

We thank all reviewers for their input, which we feel has clarified aspects of the manuscript and improved it. As we edited the text, we noted a few other areas where clarity could be increased. For instance, there were some discrepancies in significant digits that has been addressed.

A note to anonymous referees, any line numbers mentioned in the responses correspond to the track changes documents.

Review #1

Summary

In their work on vertical profiles Bromine Monoxide (BrO) in the Arctic, the authors nicely motivate their investigation, put it in context of ongoing scientific discussion and – aside from presenting an excellent data set – clearly point out the scientific novelties of their work: By adapting the new flight pattern of “porpoising” to AMAX-DOAS measurements and performing the radiative transfer simulations on a finer grid, the vertical resolution of BrO profiles is improved. The higher resolved profiles are categorized into four clusters with each being investigated on chemical and meteorological effects. While the finding of high BrO concentrations close to the surface is often reported in literature, the authors used the higher vertical resolution to identify layers of increased BrO just above the surface layer. In general, the presented paper is of “outstanding” quality. In the following suggested minor revisions and technical corrections are listed:

Minor revisions

Line 238: The authors calculate box air mass factors for 4 forward viewing angles. However, these angles are not constant and depend on variations of the pitch angle on short time scales. This becomes visible in Figure S2 where the BrO DSCD peaks during the ascent when the flight altitude becomes less steep (just before 16:24), i.e. the pitch angle is smaller. The authors should include a small discussion on the variation of the pitch angle and how it can affect the retrieved BrO profiles.

The pitch angle is accounted for by adding the average pitch angle from the 3 second integration to the relative viewing angle for each BAMF calculation. Line 249 of the track changes document now says “... by adding the mean aircraft pitch angle to the airplane-fixed viewing angles for each individual observation.”

Line 468: As the lofted BrO cluster is “clearly a large-scale event”, I wonder if it could be compared to satellite retrievals. As the authors speak about this work being the link between ground based and satellite-borne measurements, a small section on satellite comparison for this exceptional case on March 19th would further prove the arguments made in this study.

The figure below has been added to the supplement (section: March 19, 2022 Lofted BrO Profiles) showing TROPOMI BrO columns from 3-19 (via personal communication with Andreas Richter). High column values were observed over much of the region of interest,

consistent with the thought that this was a large-scale lofted event. Lines 520-521 of the main text point to this figure. A more quantitative comparison is beyond the scope of the current work.

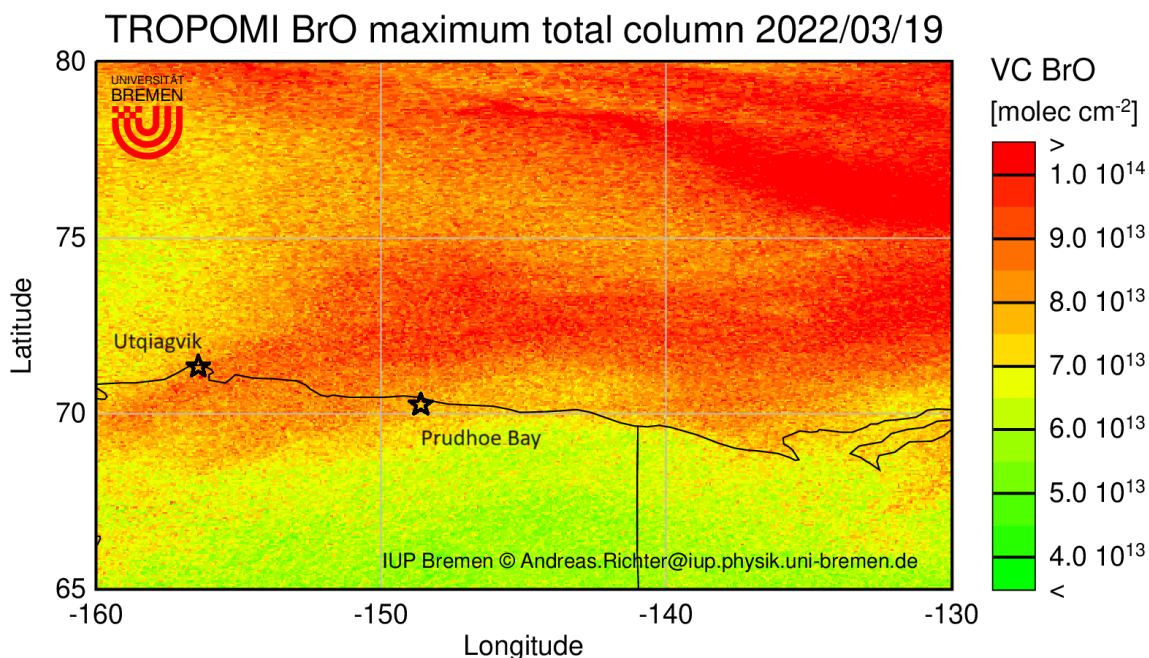


Figure S8 (from personal communication with Andreas Richter).

