Reply to Anonymous Referee # 3 Comment on 'Dynamics of gaseous oxidized mercury at Villum Research Station during the High Arctic summer'

The manuscript by Pernov et al. describes the measurement of mercury species and ancillary chemical and meteorological parameters at a site of great importance and interest in the high Arctic at 81°N in Greenland. Unlike many studies which concentrate on Spring when mercury depletion events are common, this study describes two measurement campaigns which were performed in the Summer, which is a relatively less well studied period of the year at these kinds of latitudes.

The manuscript is well written and mostly clear. There is one point however which I think could improve the clarity of the presentation and that is making it more clear when the authors are referring to the free troposphere, the planetary boundary layer or the troposphere in its entirety. As a large part of the discussion revolves around the height of origin and transport of the air masses arriving at the measurement site, it would be useful if the authors were explicit in their descriptions. For instance when they refer to the surface (see for example line 183), does this literally mean the soil or snow, or the surface layer, that is the boundary layer? Still on the same subject, it would be useful for the reader if in the Introduction, the authors could provide a brief description of the typical characteristics of the boundary layer. This would help the reader later on in the Discussion section.

I have a few more specific comments, which are listed below.

> We would like to thank Referee # 3 for the careful review of our manuscript. The comments and suggestions will improve the clarity, presentation, and discussion of the work. We have addressed the referee's concerns and corrected errors in the revised manuscript below with the author's responses in blue. Line numbers refer to the clean version of the revised manuscript. New references are highlighted in yellow.
>
> One of the referee's main points was to indicate where in the atmosphere the authors are referring to in the text. To address this, we have specified in the text which part of the atmosphere we are referring to, e.g., boundary layer, free troposphere, upper troposphere).
>
> The referee also states that a paragraph in the Introduction about boundary layer dynamics would help the reader later in the Discussion, specifically Sect. 3.6 in the previous version of the manuscript. We have made substantial changes to the text and ultimately decided to remove this section. After receiving pushback from all referees, we realize that our hypotheses were too speculative. We have opted not to include a paragraph about boundary layer dynamics in the Introduction, as this was mainly requested to aid the reader later in the Discussion on the section about possible halogen sources.

L19 does tropospheric in this sentence refer to the free troposphere?

Our original intention was to indicate free tropospheric air masses in this sentence. We have removed this sentence in the revised manuscript. We have also amended the text throughout the manuscript to be clear which part of the atmosphere we are referring to.

L24 I think it should be artisanal rather than artisan and in the list of 'natural' emissions, many would include re-emission of legacy mercury, perhaps the authors could make this clear.

We have replaced 'artisan' with 'artisanal'. We have included 'legacy mercury' in our description of previously deposited mercury.

Line 33-35: "The sources of mercury include anthropogenic emissions, e.g., fossil fuel/biomass combustion, and artisanal small-scale gold mines, in addition to natural emissions such as volcanoes, biomass burning, ocean/soil evasion, and reemission of previously deposited/legacy mercury (AMAP, 2011)."

L29 Hg(P) could include elemental mercury adsorbed (strongly) to soot particles as well maybe, which could be important in some instances.

There is ample literature demonstrating gaseous oxidized mercury is readily adsorbed onto particle surfaces. There is literature supporting the adsorption of GEM onto aerosols surfaces (Kim et al., 2012) and literature indicating this is negligible (Seigneur et al., 1998; Otani et al., 1986). Since the soot contained within Arctic aerosols are coated by sulfate and organic matter (Yu et al., 2019), the adsorption of GEM by soot containing aerosols appears negligible in the Arctic atmosphere, given this coating.

Kim, P.-R., Han, Y.-J., Holsen, T. M., and Yi, S.-M.: Atmospheric particulate mercury: Concentrations and size distributions, Atmospheric Environment, 61, 94-102, https://doi.org/10.1016/j.atmosenv.2012.07.014, 2012.

Seigneur, C., Abeck, H., Chia, G., Reinhard, M., Bloom, N. S., Prestbo, E., and Saxena, P.: Mercury adsorption to elemental carbon (soot) particles and atmospheric particulate matter, Atmospheric Environment, 32, 2649-2657, https://doi.org/10.1016/S1352-2310(97)00415-9, 1998.

