Response to Editor

Editor Comments and Suggestions for Authors:
Dear authors, I think you have changed your manuscript thoroughly and I have only a few minor points:

Author's Response: We sincerely thank the Editor for the recognition of our work. Please kindly find our point-by-point responses to the issues raised below in blue, and the changes to the manuscript in orange.

Comments:
QE.1: Please make sure that the Abstract length is as required (250 words)

AE.1: We have ensured that the Abstract is within the required 250-word limit.

QE.2: change "strong agreement" to good agreement

AE.2: We have revised the corresponding section of the manuscript. Please see P38 (lines 759–760):

"The MAX-DOAS BrO partial columns (0–4 km) show good agreement with overpassing GOME-2B tropospheric columns."

QE.3: what means "enhanced Bro chemistry"? Why is chemistry "enhanced"? Please clarify

AE.3: Thank you for pointing that out. The phrase of "enhanced BrO chemistry" is not appropriate, so we have rephrased the sentence in the relevant text. Please see P38 (lines 763–765):

"In addition, BrO partial columns exhibit a significant negative correlation with GEM, with higher BrO generally associated with lower GEM concentrations, suggesting that enhanced bromine is closely involved in springtime atmospheric mercury depletion."

QE.4: you explain what is done regridded ERA5 for use in p-TOMCAT regarding horizontal resolution. What is done regarding vertical resolution? This information should be added

AE.4: The corresponding information regarding the vertical resolution has been added to the revised manuscript. Please see P10 (lines 267–269):

"While ERA5 provides 37 levels, p-TOMCAT setups commonly use a reduced number of hybrid sigma-pressure levels (e.g., 31 levels reaching up to ~10 hPa) with a spacing of approximately 100 m in the boundary layer (~60 m in the surface layer) and 1–1.5 km in the vicinity of the tropopause."

Response to Reviewer #3

Reviewer #3 Comments and Suggestions for Authors:
The authors present a comparison of ground based BrO MAX-DOAS retrievals and satellite based BrO retrievals at Ny-Ålesund with modelling work to explore the role of meteorological conditions and time an air mass spends in sea ice regions in explaining variability in observed BrO. This work has been reviewed already, and the authors have done a nice job addressing the concerns raised by the reviewers. I have a few minor points that should be addressed prior to publication.

Author's Response: We thank Reviewer #3 for the valuable comments, which have greatly helped this manuscript. Please kindly find our point-by-point responses to the issues raised below in blue, and the changes to the manuscript in orange.

Comments:
Q3.1: In line 509, what level AERONET data are you using? Generally for publication, at least level 1.5 which is cloud screened is recommended. You should also acknowledge the site PI (https://aeronet.gsfc.nasa.gov/new_web/data_usage.html).

A3.1: We used AERONET Level 2.0 data, and the corresponding section of the manuscript has been modified as follows (P21, lines 503–504):

"In addition, we compared MAX-DOAS AOD with collocated AERONET Level 2.0 observations at Ny-Ålesund (https://aeronet.gsfc.nasa.gov/)."

Also, we have acknowledged the site PIs in the manuscript. Please see P40 (lines 830–831):

"We gratefully acknowledge Victoria E. Cachorro Revilla and Christoph Ritter for their efforts in establishing and maintaining the Ny-Ålesund site and for providing AERONET Level 2.0 data."

Q3.2: Line 241 It should probably be stated a little more clearly that passive remote sensing is not the best tool to examine BrO production during low visibility conditions. I understand it is what we have as a community for long term observations and some data are better than no data, but we need to be clear about the limitations in this context. The cloud effects on the retrieval are not trivial, and as a community we need to do more work to evaluate the quality of these retrievals in poor visibility if we want to keep using these data in this context.

A3.2: The corresponding section of the manuscript has been modified as follows (P10, lines 261–262):

"As noted above, passive remote sensing has limitations when studying BrO under low visibility conditions, as it does not account for cloud effects. However, it remains valuable for long-term observations."

Q3.3: Throughout the paper, you refer to MYI and FYI. You can't say anything about the impacts of the sea ice itself in this work. What you are discussing are the role of first year sea ice regions and multi-year sea ice regions as a whole not just the ice itself. It may seem pedantic, but it is an important distinction and should be clear throughout the paper.

A3.3: We thank the reviewer for this clarification. We agree that our analysis does not refer to sea ice itself, but rather to snow over different sea ice regions (MYI and FYI). We have revised the manuscript throughout to explicitly reflect this distinction. As an example, the revised text in the Abstract is shown below (P1, lines 20–23):

"Five-day backward trajectories (0–3 km) showed significant BrO correlation with sea-ice contact time, particularly over multi-year ice (MYI) regions, which contributes comparably to first-year ice (FYI) regions in the total blowing-snow-sourced bromine flux, highlighting the comparable importance of snow over MYI and FYI regions in driving bromine explosion events."

Q3.4: Figures 7 and 9 have a mixture of BrO columns and BrO mixing ratios. In my view it makes more sense to talk about columns throughout. The mixing ratio depends on both boundary layer dynamics and sources. If we want to get at impacts on BrO we should be talking about columns.