Technical corrections

Line 31: “should be used as prior profiles” – “should be used as a priori profiles”

This key point was removed at the request of Anonymous Referee #3.

Line 130: “the Purdue ALAR aircraft and a University of Wyoming King-Air aircraft” – Maybe swap the description of both aircrafts to get a nice transition to the next sentence.

This change has been made in the text (line 141).

Line 144/145 Suggestion to rephrase: “NO_x emissions in the area were dominated by two specific facilities during the campaign as observed with the HAIDI nadir spectrometer, and these facilities were located very close to each other (<1 km). For the purposes of this work, Prudhoe Bay will be shown as a point source centred between these two facilities.”

to

“NO_x emissions in the area were dominated by two specific facilities during the campaign as observed with the HAIDI nadir spectrometer. As these facilities are in close proximity with less

than 1km apart, Prudhoe Bay will be shown as a single point source centred between these facilities throughout this study.”

This change has been applied to the manuscript (lines 156-158).

Line 145: “two specific facilities” – Is there a reason as to why the name of the facilities is not mentioned here?

The text now specifies the two facilities as the Central Compressor Plant and the Central Gas Facility (line 156).

Line 154: “field of view of 2.8°” – Is this the FWHM or was this value calculated from the optical properties of the lens?

The text now specifies at lines 168-169, “3.3° full width at half maximum as measured in the field, though the most upward and downward views had some truncation at the edges.”

Line 190/191: “since dSCDs are relative and SCDs depend both on stratospheric trace gas concentrations as well as solar/measurement geometry, which is observation-dependent.” – I don’t think this is a fair comparison of a column vs. a height resolved quantity. Also, the later introduced lower troposphere vertical column density (LT-VCD) has the same advantages as the mixing ratios. I don’t think the use of mixing ratios needs to be motivated here as it is a height resolved quantity and thereby conveys more information than a column quantity like SCD or LT-VCD.

This first sentence of section 2.4 has been removed from the manuscript.

Line 212: “function of particle extinction” – As measurements were conducted in a cloud-free atmosphere, I’d specify this to “a function of aerosol particle extinction”. This sentence could be rephrased to “The observed O4 dSCDs are reliant on the vertical profile of O4 concentration and how light travels through the atmosphere. In a cloud free atmosphere this is mainly a function of aerosol particle extinction, so these dSCD observations can be used to retrieve the vertical profile of the aerosol extinction coefficient.”

This change has been made in the text (line 226-228).

Line 217: Why do you need to run the radiative transfer model with/without O4 absorption? Should this not be “with and without aerosol particles”?

This is explained by Equation 7 of the supplement (line 98). The math depends on how the O4 SCD is impacted by aerosol extinction, and the SCD is calculated by running VLIDORT with and without O4 absorption. The algorithm therefore depends on particle extinction Jacobians from these two model calculations. A reference to equation 7 of the supplement has been added to the text at line 234.

Line 219: “200 observations” – How many observations were there in total?

Line 236 now specifies this is "... roughly 5% of an average 4,141 observations per flight."

Line 220/221: Maybe it's helpful to include the information that on clear-sky days, a temperature inversion causes a stable layered atmosphere where the particle extinction profile is rather constant.

Lines 238-239 now states "...as the mostly clear-sky nature of the days when ALAR flew is associated with strong surface temperature inversions that result in a stable atmosphere."

Figure 2 description: Did the authors perform a sensitivity test on how the results depend on different a priori assumptions? Since it is shown later that the BrO profile correlates with enhanced aerosol extinction, I wonder if there could be an auto-correlation for cases with elevated aerosol layers.

For a quick sensitivity test, data from the lofted BrO day on March 19 were reanalyzed with the particle extinction a priori profile halved and doubled. In both cases, the root mean square difference below 1.5 km altitude between the resulting extinction profiles and the profile used in this work was roughly 0.04 km^{-1} with a mean absolute difference of 34%. This is comparable to the mean relative uncertainty of 27% produced from the original inversion and well below the 50% uncertainty assumed for the BAMF sensitivity studies now discussed in the supplement (line _). Lastly, the a priori used on this day had a maximum extinction at the surface, but each of the three retrievals resulted in the maximum extinction being located above the surface with good correlation to the originally retrieved profile (mean R^2 of 0.76). The results of this test have been added to the supplement (lines 127-132).

