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Comment on acp-2020-1287

Anonymous Referee #1

Referee comment on "Dynamics of gaseous oxidized mercury at Villum Research Station during the High Arctic summer" by Jakob Boyd Pernov et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-1287-RC2>, 2021

Review of Dynamics of gaseous oxidized mercury at Villum Research Station during the High Arctic summer by Jakob Boyd Pernow et al.

This manuscript presents original data regarding atmospheric Hg species during summertime in the high Arctic. The authors suggest that the GOM peaks (so called "events") that are measured are explained by the influence of free tropospheric production of GOM and transport to the sea-level site.

The scientific reasoning that is conducted here is presented in backwards order. The authors conclude on the influence of air masses from the free troposphere on GOM measurements. To my opinion, the supplementary data are not robust enough to support this very original but debatable hypothesis. Then the authors examine additional data to further explore the GOM origin including other GOM sources. From the data they show, it appears to me that the hypothesis of alternative GOM sources remain still valid while the authors clearly reject these sources since they do not support their initial idea. I think this is a biased way of discussing their data set, and that their initial hypothesis is not supported by clear evidence and data.

Demonstrating the direct influence of the free troposphere at an ocean-front site requires solid multiparameter measurements, on-site knowledge of the vertical structure of the atmosphere and probably vertical concentration profiles (aircraft). Although the hypothesis is attractive, there are many other possibilities to explain these peaks of GOM, which have been only partially studied in this manuscript, including local pollution sources (ships, airplanes), biomass fires, anthropogenic influence of European pollution, and transport of GOM species from the Greenland ice cap. The backtrajectory analysis is not deep enough and the statistics are not convincing.

Moreover, the introductory part contains too many approximations and requires an

obvious reformulation work.

For these reasons, this work cannot be published in ACP . It could at least be requalified as a "measurement report" with however an important work on the formulation of the different hypotheses.

Details comments.

Line 9 : « GOM, once introduced into the ecosystem, »- GOM are not really introduced in ecosystems, these species are deposited. The link with « threat to human and wildlife » is exaggerated since there is no proven direct link between GOM and wildlife contamination. This need to be rephrased

Line 11 : « the ecosystem » is not appropriate.

Line 21 « an »

Line 21 : what is « relaxation time » ?

Line 24 : artisanal small scale gold mining

Line 28 : atmospheric particles or aerosol, not a combination of both

Line 31. A reference is missing for « depletion events », at least you should cite Schroeder et al 1998.

Line 31 PHg is not a fraction of total gaseous mercury.

Line 32. The link with the previous sentence is not straightforward. « in contrast » is not appropriate here.

Line 34 – reactive halogens would be better. And especially bromine radicals. There is no real evidence for other halogens reactivity. Why coastal regions? There are many coastal sites where no reactivity is observed (e.g Mace Head in Ireland).

Line 38-39 : This statement is only valid for Alert in Canada. Has it been observed elsewhere?

Line 40 : the peak of Hg in snow – This should be clarified. What kind of peak ? is it totalHg in surface snow ? « GOM is the main deposition pathway » does not mean anything.

Line 60 : Do Ariya et al really mention a ionic pulse ? there are better references for this .

Line 61. Are you sure that GEM can be directly methylated ? PHG ? Methylation does not occur at the « earth's surface, this should be better explained.

Line 65. I do not understand why « it is pertinent to understand mercury oxidation in response to a changing climate » . There is no link with the preceding sentences.

Line 71 : GOM deposition does exist outside of AMDE since this is a major removal pathway for Hg on a global scale

Line 82-83 : I do not agree with this conclusion. Most of the studies report low GOM/PHG values.

Line 84-86 : this sentence is difficult to understand.

Line 87 – I don't understand what « will help to infer the response of mercury in the context of a changing climate » mean

Line 88 : It does not make sense "is also important to understand the dynamics of mercury to assess the effects of abatement strategies on atmospheric concentrations in the framework of the Minamata Convention (UNEP, 2013) »

What is the link between abatement strategy and Hg dynamic in the Arctic ?

Line 125. You should be consistent with the numbers.

Line 128 : How is snow depth measured ?

Line 129 : What does « averaged to 30 - minute means » mean?

GOM data

Is the use of the GOM detection limit appropriate? Why not using the Quantification limit since we all know that these speciation instruments are quite difficult to manage? Event 2 is very closed to the detection limit and the first days of Event 1 and may be excluded.

There is no discussion on the quality of GOM data obtained with denuders while the authors may know that Tekran speciation unit underestimate GOM value as shown in recent studies (Maruszczak et al 2017 – Gustin et al 2015 – Huang et al 2015, 2017)

Are the raw data available for all these events ? This is critical to make sure that all the blanks where correctly made.