Otani, Y., Kanaoka, C., Usui, C., Matsui, S., and Emi, H.: Adsorption of mercury vapor on particles, Environmental Science & Technology, 20, 735-738, 10.1021/es00149a014, 1986.

Yu, H., Li, W., Zhang, Y., Tunved, P., Dall'Osto, M., Shen, X., Sun, J., Zhang, X., Zhang, J., and Shi, Z.: Organic coating on sulfate and soot particles during late summer in the Svalbard Archipelago, Atmos. Chem. Phys., 19, 10433-10446, 10.5194/acp-19-10433-2019, 2019.

L34 references for the specific cases mentioned where rapid oxidation occurs would be better, the Angot et al. paper refers only to polar regions.

The authors agree with the referee's recommendation for additional references. We have added von Glasow (2010) which details atmospheric chemistry in volcanic plumes including halogen-induced mercury oxidation, Obrist et al. (2010) which investigated mercury oxidation over the Dead Sea, and Ye et al. (2016) which models mercury species at three sites in the midlatitudes. We have also replaced coastal regions with the marine boundary layer to be more specific and highlighted that these are reactive bromine species.

Line 35 "In locations with elevated reactive halogen concentrations (e.g., polar environments, the marine boundary layer, volcanic plumes, and salt lakes) and especially bromine radicals, GEM is quickly transformed into GOM (Obrist et al., 2010; von Glasow, 2010; Angot et al., 2016; Ye et al., 2016; Wang et al., 2019)."

von Glasow, R.: Atmospheric chemistry in volcanic plumes, Proceedings of the National Academy of Sciences, 107, 6594-6599, 10.1073/pnas.0913164107, 2010.

Obrist, D., Tas, E., Peleg, M., Matveev, V., Faïn, X., Asaf, D., and Luria, M.: Bromine-induced oxidation of mercury in the mid-latitude atmosphere, Nature Geoscience, 4, 22-26, 10.1038/ngeo1018, 2010.

Ye, Z., Mao, H., Lin, C. J., and Kim, S. Y.: Investigation of processes controlling summertime gaseous elemental mercury oxidation at midlatitudinal marine, coastal, and inland sites, Atmos. Chem. Phys., 16, 8461-8478, 10.5194/acp-16-8461-2016, 2016.

Wang, S., McNamara, S. M., Moore, C. W., Obrist, D., Steffen, A., Shepson, P. B., Staebler, R. M., Raso, A. R. W., and Pratt, K. A.: Direct detection of atmospheric atomic bromine leading to mercury and ozone depletion, Proc Natl Acad Sci U S A, 116, 14479-14484, 10.1073/pnas.1900613116, 2019.

L39 my curiosity here, to which type(s) of aerosol are the authors referring?

On Line 50, we refer to the conversion of GOM into PHg due to the cold temperatures which lowers the vapor pressure and because of the high aerosol surface area concentration. The Arctic spring experiences high surface area concentrations due to the Arctic Haze, which is anthropogenic pollution transported from the mid-latitudes caused by the expansion of the polar dome and inefficient removal processes and is characterized by higher concentrations of accumulation mode particles. This peak in surface area, volume, and mass concentration are observed across all Arctic stations in the spring. For clarity, we have specified it is aerosol surface area concentration we are referring to and added a reference to this observed peak in aerosol surface area concentration.

Lines 50-51: "(through condensational processes due to the cold temperatures and high aerosol surface area concentration from accumulation mode particles (Freud et al., 2017))"

Freud, E., Krejci, R., Tunved, P., Leaitch, R., Nguyen, Q. T., Massling, A., Skov, H., and Barrie, L.: Pan-Arctic aerosol number size distributions: seasonality and transport patterns, Atmos Chem Phys, 17, 8101-8128, 10.5194/acp-17-8101-2017, 2017.

L42-44 I was very surprised that none of the work by Dibble or Saiz-Lopez and their co-workers was included in the references here. Is there a reason for this? They would seem pertinent to me.

There is not a particular reason for the omissions of these authors' work. We acknowledge the references in this section could be more inclusive, so we have amended the text as seen in our reply to the comment below.

L50 Again, I think the references should be more inclusive here.

