Interactive comment on “Fluxes of gaseous elemental mercury (GEM) in the High Arctic during atmospheric mercury depletion events (AMDEs)” by Jesper Kamp et al.

Anonymous Referee #3
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[Summary]
This study reports the measurements of gaseous elemental mercury (GEM) fluxes over the snow cover at Station Nord, Greenland, from late April to mid-May. The authors employed a relaxed eddy accumulation (REA) technique, which is new for its application to the determination of GEM fluxes in the snow-covered polar region. They observed occasional, large emissions of elemental mercury from the snow surface, which is speculated to have resulted from the prior deposition of gaseous oxidized mercury (GOM). I believe that the data presented here are valuable additions to the literature obtained by a micro-meteorological technique rarely employed to date for the measurements of mercury fluxes over the snow cover. In particular, it is found that the mercury (re-)emissions from the Arctic snow surface could take place, at least occasionally, at substantially greater rates than reported in earlier field studies using other types of methodology. The authors convey well the description of their methodology in sufficient details and yet quite concisely so that readers can understand the scientific background of the method, the instrumental configuration in the field and the data screening criteria. On the other hand, the quality of presenting results and discussion needs some significant improvement in both the presented contents and language to make this paper more compelling than currently is, hence the rating of “fair” for “Scientific Quality” and “Presentation Quality”. I recommend major revisions before the present work is considered for final publication.

[Major comments]

1. Section 3 (Results and discussion) appears to need a thorough re-writing, as there are vague statements and unclear logical flows quite often. In addition, some of the speculative statements are given to sound as if they were evidenced in the present study. Since there were no measurements of GOM conducted in this study, any discussions related to the involvement of bromine chemistry leading to the oxidation of GEM to GOM and its temperature dependence as well as the deposition of GOM as a source of GEM re-emitted from the surface snow are all no more than speculations based on prior knowledge of AMDEs. In this regard, the illustration in figure 8 is not really based on evidence from this study and therefore should be dropped. On the same ground, the occurrence of very shallow surface inversions that did not reach the height of instruments for the field data acquisition and its connection to the temporal variations of measured GEM concentrations and fluxes remain speculative, although quite plausible. In my opinion, the sentences in section 3 must be composed more clearly to distinguish between facts and speculations. I also suggest the authors to cut back on the amount of speculative discussions and to increase fact-based and/or quantitative arguments such as those suggested below.

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Figure 8 has been removed. We have changed figure 7, so it becomes more clear. We have added numbers to Fig. 7 to refer to the events and refers to the numbers in the discussion. Furthermore, we have added an extra figure showing the relation between the fluxes and temperature.
The whole section has been thoroughly revised and rewritten.

2. One of the major findings reported in this study is that the magnitude and rate of GEM (re-)remissions from the springtime Arctic snow surface can be much larger than previously reported. In addition to difference in time and location of measurements from earlier studies, the flux estimation technique employed here is different and more sophisticated. To increase the value of the present work, the authors could elaborate more on discussions related to the technical advantage of the REA method over the aerodynamic gradient method (AGM) and whether the two methods can result in significantly different data approval/rejection characteristics, for example, under windy conditions such as during April 26-30. I suppose that small vertical concentration gradients under the enhanced turbulent mixing render the AGM more or less inaccurate as you approach the limit of instrument accuracy. Is it not possible to perform some quantitative micro-meteorological (mathematical) arguments by estimating vertical scalar (tracer) diffusion coefficients and the GEM concentration gradients between the two hypothetical heights above the ground using the present field data, by which the authors could demonstrate the potential advantage of the REA method under windy conditions? I mean, hypothetically. In other words, had the authors derived the GEM fluxes by AGM instead of (or in addition to) REA, could the results have been largely the same? Such arguments could also help decrease the amount of speculative and qualitative statements in section 3.