A3.4: Thank you for this comment. In Figures 7 and 9, the variables used for correlation analysis with BrO columns contain only a single vertical layer, making them directly comparable to BrO columns. In contrast, the variables used for correlation analysis with BrO VMR (e.g., contact time with sea ice and open ocean) contain vertical structure with multiple layers, which allows us to investigate relationships across different vertical layers. In addition, in Figures S11 and S13, we examine the relationship between BrO columns and the total contact time with sea ice and open ocean (summed over 0–3 km), yielding similar conclusions. Please see P20 (lines 495–501):

"We also analyzed correlations between BrO partial columns and AOD, using total sea-ice and open-ocean contact time summed vertically over 0–3 km, as shown in Figs. S11–14. For both 2019 and 2020, BrO partial columns exhibit stronger positive correlations with total sea-ice contact in the first half of the period (March 1–April 15) than in the second half (April 16–May 31), while correlations with total open-ocean contact tend to increase in the second half. AOD shows generally weaker correlations and a somewhat different seasonal pattern than BrO. These vertically integrated results are generally consistent with the profile analysis (Figs. S7–10) and indicate a seasonal shift in the relative influence of sea-ice and open-ocean contact on BrO and aerosol variability."

Response to Reviewer #4

Reviewer #4 Comments and Suggestions for Authors:
In the paper "Tropospheric bromine monoxide in Ny-Ålesund: source analysis and impacts on atmospheric chemistry", the authors utilize 7-year MAX-DOAS observations, alongside model simulations and surface measurements, to identify the sources of BrO in Ny-Ålesund. The study is comprehensive, and the findings are interesting, particularly the discussion regarding the distinct roles of MYI and FYI. However, the manuscript is overly lengthy, causing the focus to be obscured by excessive details. Thus, I recommend a major revision, specifically requiring the authors to significantly condense the text and sharpen the focus on their key contributions. I have some specific suggestions as follows:

Author's Response: We thank Reviewer #4 for the valuable comments. Following the suggestions from the reviewers in the previous round, we have substantially expanded the manuscript and made corresponding revisions throughout. We have also made further revisions to address the remaining comments. Please kindly find our point-by-point responses to the issues raised below in blue, and the changes to the manuscript in orange.

Comments:

Q4.1: Please separate each paragraph using a blank line.

A4.1: We have separated each paragraph using a blank line.

Q4.2: L56-57, the snow on the sea ice, rather than the sea ice itself, is actually the source of halogens.

A4.2: A similar issue was also raised by Reviewer #3, see above Q3.3 and our response in A3.3. Here we updated the corresponding text as follows (P2, lines 59–60):

"Potential sources of reactive bromine include: open-ocean sea spray (Sander et al., 2003); snow over first-year sea ice (FYI) (Jones et al., 2006; Simpson et al., 2007b) and over multi-year sea ice (MYI) (Peterson et al., 2019; Huang et al., 2020); …".

Q4.3: The structure of the introduction section is not good, so that it needs a substantial improvement. For instance, L79, the change of topic is very abrupt. Aside from that, in L56, the authors discuss the sources of halogens, but in L120, they discuss the sources again. Please combine these two paragraphs together.

A4.3: We have moved the text at L79 to the discussion in Section 3.6 ("Potential role of MYI in BEEs"). We have also combined and revised these two paragraphs (L56 and L120), now they read (P2, lines 59–74):

“Potential sources of reactive bromine include: open-ocean sea spray (Sander et al., 2003); snow over first-year sea ice (FYI) (Jones et al., 2006; Simpson et al., 2007b) and over multi-year sea ice (MYI) (Peterson et al., 2019; Huang et al., 2020); frost flowers (Kaleschke et al., 2004; Nghiem et al., 2012); sea salt aerosol (SSA) from polynyas or open leads (Kirpes et al., 2019; Criscitiello et al., 2021); tundra snowpack photochemistry (Pratt et al., 2013); blowing-snow-sourced SSA (Yang et al., 2008, 2010; Jones et al., 2009; Choi et al., 2018; Huang et al., 2020); and stratosphere-to-troposphere transport of BrO (Salawitch et al., 2010). Field studies in Alaska and laboratory experiments have shown that acidic saline snowpacks can release reactive bromine when exposed to sunlight and ozone (Wren et al., 2013; Pratt et al., 2013). Reactive bromine fluxes from snowpack in Alaska, ranging from 7 × 107 to 1.2 × 109 molecules cm-2 s-1, based on direct measurements of BrCl and Br2, were reported. These are in line with those needed in models to reproduce BEEs and ODEs (Custard et al., 2017). In contrast, Yang et al. (2024) derived a much lower average snowpack release flux of 1 × 107 molecules cm-2 s-1 or below from field measurements in Eureka, Canada (86.4°W, 80.1°N), suggesting that coastal snowpack is a weak reactive bromine source. Moreover, based on a mass balance approach, Yang et al. (2024) estimated that the lifetime of atmospheric reactive bromine as a family is 17–42 days, which is longer than the 4–10 days reported in previous studies (von Glasow et al., 2004; Yang et al., 2005). Fresh frost flowers, although highly alkaline and saline, likely make only minor or local contributions due to their limited spatial extent on open leads (Obbard et al., 2009; Lieb-Lappen and Obbard, 2015). Stratospheric influence on tropospheric BrO columns also appears limited (Theys et al., 2011).”

