

Review of Frieß et al. (2022): Source Mechanisms and transport Patterns of tropospheric BrO: Findings from long-term MAX-DOAS Measurements at two Antarctic Stations

The manuscript presents 19 and 10 years of MAX-DOAS observations of tropospheric BrO and aerosols from the Antarctic stations of Neumayer (NM, 70° 40' S, 8° 16' W) and Arrival Heights (AH, 77° 49' S, 166° 39' E), respectively. Using this extensive data series, the authors investigate linear correlations between numerous parameters such BrO at surface, aerosol extinction, wind speed, contact time with sea ice, etc. After investigating these regressions, the authors conclude that (1) there is a correlation between BrO and aerosol extinction indicating that airborne saline particles are a dominant source of bromine; (2) there is a correlation between BrO and air masses' contact time with sea ice and with ice sheet, indicating that not only the sea ice but also the snow over the ice sheet is a source of bromine; (3) when the sea ice retreats, the ocean is also a source of reactive bromine. Furthermore, the authors suggest possible source regions of reactive bromine for NM and for AH sites. In addition, the authors point out the detection of an early morning peak of tropospheric BrO and also of sustained uplifted plumes of BrO crossing the continent for several days.

Throughout the manuscript, the authors discuss sources and processes of tropospheric reactive bromine in the Antarctic, addressing relevant scientific questions. Furthermore, to the reviewer's knowledge, this is probably the largest dataset of BrO observations in the pristine Antarctic troposphere. Chemical models and the scientific community can indeed benefit from these observations.

The manuscript is well presented and the methodology is well described. Overall, publication is recommended after addressing the following comments.

General Comments:

- Although the number of variables studied in the regression analysis is outstanding, the conclusions gathered after those regressions seem sometimes overstated. For instance, a correlation of -0.13 (BrO-radiation) is stated as “anti-correlated” and a conclusion is obtained after. However, a $R = -0.13$ indicates a rather low (or even negligible) linear relationship between two variables since knowing variable A would explain only 1.69% of the variance of variable B (i.e., close to negligible correlation). Similar applies to BrO-aerosols at AH ($R=0.28$) or BrO-ice at AH ($R=0.17$), etc. A revision of statements like “this provides evidence”, “this confirms” etc throughout the manuscript is suggested along with the conclusions gathered based on not high (or even very low) R . Also, all this study is based on linear regression between pair of variables. Given the complex nature of the system and the huge amount of data and variables studied, could a multivariate regression provide additional information?
- Throughout the manuscript, some context of the obtained values of BrO and aerosols is missing (i.e., how the values at NM and AH do compare with previous published works in Antarctica and/or the Arctic region?).
- The manuscript may benefit if providing the corresponding BrO VMR (i.e., ppt) along with VCD (e.g., detection limit of BrO, P16, L342). Also including BrO ppt and aerosol ranges observed throughout all the years could provide a nice overview of the data.
- In the manuscript, aerosol extinction coefficients are discussed at both sites based on MAX-DOAS observations. Is there any cloud filter applied when retrieving those aerosols? If no cloud filter is applied, the obtained “aerosol” results might well be due to clouds instead. This limitation should be mentioned in the manuscript so the conclusions reached after the “aerosol” data could be taken with caution.
- Please, discuss the possible influence of stratospheric BrO on the observations at high SZA (see also specific comments to this regard, e.g., P9, L220-224).

Specific Comments:

P1, L19: "BrO can be sustained for several days". A range deliming "several" could assist the conclusion (e.g., ~2 days).

P2, L31: missing the reference to the review work of Angot et al. 2016.

P3, L57: also for CCM (e.g., Fernandez et al., 2019).

P5, L111: missing the reference to the 25-year climatology of Silva et al., 2022.

P6, L131-138: What is the FOV of each instrument? Why 0° is not included in the scan? Is the negative elevation angle used in the retrieval (also P10, L235)?

P7, L175-177: Does this affect to any of the conclusions reached in the manuscript?

P8, L196-198: "Weighted by the time difference", please describe a bit better this approach.

P8, L208: To avoid self-referencing, could only one of the 4 references to HEIPRO be used?

P9, L220-224: "all dSCDs measured during one hour serve as input for a single retrieval". For low SZA, this might be a good approach. However, for high SZA this most probably provide results including stratospheric information since a single a priori profile (and AMF) is assumed constant during that hour. If the measurement vector consists on observations at each given angle during 1 h, wouldn't the stratospheric information change a lot during 1h at high SZA? How could this affect the authors' results at high SZA (e.g., the BrO peak the authors observe after sunrise and before sunset; P20, L1)?

P11, L278: Why is the residence time weighted?

P13, figure 4: Please, include the error bars in the figure. Also, why are there 4 different BrO profiles retrieved for one single data and time?

P15, last paragraph: Since on 7-9th October there was very high aerosol extinction coefficient, how reliable are the BrO VMR values retrieved on those days?

P16, figure 5: Since e.g. the pressure does not seems to be addressed, for the sake of simplicity, that label and graph could be removed.

P17, L363: This section presents the spatio-temporal variability of BrO. Since aerosols are also addressed throughout the manuscript, a similar section for aerosols would benefit the draft.

P17, L364: Does BrO VCD refer to total column VCD or only tropospheric column VCD? Please, clarify in the text. Also, since as stated by the authors the MAX-DOAS observations are sensitive to the first 1.5 km (P9, L232), should not the VCD refer only to those km?

P18, L364: By the different meteorological conditions and also by the location of each research site.

P19, L390: Please, state the meaning of having higher bromide-to-sodium ratio.

P19, L401: How does the surface BrO daily evolution compare to the work of Nasse et al. 2019 where, with a LP-DOAS, they detected up to 60ppt around noon?

P21, L448: Regarding BrO getting into the free troposphere, what is the evolution of the boundary layer height?

P22, figure 10: Is this figure considering only BrO data above detection limit?

P22, L56-457: p-values close to 0 indicate the *significance* of the R obtained.

P23, figure 11: Which BrO and aerosol from MAX-DOAS are used? Surface values? (also in P24, L:461)

P25, L523: The source-receptor analysis seems very clarifying. Please detailed a bit better the procedure behind this sort of analysis (e.g., which altitude of the BrO profile is used for this analysis)?

P25, L527: Do observations at Belgrano or Halley support the FRIS as a BrO hot spot? Also P26, L542; P27, L547.

P27, L557-558: Do observations at Dumont d'Urville support the TNB as an aerosol hot spot?

P30, figure 17: The time referred to is 00:30. Given the high SZA, how can the stratospheric influence be excluded?

P30, L586-588: Can a stratospheric intrusion be ruled out? Are O3 values available?

P32, L610: Please provide the BrO VCD also in ppt (easier to compare with other ground-based observations).

P32, L613-615: (Snow surface as a BrO sink during the day). This may contradict with e.g. the work of Saiz-Lopez et al. 2007 with a LP-DOAS. Same applies with the observed diurnal variation at NM with an early morning peak.

Technical Corrections:

P1, L1: Polar Regions shall be polar regions (no block letters).

P5, L109: Katabatic shall katabatic (no block letter).

P15, L319: 28. August shall be 28th August. This happens to all dates throughout the manuscript.

P31, L591: A “,” is missing after Thus

P31, L598: The “dynamics chemistry”? do the authors refer to the chemistry and the dynamics?