For the lofted BrO case on March 19, we are not concerned with auto-correlation, as the dSCDs reveal high BrO values aloft. The figure below has been added to the supplement (Section: March 19, 2022 Lofted BrO Profiles) and shows dSCDs from a single viewing angle for all descents/ascents from all porpoises on March 19. The specific viewing angles were chosen so that the mean viewing angle was as near-limb as possible. As can be seen, the dSCDs are high throughout much of the lowest several hundred meters.

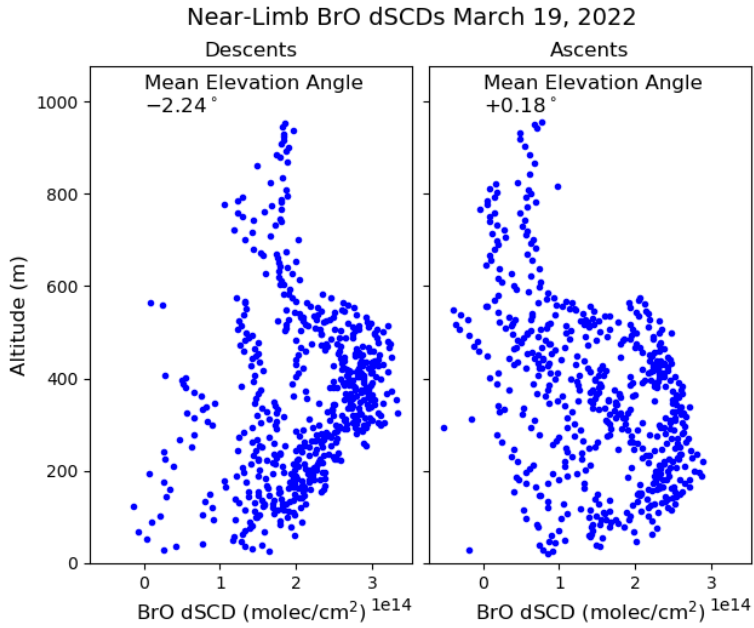
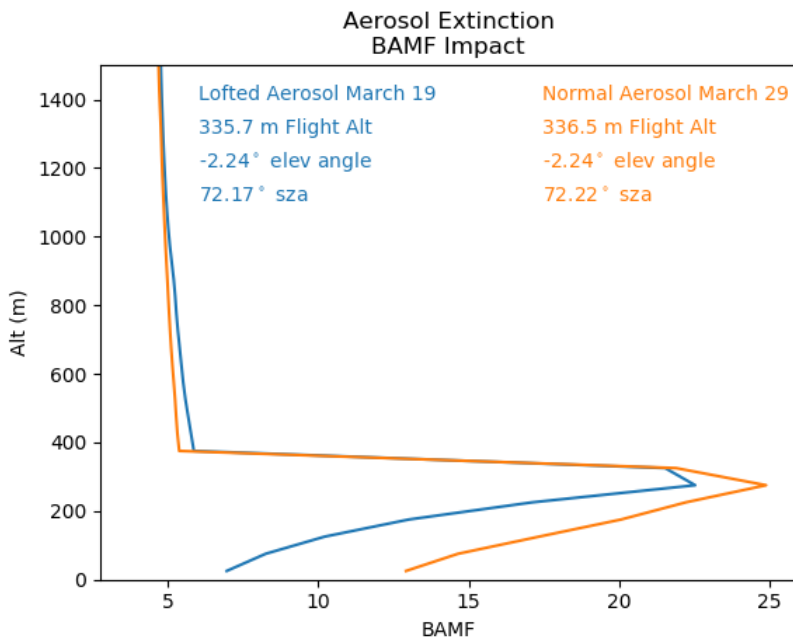