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We agree that a more detailed argument for choice of measurement method is needed. Therefore following text has been inserted into the manuscript at page 3 line 5:
"*Chamber methods are attractive methods for measuring fluxes because of their low cost and simplicity but suffers from a number of weaknesses. They only capture the flux over a small area, the chamber affects the surface over which the measurement is taken and they can modify physical properties such as light and temperature (Bowling et al., 1998, Fowler et al., 2001). This implies that the measured flux will differ from the natural flux. The AGM is not altering the surface; however, it requires a homogeneous surface several hundred meters upstream the measurement site. Furthermore, it is assumed that the vertical profile is only a product of the vertical turbulent transport; nevertheless fast chemical reactions can affect the profile. The most direct flux measurement technique is the eddy covariance (EC) technique (Buzorius et al., 1998) but close to the surface this technique only works for fast responding monitors (sampling frequency >5 Hz), which is not available for Hg. Therefore, we chose to employ the relaxed eddy accumulation (REA) method (Businger and Oncley, 1990) which is based on EC and the method does not affect the surface. Oncley et al. (1993) reported results with agreement within 20% for EC and REA and a study by Hensen et al. (1996) shows agreement between EC and REA within 10%, a difference that is reported not to be significant because the main error for REA is the determination of the concentration difference.*"

3. Given the orders of magnitude greater re-emission fluxes of GEM than reported previously, the authors should provide a more detailed description of synoptic meteorology during April 26-30 when the episodes of large GEM emissions occurred. Showing a synoptic weather map or two if available and briefly explaining synoptic conditions around the study site (e.g., passage of cyclones) would be great; even greater if such weather maps could be associated with the time series of meteorological data presented in figure 6 and backward trajectories presented in figure 3. The passage of cyclones could also enhance bromine chemistry and hence the production of GOM in the atmospheric boundary layer, potentially serving as a fresh source of oxidized mercury in the

surface snow (e.g., Zhao et al., ACPD, 2017, https://doi.org/10.5194/acp- 2017-427; Toyota et al., ACP, 2014, https://doi.org/10.5194/acp-14-4135-2014); it may be interesting to check with satellite BrO data if the authors can manage within the time frame of manuscript revision.

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The event on April 30 is an extreme event caused by a strong change in the meteorological conditions (possible a front passing) and as we have pointed out in the text this should not be a part of the general analyses. We have tried to make it more clearly in the text, thus we do not thing synoptic weather maps for this period is relevant.

[Minor comments]

1. Equation (2): Something seems to be missing in these equations; as currently formulated, $C_{up} = C_{zero\ air}$ and $C_{down} = C_{zero;}$. I guess C_{up} and C_{down} on RHS must be multiplied by $_{up}$ and $_{down}$, respectively. Please double check.
>>>>> **The equation is as it should be.**

2. Throughout section 3, the authors use the term "GOM" to refer to oxidized mercury retained in the snow after its deposition from the atmosphere. It should have been referred to differently, perhaps simply by "oxidized mercury".
>>>>> **We agree, and have changed the use of GOM to oxidized mercury when it is retained in the snow.**

[Technical suggestions]
P1, L29: inexplicit -> uncertain
>>>>> **Sentence removed**

P1, L29: relaxation -> residence
>>>>> **Sentence removed**

P2, L1: the sea -> seawater
>>>>> **Changed as suggested.**

P3, L30: backwards -> backward
>>>>> **Changed as suggested.**

P4, L9: the vertical turbulent flux of transported quantity is
>>>>> **Changed as suggested.**

P4, L23: must be larger than this threshold FOR AIR SAMPLES to be collected.
>>>>> **Changed as suggested.**

P9, L3: strongLY stable
>>>>> **Changed as suggested.**

P10, L6: extant -> of mercury chemistry and transport dynamics
>>>>> **Changed as suggested.**

P16-17, Figure 3: Add (a), (b), (c) and (d) on top of the trajectories maps.
>>>>> **Changed as suggested.**

P18, Figure 4: Add (a) and (b) on top of the trajectory frequency maps.
>>>>> **Changed as suggested.**

P21, Table 1: The range of GEM fluxes reported in the present study should be -8.0 to 190 ng m^{-2} min^{-1}. Also, it seems useful to include the time (season) of data collection for each study.
>>>>> **There is an error in the table, which has been corrected.**