Lines 55-67: "GEM oxidation has been demonstrated to be initiated via photochemical reactions with the Br radical (R1-R2) through modeling studies (Holmes et al., 2006; Holmes et al., 2010; Horowitz et al., 2017), kinetic studies (Donohoue et al., 2006), theoretical studies (Goodsite et al., 2004; Dibble et al., 2012; Goodsite et al., 2012), and observations (Skov et al., 2004; Stephens et al., 2012; Wang et al., 2019).

$$Br + Hg^0 \leftrightarrow HgBr \quad (R1)$$

$$HgBr + Y \rightarrow HgBrY \quad (R2)$$

Where Y could be OH, O_3, NO_2, HO_2, Br, Cl, BrO, ClO, I, IO (Holmes et al., 2006; Hynes et al., 2009; Holmes et al., 2010; Dibble et al., 2012; Jiao and Dibble, 2017a, b). Of which Br, I, and OH have been postulated to be the main species for Y, both globally and in the Arctic (Goodsite et al., 2004, 2012), while NO_2, HO_2, ClO, or BrO have been demonstrated to be candidates for Y by Dibble et al. (2012). Recently, ozone was proposed to be a missing oxidation pathway of HgBr (Saiz-Lopez et al., 2020)."

Horowitz, H. M., Jacob, D. J., Zhang, Y., Dibble, T. S., Slemr, F., Amos, H. M., Schmidt, J. A., Corbitt, E. S., Marais, E. A., and Sunderland, E. M.: A new mechanism for atmospheric mercury redox chemistry: implications for the global mercury budget, Atmos. Chem. Phys., 17, 6353-6371, 10.5194/acp-17-6353-2017, 2017.

Dibble, T. S., Zelie, M. J., and Mao, H.: Thermodynamics of reactions of ClHg and BrHg radicals with atmospherically abundant free radicals, Atmos. Chem. Phys., 12, 10271-10279, 10.5194/acp-12-10271-2012, 2012.

Holmes, C. D., Jacob, D. J., and Yang, X.: Global lifetime of elemental mercury against oxidation by atomic bromine in the free troposphere, Geophysical Research Letters, 33, 10.1029/2006gl027176, 2006.

Hynes, A. J., Donohoue, D. L., Goodsite, M. E., and Hedgecock, I. M.: Our current understanding of major chemical and physical processes affecting mercury dynamics in the atmosphere and at the air-water/terrestrial interfaces, in: Mercury Fate and Transport in the Global Atmosphere: Emissions, Measurements and Models, edited by: Mason, R., and Pirrone, N., Springer US, Boston, MA, 427-457, 2009.

Holmes, C. D., Jacob, D. J., Corbitt, E. S., Mao, J., Yang, X., Talbot, R., and Slemr, F.: Global atmospheric model for mercury including oxidation by bromine atoms, Atmos. Chem. Phys., 10, 12037-12057, 10.5194/acp-10-12037-2010, 2010.

Jiao, Y., and Dibble, T. S.: First kinetic study of the atmospherically important reactions BrHg˙ + NO2 and BrHg˙ + HOO, Physical Chemistry Chemical Physics, 19, 1826-1838, 10.1039/C6CP06276H, 2017a.

Jiao, Y., and Dibble, T. S.: Structures, Vibrational Frequencies, and Bond Energies of the BrHgOX and BrHgXO Species Formed in Atmospheric Mercury Depletion Events, The Journal of Physical Chemistry A, 121, 7976-7985, 10.1021/acs.jpca.7b06829, 2017b.

L53 Some recent work by Gustin and her co-workers seem to be making progress on this front, see for example, Development of an Understanding of Reactive Mercury in Ambient Air: A Review MS Gustin, SM Dunham-Cheatham, J Huang, S Lindberg, SN Lyman, Atmosphere 12 (1), 73.

We thank the referee for sharing this reference. We have included a reference to this work to inform the reader that progress in this area is being made.

Line 69-71 "The exact chemical formulas for GOM and PHg are currently unknown so both species are operationally defined by their detection methods (Landis et al., 2002; Angot et al., 2016), although the development of improved analytical systems for their detection is currently underway (Gustin et al., 2021)."

Gustin, M. S., Dunham-Cheatham, S. M., Huang, J., Lindberg, S., and Lyman, S. N.: Development of an Understanding of Reactive Mercury in Ambient Air: A Review, Atmosphere, 12, 73, 2021.

L55 The sentence beginning Recently would be better at the end of the paragraph, where it is it interrupts the flow

The authors have agreed to the referee's suggestion and moved this sentence to the end of the paragraph.

L63 … thus posing a threat to … ?