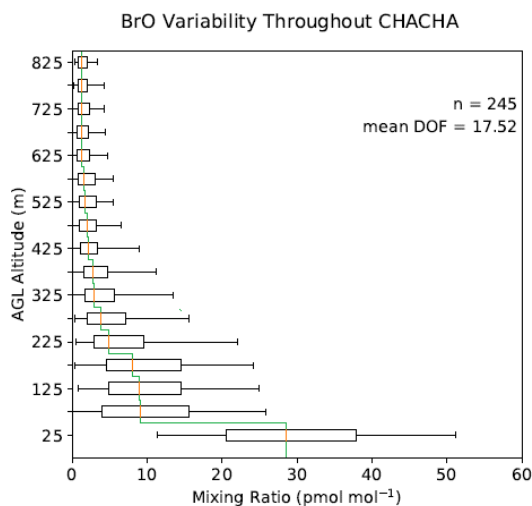
Figure S7

An update of Fig. 2 in the main text now shows the impact of Rayleigh extinction along with Mie extinction, and the two are comparable throughout the lower atmosphere for an average particle extinction day. Therefore, heightened particle extinction will not necessarily have a dominant impact on BAMF calculations.



This figure shows two BAMF calculations for very similar measurement geometries on a lofted aerosol (March 19) day and an average aerosol (March 29) day. BAMFs are generally lower on March 19 due to an optically thicker atmosphere with higher particle extinction. However, the change is likely not drastic enough to create a lofted BrO profile without large BrO dSCDs. Further, the shape of the BAMF profiles is similar, indicating that the larger impact is likely on the magnitude of the retrieved BrO profile than on the shape.

Figure 4: If the y-axis ticks would be at 0, 50, 100, 150, ... it would be easier to identify which data points are mentioned in the text. If the result of the profile retrieval is a box-profile, maybe the box-whisker could be combined with a box-profile depiction, further enhancing the understanding of this plot. See sketched example below (green line added)



This figure has been adapted in the manuscript. The new figure is attached below, with the mean BrO plotted as a box profile.

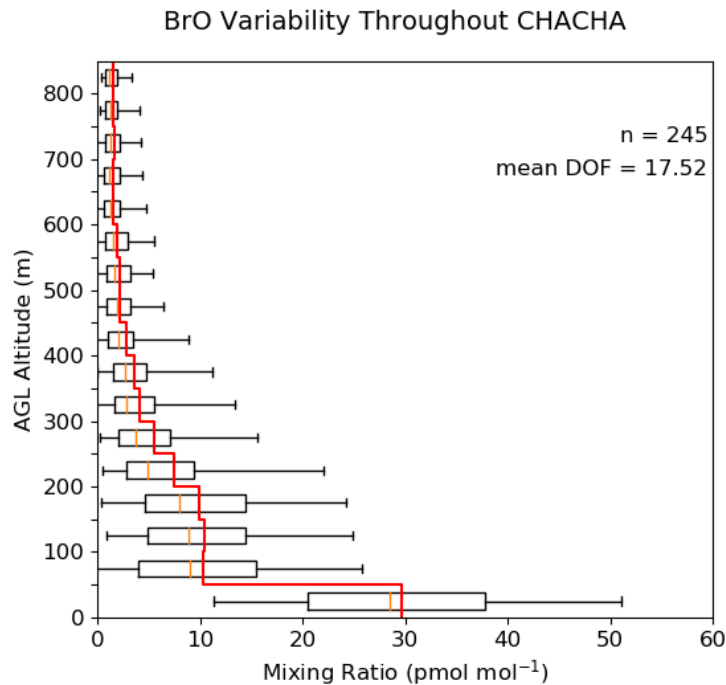


Figure 4

Figure 5: Is it possible to introduce LT-VCD and f200 before this figure appears?

These terms are now defined on line 296, before Fig. 5.

Line 336: “from those surfaces” – “on those surfaces”

This change has been made in the text (line 380).

Line 370 to 374: A small table of plot depicting the distribution of values would help to follow the argumentation in these two sentences.

A table has been put in the text below line 413.

Figure 10: Maybe it is helpful to add the number of cases in each profile cluster – while these numbers should be clear at this point it is quite helpful for understanding the significance of each cluster.

The number of observations has been added to this figure and Fig. 11.

Line 384: The authors should add how to identify the “two most stable potential temperature profiles” to make this sentence easier to understand.

The manuscript now discusses the “profiles with the largest increases in potential temperature with altitude” at lines 428-429.

Line 405: I don't find this part convincing - the lofted BrO cluster is difficult to compare here as it might just show completely different airmasses near ground and above. The Ozone could also just be depleted by other processes or on the particles itself (e.g. by speeding up the sink-term reactions) and thus not be linked to bromine chemistry.

Line 449 now specifies that “these may be cases where reactive bromine was activated by multiphase chemistry on the surface of the particles.”