic emissions on GEM concentrations at Station Nord? I would like to see a Table summarizing which simulations were done in order to address which question/hypothesis, and a list of sensitivity simulations. See for instance Table 2 in Giang et al. (2018) or Table 2 in Travnikov et al. (2017).

3. Line 133: Can you please clarify what the “prescribed boundary conditions on 1.5 ng/m3” is? How did you come up with this value of 1.5 ng/m3? Is it based on a run with a global model? Or is it what you consider the Northern Hemisphere background concentration? If so, where does this value come from? I also do not understand why you refer to this value as a hemispheric background while it also represents “transport from sources in the southern hemisphere”. This is confusing. In addition, can you please perform a sensitivity analysis here? Based on your simulations, direct anthropogenic transport only accounts for 14-17% of GEM. Does this direct anthropogenic influence changes if you decrease the prescribed boundary condition?

Line-by-line comments

Lines 42-43: “natural, anthropogenic, and reemission, accounting for roughly 10, 30, and 60% of the emissions, respectively”. The authors should cite Amos et al. (2013) here.

Lines 43-51: Please discuss the most recent estimate prepared for the Global Mercury Assessment 2018 by Outridge et al. (2018). Anthropogenic emissions are ∼2500 tons per year, natural and secondary emissions ∼5500 tons per year, and evasion from ocean accounts for ∼60% of the sum of natural and secondary emissions.

Line 59: "evidence has been obtained from experimental and theoretical studies for a much shorter lifetime of GEM". What do you mean by "much shorter"? Please provide a range of values. Additionally, please considering adding Horowitz et al. (2017) to the list of references here.

Line 68-69: "Today China accounts for about 40% of the global Hg emission (Jiskra et al., 2018; Muntean et al., 2014)". Citing Jiskra et al. (2018) is not appropriate here. I suggest the following papers instead: Streets et al. (2019, 2018, 2017).

Line 70: "decline in the GEM concentration of between -1.5 and 2.2% per year (Obrist et al., 2018)". Citing Obrist et al. (2018) is not appropriate here. Please consider citing Zhang et al. (2016) instead.

Line 84: "2015 when the measurements were moved to the newly build Air Observatory". Figure 2 says 2014 (caption).

Lines 93-95: "the reproducibility for concentrations above 0.5 ng/m3 is within 20% based on parallel measurements with two Tekran 2537A mercury analyzers". Is it something you did as part of this study or are you referring to another study? If so, the reference is missing. You could cite Slemr et al. (2015) here.

Lines 154-156: "A seasonal pattern is observed for each year. In January and February, the level of ozone and GEM is rather stable. After the polar sunrise, the concentration starts to fluctuate strongly". It is pretty hard to see anything on Figure 3. Could you add a Figure giving the mean seasonal cycle of GEM (e.g. Figure 4 in Angot et al. (2016))?

Line 163: "A test of the importance of the value in 2000 . . .". How about the value in

2017?

Line 169: typo. Dommergue.

Lines 192-194: the end of that sentence is missing.

Lines 202-204: “Frequency of AMDE and GEM concentration in summer showed a poor negative correlation. If the deposited Hg during AMDE should be released again during snowmelt, a positive correlation would have been expected, but this was not observed”. This hypothesis has already been tested, and the same conclusion reached, by Angot et al. (2016) based on data from Alert. “The increase of Hg(0) concentrations in summer could be due to the reemission of Hg deposited during springtime AMDEs. However, the comparison of the magnitude of the summer enhancement at Alert suggests otherwise”. This is worth mentioning since the same conclusion is reached here.

Line 208: “The present study indicated that atmospheric input can be significant as well”. I’m not sure I followed how you arrived at this conclusion.

Line 223: “The highest concentration of GEM was found in 2013”. Isn’t it in 2014 according to Figure 4?

Lines 225-226: “The DEHM model, using variable anthropogenic emissions as described above, shows a slightly decreasing concentration trend (see Figure 7)”. I assume you mean Figure 8 here. Figure 7 shows the contribution of the various regions to the annual average GEM concentration at Villum. Also, you mention “variable anthropogenic emissions” while using constant emissions after 2010 according to the Material and Methods Section. If you observed a decreasing trend with constant emissions, that means the decreasing trend is not due to decreasing anthropogenic emissions. This modeling section is quite confusing, reason why I suggest major changes in the Material and Methods section to clarify what you are doing.

Line 234: missing parenthesis after Dibble et al., 2012.

Line 242: “in separate calculations with DEHM”. What exactly did you do? Please

improve the Material and Methods section accordingly and provide a summary of all the simulations performed (and why).

Lines 242-257: Chen et al. (2018) also found that Hg deposition in the Arctic is mainly due to emissions from Asia. Additionally, the source-apportionment analysis performed with four global models for the Global Mercury Assessment 2018 shows that the Arctic is predominantly influenced by long-range transport from East Asia.

Lines 259-264: How do you results compare to the study by Dastoor et al. (2015): the authors found that changes in meteorology and decline in emissions in North America and Europe contribute equally to the decrease in surface air Hg(0) concentrations. Additionally, you mention here a simulation where emissions are kept constant at the 2005 level, while meteorology is varying. Please clarify this in the list of simulations in the Material and Methods.

Lines 265-278: this entire paragraph is unclear because Table 2 is missing.

Line 278: “The DEHM model predicts that there is a maximum in GEM. . .”. Could you please provide a direct comparison of observed vs. modeled time-series?

Lines 279-281: “In fact, the highest concentration of GEM is observed during summer and is attributed to release of GEM from the melting snow and ice pack from Hg deposited during AMDEs in spring”. I do not understand. You say earlier in the manuscript that springtime AMDEs cannot explain the summertime enhancement: “Frequency of AMDE and GEM concentration in summer showed a poor negative correlation. If the deposited Hg during AMDE should be released again during snowmelt, a positive correlation would have been expected, but this was not observed”.

Line 288: “Simulations of the concentrations at Villum using the DEHM model using fixed emission inventory show no significant trends”. You say earlier that the simulations give a decreasing trend: “shows a slight decreasing concentration trend, -0.7% per year”. Or, by "slight", do you mean that this trend is not significant?

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