> The authors agree with the referee's suggested change and have amended the text accordingly.

L154 Here is a point where a little more description of local boundary layer dynamics would help the reader.

> We have added a short statistical description of the modeled mixed layer depth from HYSPLIT during each campaign in the Methods and Materials sections to help the reader.
>
> Lines 182-185: "For the 2019 campaign, the mixed layer varied from 25 to 554 m, with a median ± median absolute deviation (m.a.d.) of 74 ± 131 m, and a bimodal diurnal profile with minima at night and peaks at 5:00 and 15:00 of ~80 and ~85 m. For the 2020 campaign, the mixed layer varied from 25 to 204 m, with a median ± median absolute deviation (m.a.d.) of 34 ± 21 m, and a bimodal diurnal profile with minima at night and peaks at 13:00 and 18:00 of ~40 and ~50 m."

L163 to be honest I haven't checked all the references, but Greene 2020 is definitely missing.

> The Greene, 2020 reference refers to
>
> 'Arctic Sea Ice: https://www.mathworks.com/matlabcentral/fileexchange/56923-arctic-sea-ice), access: 2020-01-26, 2020.'
>
> Which was listed after Greene, 2017 in the bibliography. This is a webpage reference, so EndNote formatted it as such but without the author, year in the bibliography. This reference has been reformatted in Endnote to reflect the author and year, it is now properly referenced on Line 695.
>
> Greene, C. A.: Arctic Sea ice: https://www.mathworks.com/matlabcentral/fileexchange/56923-arctic-sea-ice), access: 2020-01-26, 2020.

L168 In this section would it be possible to have a small summary table with the parameters listed simply as High or Low, when compared to the averages?

> We have added two tables to the SI, Table S1 and S2 which lists the median and median absolute deviation of ground-level parameters and air mass history, respectively, for each event as well as event and non-event periods for each campaign.

L232 The reference list is rather scarce again.

> We amended the text to include additional references.

Lines 432-436: “Interestingly, none of the events were linked to cold temperatures, which has been previously demonstrated to be associated with mercury oxidation through observations in the Arctic (Cole and Steffen, 2010; Ariya et al., 2015; Steffen et al., 2015), theoretical studies (Shepler et al., 2007), and modeling (Toyota et al., 2014). The stability of the Hg-Br intermediate is highly temperature-dependent (Goodsite et al., 2004; Donohoue et al., 2006; Dibble et al., 2012; Goodsite et al., 2012).”

Cole, A. S., and Steffen, A.: Trends in long-term gaseous mercury observations in the Arctic and effects of temperature and other atmospheric conditions, Atmos. Chem. Phys., 10, 4661-4672, 10.5194/acp-10-4661-2010, 2010.

Shepler, B. C., Balabanov, N. B., and Peterson, K. A.: Hg+Br→HgBr recombination and collision-induced dissociation dynamics, The Journal of Chemical Physics, 127, 164304, 10.1063/1.2777142, 2007.

Toyota, K., Dastoor, A. P., and Ryzhkov, A.: Air–snowpack exchange of bromine, ozone and mercury in the springtime Arctic simulated by the 1-D model PHANTAS – Part 2: Mercury and its speciation, Atmos. Chem. Phys., 14, 4135-4167, 10.5194/acp-14-4135-2014, 2014.

Donohoue, D. L., Bauer, D., Cossairt, B., and Hynes, A. J.: Temperature and Pressure Dependent Rate Coefficients for the Reaction of Hg with Br and the Reaction of Br with Br: A Pulsed Laser Photolysis-Pulsed Laser Induced Fluorescence Study, The Journal of Physical Chemistry A, 110, 6623-6632, 10.1021/jp054688j, 2006.

Dibble, T. S., Zelie, M. J., and Mao, H.: Thermodynamics of reactions of ClHg and BrHg radicals with atmospherically abundant free radicals, Atmos. Chem. Phys., 12, 10271-10279, 10.5194/acp-12-10271-2012, 2012.

L263-4 Are there not any more recent studies which support or further clarify the results from these studies?

We have updated the text to include more recent references about the link between cold temperatures and mercury oxidation.

