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 #3

This manuscript offers new DOAS-measured BrO profiles from an aircraft, making observations at various altitudes to profile profiles at a high-altitude resolution. They observe different concentrations and profiles of BrO and report a lofted BrO profile. The paper is well written with a detailed discussion on the meteorological effect, although implications on chemistry are not explored in detail. Overall, the paper adds to the current literature but needs a few details before publication:

Comments:

Line 15: 'at the Earth's surface' is not necessary.

We wanted to be careful to differentiate the Earth's surface from the surface of particles, which is important in this chemistry. This sentence remains unchanged to keep that clarity.

Line 20: MAX-DOAS profile retrievals do not necessarily depend on prior BrO profiles. This depends on the method used for profile retrievals. Please remove this claim from the abstract and clarify this in the text.

The text (lines 20-21 of the revision) now specifies that these profiles and their uncertainties will help some future studies that rely on optimal estimation inversion algorithms.

Key point number 4: This is not a key point from the study but a future outlook – it does not belong in the key points.

This text has been removed.

Line 66: This is mainly driven by chlorine chemistry, with some contribution from bromine chemistry.

This has been specified in the text (line 67).

Line 80: Also mention how climate change is leading to increased iodine chemistry impacts (Benavent et al., 2022) along with bromine and chlorine.

Benavent et al., 2022 and the possible role of iodine in Springtime Arctic chemistry has been added (lines 83-85).

Line 91: Add papers (Tuckermann et al., 1997; McElroy et al., 1999; Carlson et al., 2010; Liao et al., 2011; Benavent et al., 2022; Zilker et al., 2023)

The references have been added (lines 95-97).

Line 93: Please cite original papers that developed profile inversions rather than a later self-cited work.

This sentence is not meant to discuss the development of methods, rather the use of MAX-DOAS to retrieve BrO profiles in the Arctic. This reference has been changed to Frieß et al., 2011 to avoid self-citation (line 99).

Line 96: Please cite the original work that led to the inclusion of halogens in chemistry models instead of only citing your own works, e.g. (von Glasow et al., 2002).

The reference has been added (line 103).

Line 103: 'the same instrument is used in this study.'

This change has been made in the text (line 110).

Line 105-115 –the text is dedicated to the BROMEX campaign that the authors participated in, but all the subsequent studies by other groups that have increased our understanding of bromine chemistry have been ignored.

This paragraph is meant to motivate the CHACHA field campaign, which bears many similarities to the BROMEX field campaign from 10 years prior, hence the focus on the BROMEX campaign. More references have been added to this paragraph to reflect discoveries made outside of this field campaign (lines 120-123).

Line 140: Few flights went much south of Atqasuk, Alaska, at which point the topography started to rise, so the ground elevation was often close to sea level – not clear how the ground level is close to sea level if the topography is rising.

Since there were only a few observations south of Atqasuk where the ground elevation starts to rise, the ground elevation of most observations was close to sea level. The text now states, "Few flights went further south than Atqasuk, Alaska, where the topography starts to rise, so the ground elevation was close to sea level for most observations." (lines 152-153)

DOAS settings – not including HCHO is not standard due to the substantial interference between BrO and HCHO. The authors mention that it did not have any effect, but no evidence for this is provided. Please demonstrate that the exclusion of HCHO did not affect the BrO fits and present a correlation plot between HCHO and BrO through the campaign in high and low HCHO regions.

HCHO in the Arctic springtime is typically very low, at 100-300ppt, at the surface (Sumner et al., 2002), so HCHO was omitted from the fit algorithm due to the noted substantial interference. Adding a HCHO reference to the fit routine for the flight on April 1, 2022 leads to insignificant,

increased BrO dSCDs and almost entirely negative HCHO dSCDs. A detailed reasoning for this omission can be seen in the response to Anonymous Referee #2.

How the authors deal with short-term variations of the aircraft pitch angle is unclear. It would be nice to see some sensitivity analysis or a discussion on the effect of short-term variations of the pitch angle.

The average pitch angle of each observation is added to the relative viewing angles for each BAMF calculation. The mean pitch angle change between observations is 0.3°, so it is not accounted for in the radiative transfer calculations. The field of view of the telescope is much larger than this and is used as the basis for a sensitivity study on the propagation of the elevation angle uncertainty. Impacts of viewing angle uncertainty have been added to a new section of the supplement (BrO Error Propagation and Sensitivity Studies) (lines 188-192).

Line 183: Is the horizontal distance just the flight path or includes the light path?

The text now specifies that the horizontal distance is only from the flight path (line 199).

It is not clear where the reference spectra were collected from – were they collected for each flight individually – how was the area with 'low' trace gas concentration determined? If not, what is the effect of this?

