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# Comment on acp-2020-1287

Anonymous Referee #4

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-RC3, 2021

The authors present data from two summer measurement campaigns at a high Arctic station. The presentation and discussion of the data is focused on the origin of the gaseous oxidized mercury (GOM). They arrive at the conclusion that high GOM concentrations during all 5 events are due to the transport from the free troposphere. At a very remote site this is almost always the case and, as such, too unspecific. This is a pity because the observed events display different patterns which would enable more specific conclusions.

The data are valuable and worth of detailed analysis. Unfortunately, the manuscript is difficult to read because GEM, GOM, PHg, $O_3$, BC, $H_2O$, and $N_{coarse}$ are all discussed in different chapters. A change of perspective in $O_3$ chapter from the analysis of individual events to the whole campaign analysis does not make the understanding easier. Ideally, the air mass of each event would be characterised by all of the available data, including backward trajectories (the consideration of the latter would probably result in splitting the event 1 into two events). This would be followed by detailed discussion of GOM origin in each of the characterised events separately before making final conclusions.

I recommend the publication of the manuscript after a thorough reorganisation of the text along the lines proposed above. Some of the comments below are also to be considered:

Lines 12-13: „we performed measurements of ……air mass history”?

Line 24: Biomass burning is partly natural and partly man-made.

Lines 31-33: Wording: During depletion events… GOM and PHg can constitute large

fractions of… In contrast,….. GOM and PHg can constitute large fractions of. ??

Line 41: "oxidation" instead of "oxidization"

Line 51: What does it mean "liable halogen reservoir species"?

Lines 56-57: Uptake by vegetation and soil in the high Arctic? Is there enough vegetation and soil not covered with snow for that in the high Arctic?

Line 65: "important" instead of "pertinent"?

Line 80: CA stands usually for California, not Canada.

Section 2.2: Duration of GOM and PHg sampling time should be given. This is important because the usual 2h sampling at 10 L/min usually provides too small Hg amounts for analysis unbiased by the internal Tekran signal integration routine (Slemr et al., Atmos. Meas. Tech., 9, 2291-2302, 2016; Ambrose, Atmos. Meas. Tech., 10, 5063-5073, 2017). This artefact applies also for 5 min GEM sampling time which probably leads to a small underreporting of the GEM concentrations.

Line 181: RH at a given $H_2O$ content of air is inversely related to temperature and as such provides a redundant information on temperature (as can be clearly seen in Figures 1 and 2). Consequently, it is not a suitable variable for the characterisation of the air mass. $H_2O$ content of air, as shown in Figure S5 and discussed elsewhere in the manuscript, should be used mostly through the paper. The use of RH makes sense only when discussing the GOM attachment to particles.

Section 3.1: Figures S1 and S3 show that local meteorological parameters essentially do not matter. What matters are the times of the air mass exchanges characterized more clearly by their specific chemical fingerprints and the question where they come from. As already mentioned, RH should be replaced by $H_2O$ content of the air.

Figures 1 and 2: I think that these figures should include all measured parameters, i.e. additionally $O_3$, BC and $N_{coarse}$ . Measurements of BC and $N_{coarse}$ shown in Figure 7 are especially important because these species are specific tracers for anthropogenic activities and biomass burning. GOM and PHg below detection limit are plotted as zero concentration which is misleading because "below detection limit" does not mean zero. As mentioned above, their concentrations are underreported due to the internal Tekran

integration procedure. I would plot only the measured GOM and PHg concentrations above the detection limit.

Figure 3: RH in panel b provides hardly any additional information to T in panel a. Water content could be more useful because it would reveal the precipitation along the trajectory as discussed in the related text. A comparison of panel a (T) with panel d (altitude) shows an inverse relationship which is not mentioned in the text.

Figures 1, 5a and S2 show that event 1 consists essentially of two events with different trajectories which should be perhaps treated separately.

Figure 5: The addition of trajectory altitudes, as in Figure S4, could provide support for the claim of GOM arriving from the free troposphere.

Line 325: "Influence of the troposphere on mercury concentrations" sounds like "water has influence on fish". Please reword.

Lines 328-332: Measurements of GEM by Talbot et al. (2007) are subject to two experimental artefacts: a) with their specific inlet system they measured most likely GEM + GOM, not only GEM as they claim, and b) their reported concentrations are too low because they relied on internal Tekran signal integration procedure which was demonstrated to underreport Hg concentrations leading to numerous zero concentrations in the paper which are incorrect. With these problems, Talbot et al. does not provide any usable information about GOM. A reference to Lyman and Jaffe (Nature Geosci., 5, 114-117, 2012) and Slemr et al. (Atmos. Meas. Tech., 9, 2291-2302, 2016; Atmos. Chem. Phys., 18, 12329-12343, 2018) would be more appropriate.

Line 355: Do you mean $O_3$ median at Villum station?

Section 3.4: Elevated $O_3$ mixing ratios could be due to a transport from free troposphere but also due to $O_3$ formation in polluted air masses. The latter applies clearly due to anthropogenic pollution during the events 1 and 3 and to a smaller degree to event 2 as indicated by BC and $N_{coarse}$. A discussion of the elevated GOM concentrations during these events in terms of anthropogenic pollution would be more appropriate.

Figure 6: Putting all data for 2019 and 2020 campaigns into each GEM vs $O_3$ and GOM vs $O_3$ diagrams is inconsistent with the discussion of individual events 1-5 in other chapters and obscures the origin of GEM instead of revealing it. What would be the GEM vs $O_3$ and GOM vs $O_3$ correlations for each of the individual events 1 – 5?

Section 3.5: This discussion is muddled. GOM correlates well with BC and $N_{coarse}$ during the events 1, 2, and 3 but not during the events 4 and 5. Despite the different patterns, the authors conclude that emissions from biomass burning and combustion have little to no influence on GOM levels at Villum station. That may be true for events 4 and 5 but obviously not for events 1, 2, and 3. The discussion in lines 445 – 454 may be used as an argument against biomass burning being the source of GOM in some of the events but not in all.

Section 3.6: This section is highly speculative and confusing. As in the discussion of GOM vs $O_3$ the different pattern of each of the five events is not mentioned. Especially that of event 5 with no relation between GOM and $N_{coarse}$ and BC, whatsoever. A weak correlation with of GOM with $O_3$ in this event could be interpreted as transport from the free troposphere. A general problem is that the authors attribute high GOM to free troposphere in all events while the halogens claimed for GEM oxidation are produced more likely in the boundary layer. I would skip this section.

Line 491: The statement about "the positive correlation between GOM and $N_{coarse}$ observed in our study (Figure 7).." is valid only for events 1, 2, and 3, not for the events 4 and 5. The statement is thus wrong without a qualifier.

Section 4: The conclusions are too unspecificl, they do not reflect on the different patterns of the individual events.

Figure S4: I could not find a reference to it in the manuscript.