Lines 432-442: “Interestingly, none of the events were linked to cold temperatures, which has been previously demonstrated to be associated with mercury oxidation through observations in the Arctic (Cole and Steffen, 2010; Ariya et al., 2015; Steffen et al., 2015), theoretical studies (Shepler et al., 2007), and modeling (Toyota et al., 2014). The stability of the Hg-Br intermediate is highly temperature-dependent (Goodsite et al., 2004; Donohoue et al., 2006; Dibble et al., 2012; Goodsite et al., 2012). Lower temperatures aid in the formation of GOM from HgBr, for example, Skov et al. (2004) and Christensen et al. (2004) modeled a surface temperature below -4 °C for mercury depletion to occur in the Arctic, while Brooks et al. (2011) observed a

temperature threshold of -15 °C for mercury oxidation to occur at Summit Station, atop the Greenlandic ice sheet. It should be noted that Brooks et al. (2011) detected oxidized mercury at temperatures above this threshold but not above 0 °C. Tarasick and Bottenheim (2002) analyzed ozonesonde records and observed surface temperatures below -20 °C were required for the occurrence of ozone depletion events. Furthermore, Halfacre et al. (2019) and Burd et al. (2017) demonstrated that a frozen heterogeneous surface is required for the propagation of halogen explosion events."

Cole, A. S., and Steffen, A.: Trends in long-term gaseous mercury observations in the Arctic and effects of temperature and other atmospheric conditions, Atmos. Chem. Phys., 10, 4661-4672, 10.5194/acp-10-4661-2010, 2010.

Shepler, B. C., Balabanov, N. B., and Peterson, K. A.: Hg+Br→HgBr recombination and collision-induced dissociation dynamics, The Journal of Chemical Physics, 127, 164304, 10.1063/1.2777142, 2007.

Toyota, K., Dastoor, A. P., and Ryzhkov, A.: Air–snowpack exchange of bromine, ozone and mercury in the springtime Arctic simulated by the 1-D model PHANTAS – Part 2: Mercury and its speciation, Atmos. Chem. Phys., 14, 4135-4167, 10.5194/acp-14-4135-2014, 2014.

Tarasick, D. W., and Bottenheim, J. W.: Surface ozone depletion episodes in the Arctic and Antarctic from historical ozonesonde records, Atmos. Chem. Phys., 2, 197-205, 10.5194/acp-2-197-2002, 2002.

Halfacre, J. W., Shepson, P. B., and Pratt, K. A.: pH-dependent production of molecular chlorine, bromine, and iodine from frozen saline surfaces, Atmos. Chem. Phys., 19, 4917-4931, 10.5194/acp-19-4917-2019, 2019.

Burd, J. A., Peterson, P. K., Nghiem, S. V., Perovich, D. K., and Simpson, W. R.: Snowmelt onset hinders bromine monoxide heterogeneous recycling in the Arctic, Journal of Geophysical Research: Atmospheres, 122, 8297-8309, 10.1002/2017jd026906, 2017.

L284 mixed layer height which is how high exactly and how does it vary over time, see introductory remarks

The mixed layer height is a modeled parameter that is included in the output of the HYSPLIT model along each step of the trajectory. The percentages above the mixed layer height were calculated by summing the number of hourly steps in which the modeled altitude of the trajectory was above the modeled mixed layer height and dividing by the total number of hourly steps during the event periods. We have included a statistical description of the boundary in the Methods section as described above.

L291 the characteristics of the trajectories for 2019 and 2020 seem very comparable, give or take a few hours, I would have emphasised this point, I think.

> We thank the referee for highlighting this point. One area of concern from other referees was the trajectory length was too short, we have recalculated the trajectory length from 120 to 240 hours to capture the lifetime of aerosols and to obtain a synoptic view of the air mass history. Calculating these statistics for 240 hours produced less clear results, and our analysis of individual events yielded unique features that were not revealed by grouping all event periods. Therefore, we have removed this paragraph from the discussion.

L312 Could the authors add a reference to Section 3.5 and or just include a short explanation of why fires might be important, otherwise the reader is at a bit of a loss as to why they are mentioned here.

> We have indicated in the text why we have included active fires in our analysis of air mass history.
>
> Lines 329-331: "Biomass burning (one of the possible causes of active fires) can emit aerosols covering a large size range and varying chemical composition (Reid et al., 2005), therefore active fires were included to analyze their effect on the air mass history during event periods."
>
> Reid, J. S., Koppmann, R., Eck, T. F., and Eleuterio, D. P.: A review of biomass burning emissions part II: intensive physical properties of biomass burning particles, Atmos. Chem. Phys., 5, 799-825, 10.5194/acp-5-799-2005, 2005.