Fifteen reference spectra were used throughout the campaign to account for changing solar zenith angles and stratospheric BrO throughout the campaign. These references are observations from higher-altitude (~1000 m) portions of different flights. Low trace gas concentrations were generally determined by using the lowest SZA observations at the highest flight altitudes. Though some trial and error was involved if large negative dSCDs were present. This is now stated in the supplement at line 6.

It would also be nice to see the mean vertical ozone profiles in the lower 100 m, as bromine chemistry is highly active there. The authors have the data, why not show it?

Ozone data was measured in-situ on the aircraft. As the bottom altitude of the porpoises varied, and only rarely approached the surface during missed approaches at nearby airports, this data is not shown as it skews the shape of the profiles based on available data. Therefore, we only show data above 100 m so that the mean at each altitude is based on a consistent dataset.

Looking at the plots, it is unclear how the 4 clusters differ. The lofted BrO profile is indeed different, but aside from that, the other profiles are not very different when considering the variation.

The K-means clustering algorithm used to create the 4 clustered profiles is an unsupervised algorithm that combines profiles based on how alike they are. The silhouette score test used to choose four clusters shows that three and five clusters fit the data worse than four clusters. The shapes of each of the first three profiles are similar, indicating the surface-based chemistry claimed in this manuscript. However, the magnitude and rate of decrease with altitude are clearly

different for all three cases. The difference between 20 and 40 pmol mol^{-1} of BrO at the surface is significantly different from a chemistry perspective.

The low-BrO day still has 20 pptv at the surface – does the ozone profile reflect this? The ozone profiles in the supplementary text show ozone mixing ratios only above 100 m.

The available mean ozone profile is shown in Figure 11. Ozone data is not available at the surface for most observations. There are instances of Low BrO profiles associated with both high and low ozone concentrations, indicating that this profile can be associated with cases of low reactive bromine chemistry as well as ODEs that can result in low BrO concentrations.

The authors should include a comparison with satellite observations, especially for the lofted BrO day, which looks like a widespread event.

A new figure (S8) has been added to the supplement showing TROPOMI BrO observations (via personal communication with Andreas Richter) that shows high BrO columns on March, 19 over the measurement region, agreeing with the assertion that this is a large scale event. A full satellite comparison is beyond the scope of this work.

If inhibited vertical mixing explains values close to the surface, does that mean that the surface ozone was completely depleted?

As seen in figure 11, ozone was generally depleted towards the surface. Although O_3 measurements from the aircraft were not generally made below 100m, this profile shape was common throughout the campaign, consistent with inhibited vertical mixing. Previously, Peterson et al. (2016) reported full ozone profiles for several BROMEX profiles over Utqiaġvik and Atqasuk, with comparisons to ground-based data, showing that significant changes in ozone levels can occur within the lowest 100 m. Oltmans et al. (2012) previously reported ozone vertical profiles up to 2 km using ozonesonde data at Utqiaġvik.

Why is there a cutoff effect for O4 but not BrO or NO2?

The dSCDs of O_4 were often negative due to light path truncation by the surface. The same effect occurs in the observations of BrO and NO_2. In this observation however (from figure S1), the concentration of BrO and NO_2 was considerably higher at the surface than at the altitude at which the reference spectrum was recorded. Therefore, the higher concentrations overcome the shorter path length, resulting in positive absorption. This has been added to the caption of Fig. S1.

References:

Frieß, U., Sihler, H., Sander, R., Pöhler, D., Yilmaz, S., and Platt, U., The vertical distribution of BrO and aerosols in the Arctic: Measurements by active and passive differential optical absorption spectroscopy, J. Geophys. Res., 116, D00R04, doi:10.1029/2011JD015938, 2011.

Oltmans, S. J., Johnson, B. J., and Harris, J. M., Springtime boundary layer ozone depletion at Barrow, Alaska: Meteorological influence, year-to-year variation, and long-term change, J. Geophys. Res., 117, D00R18, doi:10.1029/2011JD016889, 2012.

Peterson, P. K., Pratt, K. A., Simpson, W. R., Nghiem, S. V., Pérez, L. X., Boone, E. J., Pöhler, D., Zielcke, J., General, S., Shepson, P. B., Frieß, U., Platt, U., and Stirm, B. H.: The role of open lead interactions in atmospheric ozone variability between Arctic coastal and inland sites, Elementa, 2016, https://doi.org/10.12952/journal.elementa.000109, 2016.

Sumner, A. L., Shepson, P. B., Grannas, A. M., Bottenheim, J., Anlauf, K. G., Worthy, D., Schroeder, W. H., Steffen, A., Domine, F., Perrier, S., and Houdier S.: Atmospheric chemistry of formaldehyde in the Arctic troposphere at Polar Sunrise, and the influence of the snowpack, Atmos. Environ., 36, 2553–2562, 2002.