Comments from anonymous Referee #1:

We would like to thank the reviewer for his/her helpful comments. We hope that we could address all questions and unclear points satisfactorily.

Legend: Author comments in blue, Referee comments in black.

General Comments

This manuscript investigates the meteorological conditions spatial distributions of BrO in the Arctic leading to ozone depletion events as observed by ozone sondes and an in situ ozone monitor in Ny-Alesund, Spitsbergen. This is done by separating the ozone time series into ODE and non-ODE periods using a threshold value, and by calculating maps of the anomaly of meteorological parameters, sea ice conditions and BrO VCDs over the Arctic for both situations. Based on these anomaly maps, the impact of the spatial distribution of temperature, wind speed, boundary layer height and pressure as well as BrO and sea ice coverage has been investigated. The manuscript confirms many findings from previous studies, such as the impact of polar cyclones on ozone depletion and the occurrence ODEs during high wind speeds, which are probably due to heterogeneous release of reactive bromine from saline aerosols. It is found that certain distributions of polar low- and high-pressure systems lead to the southward transport of ozone depleted air towards Ny-Alesund. Furthermore, the seasonal and inter-annual variation of these anomalies is investigated and a case study on a particular ODE is presented.

The meteorological conditions leading to a release of reactive bromine and a subsequent ozone depletion are still not fully understood. Therefore, this manuscript provides a valuable contribution to this field of research and fits well into the scope of ACP. The results of the study are described appropriately, but I feel that the description of the methods requires substantial revision. In particular, the “composite analysis” method presented in Sect. 2.7., which represents the key method of the study, should be re-written since it is lacking conciseness and is difficult to understand (see the specific comments below).

Following the suggestion of the reviewer, Section 2.7 has been rewritten (see below).

Sect. 3.2.1. describes in detail the impact of the ODE threshold values on the resulting anomaly maps, and concludes that there is only little impact on a qualitative basis. I therefore suggest to skip this section, and simply add the final sentence of this section (“No major differences in BrO and meteorological anomalies are observed when changing the ozone threshold value”) to the methods section.

We have decided not to remove Section 3.2.1 as an additional discussion point was added there and as we think it is important to show the effects of different thresholds as these are chosen more or less randomly.

Specific Comments

P2, L47: In addition to chlorine, I think it would be worth mentioning iodine as a potential booster for ozone depletion (Benavent et al, 2022).

We agree that iodine could be relevant, and therefore already mentioned it further below in the text using the same reference (P3, L58-60).

P3, L68: Please add a reference for the lifetime of BrO and specify what exactly is meant with this value. While the photolytic lifetime of a BrO molecule is quite short, the lifetime of BrO in a certain air mass depends on various parameters, such as the presence of saline surfaces for recycling.

Following the suggestion of the reviewer, we have updated this sentence and added references:

The photolytic lifetime of BrO is approximately one minute (e.g., Lehrer et al., 2004; Pratt et al., 2013)

Here, we only refer to the photolytic lifetime of BrO. Although lifetimes are given in several publications (see below), we did not find a satisfactory reference describing how these values were determined.

Liao, J., et al. (2011), A comparison of Arctic BrO measurements by chemical ionization mass spectrometry and long path-differential optical absorption spectroscopy, *J. Geophys. Res.*, 116, D00R02, doi:10.1029/2010JD014788.

Lehrer, E., Hönninger, G., and Platt, U.: A one dimensional model study of the mechanism of halogen liberation and vertical transport in the polar troposphere, Atmos. Chem. Phys., 4, 2427–2440, https://doi.org/10.5194/acp-4-2427-2004, 2004.

Pratt, K., Custard, K., Shepson, P. *et al.* Photochemical production of molecular bromine in Arctic surface snowpacks. *Nature Geosci* 6, 351–356 (2013). https://doi.org/10.1038/ngeo1779

The detection limits of the ozone measurements, as well as the sensitivity of the MAX-DOAS vertical profile measurements, should be briefly discussed in Sections 2.1 and 2.2, respectively.