L325 troposphere, all of it or just the free troposphere?

> We have amended the text on Lines 480-481 and throughout the manuscript to specify the region of the atmosphere we are referring to.
>
> Lines 480-481: "Previous studies have demonstrated the influence of the free troposphere on mercury concentrations within the boundary layer."

L330 This article might be useful in the discussion of free tropospheric Hg dynamics/reactions Weiss-Penzias, P., Amos, H. M., Selin, N. E., Gustin, M. S., Jaffe, D. A., Obrist, D., Sheu, G.-R., and Giang, A.: Use of a global model to understand speciated atmospheric mercury observations at five high-elevation sites, Atmos. Chem. Phys., 15, 1161–1173, https://doi.org/10.5194/acp-15-1161-2015, 2015.

> We greatly appreciate the referee's suggested article. This has provided valuable discussion for our results.

L355 and 358 both parentheses refer to median O3 from August 2010-2018 but the numbers are different, should one be July?

> This is a good catch. The month on Line 369 of the previous manuscript was incorrectly stated. It now correctly reads "July" on Line 376. We have also included another year of ozone observations into this average, which was not available at the time of submission.

L380 Good point, it is unlikely that small decreases in O3 would be noticed in the FT. But as we seem to be lacking oxidants

(Photochemistry of oxidized Hg(I) and Hg(II) species suggests missing mercury oxidation in the troposphere, Alfonso Saiz-Lopez, Oleg Travnikov, Jeroen E. Sonke, Colin P. Thackray, Daniel J. Jacob, Javier Carmona-García, Antonio Francés-Monerris, Daniel Roca-Sanjuán, A. Ulises Acuña, Juan Z. Dávalos, Carlos A. Cuevas, Martin Jiskra, Feiyue Wang, Johannes Bieser, John M. C. Plane, Joseph S. Francisco, Proceedings of the National Academy of Sciences Dec 2020, 117 (49) 30949-30956; DOI: 10.1073/pnas.1922486117)

Can we really ignore the possibility of O3/OH being an Hg oxidant, maybe through heterogeneous reactions? Just a thought.

> We thank the referee for bringing this to our attention and have incorporated this paper into the discussion.

> Lines 448-455: "While ozone mixing ratios were high during GOM enhancement events, they are an order of magnitude below levels reported in the upper troposphere/lower stratosphere (Talbot et al., 2007), and given the slow rate reaction coefficient (Pal and Ariya, 2004), ozone is an improbable first oxidant of mercury during these campaigns (Calvert and Lindberg, 2005), although ozone has recently been identified as a second oxidant of HgI (Saiz-Lopez et al., 2020). While ozone might be acting as a second oxidant of HgI, any depletions of ozone during GEM oxidation, either through reaction with the HgI intermediate or with halogen species, is likely masked by the elevated levels of ozone in the free troposphere. Therefore, the high ground-level temperatures, the increased ozone mixing ratios, and the positive correlations observed between ozone and GOM during all events cast doubt on the local in situ production of GOM in the boundary layer."

> Saiz-Lopez, A., Travnikov, O., Sonke, J. E., Thackray, C. P., Jacob, D. J., Carmona-García, J., Francés-Monerris, A., Roca-Sanjuán, D., Acuña, A. U., Dávalos, J. Z., Cuevas, C. A., Jiskra, M., Wang, F., Bieser, J., Plane, J. M. C., and Francisco, J. S.: Photochemistry of oxidized Hg(I) and Hg(II) species suggests missing mercury oxidation in the troposphere, Proceedings of the National Academy of Sciences, 117, 30949-30956, 10.1073/pnas.1922486117, 2020.

Calvert, J. G., and Lindberg, S. E.: Mechanisms of mercury removal by O3 and OH in the atmosphere, Atmospheric Environment, 39, 3355-3367, https://doi.org/10.1016/j.atmosenv.2005.01.055, 2005.

L390 common source as in emission source? Or do you mean source region or regions?

We acknowledge that the wording could create confusion. The intended meaning was to illustrate a common formation source (both GOM and ozone being formed in the free troposphere) and a common source region (the free troposphere). While we cannot distinguish whether these species were formed in the free troposphere, they were both transported from the free troposphere. During our reorganization of the text, this sentence was however removed.