Q4.4: Please better highlight the key findings of this study. Specifically, the analysis of the roles played by FYI, MYI, and blowing snow is particularly interesting in my mind, and I suggest focusing the manuscript more heavily on these aspects.

A4.4: We have revised the Summary section to highlight the key findings of this study, with particular emphasis on the roles of snow over FYI regions, snow over MYI regions, and blowing snow. Please see P38 (lines 757–805):

“In this study, we integrated seven years (2017–2023) of MAX-DOAS BrO observations in Ny-Ålesund with GOME-2B measurements, meteorological data, p-TOMCAT simulations, and HYSPLIT backward trajectories to explore the sources and causes of variability in tropospheric BrO during polar spring (March to May). The MAX-DOAS BrO partial columns (0–4 km) show good agreement with overpassing GOME-2B tropospheric columns. The mean monthly BrO partial column exhibits a decreasing trend from March to May (1.97 × 1011 molecules cm−2 d−1) with pronounced interannual variability. The most substantial short-term perturbations occur in early spring (March). Episodes of enhanced BrO frequently coincide with ODEs, highlighting the role of reactive bromine in Arctic atmospheric chemistry. In addition, BrO partial columns exhibit a significant negative correlation with GEM, with higher BrO generally associated with lower GEM concentrations, suggesting that enhanced bromine is closely involved in springtime atmospheric mercury depletion.

Strong correlations between MAX-DOAS–retrieved BrO and aerosol extinction were observed at Ny-Ålesund, indicating a potential link between airborne particles and enhanced reactive bromine. Sensitivity simulations using p-TOMCAT further demonstrate that heterogeneous recycling of inactive bromine species (like HBr, HOBr and BrONO2) on aerosols plays a dominant role in sustaining elevated BrO levels. For example, when heterogeneous reactivation was disabled, BrO partial columns were only about 15% of those when heterogeneous reactivation was enabled, and BrO/Bry ratios changed by a factor of 6.7 times, strongly indicating that, without heterogeneous reactivation, pure gaseous-phase photochemical reactions alone are insufficient to maintain BEEs.

Backward trajectory analyses further revealed that enhanced BrO during BEEs is positively correlated with total sea-ice contact time. Notably, the correlation coefficients between BrO and sea-ice contact time are substantially larger (0.26–0.42) under high wind speeds (> 7 m s−1) than under low wind speeds (0.10–0.19, ≤ 7 m s−1). These results suggest that strong winds enhance reactive bromine release from the sea ice surface, likely through the production of SSA from blowing snow, as previously proposed. This interpretation is further supported by the significant correlations between p-TOMCAT modelled SSA and retrieved BrO, and between the corresponding bromine emission fluxes from blowing snow and retrieved BrO, as shown in Figure 13.

During BEEs, total sea-ice surface snow contact accounts for more than 50% of air-mass contact time, whereas open-ocean contact accounts for less than 10%. Moreover, more than half of the air masses within the boundary layer originate from regions covered by MYI during BEEs, particularly areas north of Greenland and the Canadian Arctic Archipelago. This is likely due to Svalbard’s geographic location, which places it downwind of southward-flowing air masses approaching Ny-Ålesund. Further analyses show that air masses during BEEs spend approximately 2.4 times longer in contact with MYI regions than with FYI regions; this ratio increases to about 2.8 under strong wind conditions (> 7 m s−1). Accumulated blowing-snow-sourced bromine emissions over MYI regions contribute, on average, about 54% of the total sea-ice-sourced bromine. These results indicate that snow over MYI regions plays an important, previously underappreciated role, in determining BEEs at Ny-Ålesund, comparable to snow over FYI regions. Due to limitations in the model’s representation of sea-ice types and the lack of field data to constrain key parameters, such as snow salinity over MYI and FYI, we could not quantify their relative contributions to Ny-Ålesund BEEs or to BEEs across the whole Arctic. However, our findings still provide informative insights into the mechanisms and controlling factors of polar BEEs, which may be relevant for other coastal sites or across the pan-Arctic.

Note that this work could not rule out direct emissions of reactive bromine from the sea-ice surface or the snowpack above it, given positive correlations between observed BrO and ice contact time, and between BrO and the reactive bromine flux from the snowpack. Notably, under strong wind conditions, the correlation coefficients increase markedly relative to calm conditions, underscoring the dynamical influence on reactive bromine release, though the underlying microphysical processes remain unclear. Currently, the debate centres on two compelling mechanisms: the so-called "air-pumping" effect (Toyota et al., 2011) and the blowing-snow effect (Yang et al., 2008). Our findings highlight the need for quantitative measurements of snowpack emissions over sea ice, as well as polar boundary layer bromine budget analyses (Yang et al., 2024), to improve our understanding of the relevant processes. This is crucial in terms of accurately constraining models in reproducing polar springtime BEEs and ODEs, and assessing their climate impacts through effects on atmospheric oxidizing capacity."