Line 204 : yes but there is no evidence that there are open leads on the way ?

Line 218 : snow cover is not displayed in figure S2

By the way the color scale for the contour is quite difficult to read.

Line 230 : What is considered as high radiation and low RH ? How are those threshold defined ? Event 1 had some low radiation too (<200). Was there a snow/rain fall ? on august 21st ?

What is the dynamic of the boundary layer ? This is an important factor that can explain some variation in your concentrations.

Line 230 : your suggestion that cold temperatures are associated to mercury oxidation is not valid in volcanic plumes or in salt lake regions. To me, the temperatures are not a solid argument to reject in situ production. Halogen measurements could give a major evidence to demonstrate this -although I do not believe that this is likely to happen at this time of the year.

Line 249-253 : How are calculated those averages ? on the whole trajectory duration ?

Given the uncertainties of all these measurements and of the average, are these air masses statistically different ? Given the shown interval, I do not see any evidence of a robust difference. The Wilcoxon test is used for comparing independant populations. Here you have an overlap on « event » and « non-event » trajectorie : for example the 28/08 -120h trajectories overlap and are supposed to be included with the one from the 26/08.

Where the test conducted on each separate observation? (ie RH in « event » with RH in « non event »). Btw, water mixing ratio if a function of temperature and RH ?

Line 276 : The meaning of this sentence is not clear. What does « these air masses » refer to ?

Line 280 : Figure 3d and 4d With this figure the author suggest that GOM event air masses spent more time at high altitude. The overall picture is not that clear. Event 3 and 4 are not very different from the non-event period (27/07-31/07). Regarding the strongest event in 2020 (on the 23/07) these air masses are not different from the day before and the altitude is close to what one can expect as a marine boundary layer. In summer time it can be several hundreds of meters thick, even more when passing over turbulent and convective areas.

Line 282 : for the 2019 campaign : what is the value show with the median height ?

1sigma ? min-max ? For this campaign, there are important overlap between « event » and « non event » periods.

Line 285 : how is retrieved the mixed layer height ? How robust is it in Hysplit ? Why is it no plotted on your figures ?

Overall with the presented data, I do not come to the same conclusion (line 286-289).

Line 295 : you mentioned earlier than cold temperature below -15°C are likely needed.

Looking at temperature along the BT ways, it is not the case here ?

In the free troposphere what could cause the formation of GOM is mainly the supposed abundance of Br concentrations ?

Line 298 what is the low surface resistance ?

Line 299 There are two verbs in the sentence.

Line 301 : survival is not appropriate for a chemical species

Line 31 – 5 days backtrajectories are clearly not long enough. For BC studies or aerosols 10 days are usually used (see Thomas et al 2017 in GRL). The life time of GOM is poorly known and could be of several days to weeks in dry conditions?

This approach is not relevant. CO data and BC may be a more straightforward approach to track fires. This whole paragraph does not bring any relevant information and fires may contribute to these GOM events, and/or GEM.

Line 325 : this first sentence has no meaning in atmospheric chemistry.

The Shah and Jaeglé study point sub-tropical areas and this is an important difference from mid-latitudes.

Line 340 : Why not using aerosols (check Uge et al in GRL 2017). The paragraph 3.3 is only a short review of FT measurements and is of no interest for the discussion.

Line 390 – and after. The authors make a confusion between the influence of free troposphere and a stratosphere intrusion. Stratospheric intrusion would bring very dry air masses and very high ozone. It is likely that the Biomass burning may have an influence on ozone.

There is not ozone data presented in Jacob et al 2010. I don't understand what do the authors find in Monks et al 2015 to support their statements. Monks et al suggest that European anthropogenic emissions may be important for lower tropospheric summertime ozone and that PAN reactivity may be a source of ozone.

Paragraph 435 – 444 . The correlation of GOM and BC looks very interesting in 2019 so I do not understand why the conclusion is that combustion sources has no influence – As said earlier the 5 days trajectories are too short to reach this conclusion. For year 2020, event 3 and 4 may be as well related to combustion sources (the scale is different on figure 7a and b). Event 5 could be due to production of GOM over the Greenland ice cap (as mentioned in Brooks et al 2011 – although earlier in the season), or as proposed in Angot et al 2016 10.5194/acp-16-8265-201. This hypothesis would also lead to low BC.

Then I do not agree that this airmasses comes from the upper troposphere. The GOM peak on August 1st show trajectories around 1000-2000 m ? This could only be air masses leaching the Greenland icecap before arriving to VRS.

453-454 – This is not because a high BC is observed without GOM for a single event that biomass burning cannot influence arctic GOM concentration. This is too speculative.

508-511 – This is a very vague speculation.