L405 surface level, is this literally ground level or within the boundary layer?

Schulz et al. (2019) used aircraft campaigns to measure the vertical distribution of BC, they divided the vertical profiles into different levels based on potential temperature. The BC concentration was lowest in the lowermost level and compared well with measurements made over the Arctic Ocean. While these observations were not made at the surface, it is unclear if the lowermost level was within the boundary layer. Therefore, we have used their terminology of the surface level to represent their lowermost observations of BC.

L411 sources or source regions?

Similar to the comment above, we acknowledge this wording can create confusion. We intended to show the different sources regions of aerosol particles and BC (the free troposphere) as well as a common source of GOM, aerosol particles, and BC (biomass burning). During our reorganization of the text, this sentence was however removed.

Section 3.6

In this discussion of possible halogen sources it would be useful to understand the dynamics of the mixing between the boundary layer and the free troposphere. The authors mention a number of surface sources and suggest that these are unlikely to play a role given that Hg oxidation seems to be occurring in the free troposphere L487.

However it is then postulated that biogenic halogen compound emissions and cycling on coarse mode aerosols are a potentially widespread source in the Arctic.

I would have thought that the coarse mode aerosol was less likely to reach the free troposphere than gaseous halogen containing compounds.

Some clarification of the reasoning behind the last two paragraphs of this section is required

We have opted to remove this section from the revised manuscript, although we have tried to explain our reasoning for this mechanism below:

While coarse mode aerosols are less likely to reach the free troposphere than gaseous compounds, the stability stratified atmosphere and reduced moisture in the Arctic inhibits its removal via gravitational setting and removal, respectively, compared to the mid-latitudes. The Arctic boundary layer is also different from mid-latitudes, where thermal convection drives the boundary layer structure. In the Arctic, heat fluxes from open leads, polynya, and wind shear cause convective mixing surface to free troposphere. Coarse mode sea salt can be emitted from these open leads and polynya as well as from sublimation of blowing snow at higher wind speeds. These aerosols are then transported via the free troposphere and subsided into the boundary layer through entrainment and frontal passages.

We realize this hypothesis, while based on literature, is highly speculative. We, therefore, removed this section to make the interpretation of the results more concise.

AMAP: AMAP Assessment 2011: Mercury in the Arctic, Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, xiv + 193 pp., 2011.

Angot, H., Dastoor, A., De Simone, F., Gårdfeldt, K., Gencarelli, C. N., Hedgecock, I. M., Langer, S., Magand, O., Mastromonaco, M. N., Nordstrøm, C., Pfaffhuber, K. A., Pirrone, N., Ryjkov, A., Selin, N. E., Skov, H., Song, S., Sprovieri, F., Steffen, A., Toyota, K., Travnikov, O., Yang, X., and Dommergue, A.: Chemical cycling and deposition of atmospheric mercury in polar regions: review of recent measurements and comparison with models, Atmos Chem Phys, 16, 10735-10763, 10.5194/acp-16-10735-2016, 2016.

Ariya, P. A., Amyot, M., Dastoor, A., Deeds, D., Feinberg, A., Kos, G., Poulain, A., Ryjkov, A., Semeniuk, K., Subir, M., and Toyota, K.: Mercury physicochemical and biogeochemical transformation in the atmosphere and at atmospheric interfaces: a review and future directions, Chem Rev, 115, 3760-3802, 10.1021/cr500667e, 2015.

Brooks, S., Moore, C., Lew, D., Lefer, B., Huey, G., and Tanner, D.: Temperature and sunlight controls of mercury oxidation and deposition atop the Greenland ice sheet, Atmos Chem Phys, 11, 8295-8306, 10.5194/acp-11-8295-2011, 2011.

Burd, J. A., Peterson, P. K., Nghiem, S. V., Perovich, D. K., and Simpson, W. R.: Snowmelt onset hinders bromine monoxide heterogeneous recycling in the Arctic, Journal of Geophysical Research: Atmospheres, 122, 8297-8309, 10.1002/2017jd026906, 2017.

Christensen, J. H., Brandt, J., Frohn, L. M., and Skov, H.: Modelling of Mercury in the Arctic with the Danish Eulerian Hemispheric Model, Atmos. Chem. Phys., 4, 2251-2257, 10.5194/acp-4-2251-2004, 2004.

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