Both Sections have been updated and now include the detection limits of the ozone sondes (2 ppb in the boundary layer), Zeppelin instrument (1 ppb), and a brief discussion about the MAX-DOAS vertical profile sensitivity.

Section 2.1: It is not entirely clear how the number of ODEs is determined, in particular for the in-situ instrument. My understanding of a single ODE is a continuous period in time during which the ozone VMR remains below a certain threshold value. Here, it is not clear whether the number ozone depletion events are counted, or rather the number of hours during which ozone VMR remains below the threshold. This needs to be clearly defined. I think it would be inappropriate to count each hour of low ozone as a single ODE.

Thank you for pointing this out. In this study, consecutive hours below the threshold are each marked and individually counted as ODE, which is not really consistent with the definition of ODE. However, to avoid introducing a new abbreviation and jumping back and forth between the new abbreviation and ODE, it has been decided to still use ODE. The following sentences have been included in Section 2.1 to avoid confusion:

We here use the name ODE although it is not quite correct in this context, since during a longer ODE, all consecutive hours below the threshold are individually marked with the abbreviation ODE, which is not consistent with the definition of ODE. However, in order to avoid introducing a new abbreviation, we kept the term ODE when referring to individual hours having ozone values below the threshold.

P5, L122: “The sensitivity to the choice of the threshold value”: Sensitivity of what?

This sentence has been extended: *The sensitivity* *of the meteorological conditions and BrO […]*

Figure 1 is very hard to read. In the left panel, it is impossible to recognize individual non-ODE profiles due to the large number of overlapping profiles, and it is hard to see any patterns in the time series shown in the right panel since the x-axis covers a large time range of more than 10 years. It is therefore impossible to recognize any seasonality. I would therefore appreciate if some other way of presenting the data could be found. For example, the vertical profiles could be shown as box-whisker-plots, and the time series could be shown as a separate figure with a larger width. For the time series, it appears that only springtime values are shown. I suppose there are also measurements during the rest of the year, and it would be nice to show all data in order to give an idea about the complete seasonality of ozone.

Following the suggestion of the reviewer, we have updated Figure 1. The data measured on Zeppelin mountain during the rest of the year has been inserted as black dots and is shown with a larger width.

Regarding the ozone sonde data, it is correct that it is not possible to read the individual profiles labeled as no ODE. The focus of this plot is on the individual vertical profiles of the ODE sondes and to have all the no ODE sondes for comparison in the background.

In order to perform a box-whisker-plot for the ODE sondes and the no ODE sondes as suggested, each ozone sonde measurement would have to be interpolated to the values of a fixed altitude grid. The information on the individual ODE profiles which we consider to be essential would be lost. We agree that the figure is busy but still think that it is the better way to provide the important information.

P8, L174: I think it is not appropriate to call the (310-500) nm channel “visible” since light below 380 nm is not visible.

We agree and changed the formulation to: [..] *near-ultraviolet and visible (310–500 nm)* [..]

P8, L198: Can you be more specific with the location of the WRF domains, e.g. by providing coordinates of the centres of the domains?

An additional Figure has been inserted, containing the location of both WRF domains.

The “composite analysis” method described in section 2.7 is difficult to understand and this section should be re-written (see also general comments). A very simple approach (anomaly = deviation from averages of maps of meteorological, chemical and sea ice parameters for ODE and non-ODE conditions) is described in a very complicated way:

- It does not become clear that the approach is applied to the spatial distribution of the observables over the entire Arctic region. During my first read, I thought this would refer to local parameters at the measurement site.
  The following sentence has been expanded:
  To investigate the anomalies of 1. meteorological conditions, 2. BrO, and 3. SIC in the Arctic region during ODEs in Ny-Ålesund, a composite analysis was conducted.
- Related to the ozone soundings, how do you define a "data point" (L216)? Is this the O3 VMR at a certain altitude (meaning that one profile consists of many data points), or a single ozone profile?
  Regarding the ozone sondes, a data point is defined as a single ozone profile. The word 'data point' as been removed, as it is misleading and a reference to Section 2.1 has been included, where the separation into ODEs an noODEs is described.
- It is not clear how you apply the threshold value to the ozone vertical profiles. Do you consider a measurement as ODE if the O3 VMR is below the threshold at some altitude, or does it need to be depleted over a certain altitude range?
  Described in Section 2.1.: All ozone sondes that contain ozone values below 15 ppb at altitudes between 0 and 2 km are marked as ODE and displayed in red.
- The number of ODEs stated in section 2.7 does not agree with the numbers in Table 2
  Changed in text to 1237 ODE and 24409 no ODE hours in the Zeppelin data set.
- How do you define an "ODE day" (L216)? An ozone depletion at the time of the balloon sounding does not necessarily mean that ozone is depleted during the entire day.
  The word 'day' is misleading and has been removed, since an ODE 'day' only covers the time of the ozone sonde measurement.
- The calculation of the anomalies 24 h and 48 h before and after the time of O3 observation is explained in a quite cumbersome way and should be rewritten.
  This part has been shortened and rewritten.

P9, L227: It is not mentioned that Y* is also calculated for the sea ice coverage parameter. It is not clear what you mean with "To obtain Y*(bar)_ODE, all selected Y* were averaged". How is this selection performed?

Y* and further the anomaly has not been calculated for sea ice, since it is not expected (and results not shown here confirmed it) that there will be much change in SIC during this time period.

With 'selected' all points in time 24 or 48 hours before/after an ODE were meant. This section has been rephrased.

P10, L260: It is not clear to me how you it can be concluded that "already ozone poor air is transported to the measurement site" if there are indications that recycling of Br_x on blowing snow took place. I think the opposite is likely as well, namely that saline particles are transported to the measurement site and ozone destruction took place all the time along the trajectory, and probably still takes place in situ.

This sentence has been adapted: *These findings indicate, that Br is likely recycled on aerosol or blowing snow on its way to Ny-Ålesund and therefore ozone is continuously depleted along the trajectory to and in Ny-Ålesund.*

P11, L262: Here you discuss an increase of the SIC on ODE days. It is hard to imagine that sea ice cover changes that rapidly, since sea ice formation is a very slow process, while ODEs occur over time scales of only a few hours.

The discussion of SIC is based on a publication from Aue et al., 2022 where they found a change in SIC in the Arctic region due to cyclones. Since we see this low pressure anomalies during ODEs, we thought that this might be also the case for ODEs. Strong winds and a cold air outbreaks could lead to fast sea ice change or formation. But as already mentioned, these anomalies might be due to seasonal effects. Camera images from Zeppelin mountain during the time of the case study showed freshly formed sea ice in the Kingsbay after the cyclone passed Ny-Ålesund.

Aue, L., Vihma, T., Uotila, P., & Rinke, A. (2022). New insights into cyclone impacts on sea ice in the Atlantic sector of the Arctic Ocean in winter. Geophysical Research Letters, 49, e2022GL100051. https://doi.org/10.1029/2022GL100051

Section 3.2.1: I suggest to skip this section as already detailed in the general comments

See answer in the general comments.

Sections 3.2.2 and 3.2.3 discuss seasonal and inter-annual variations of the anomalies. These are not sensitivity analyses, as the title of Section 3.2 suggests.

The title of Section 3.2. has been changed to: *Sensitivity and temporal analysis*

P17, L374: Please quantify the detection limit of the ozone measurements.

That has been done in the updated Section 2.1 (see 3rd comment).

Section 3.3: Here vertical profiles of ozone from balloon soundings are compared to vertical profiles of BrO from MAX-DOAS. It is speculated that blowing snow plays a role in the release of reactive bromine. To further support this hypothesis, it would be important to also show and discuss vertical profiles of the aerosol extinction, which should be available from the MAX-DOAS measurements.

We thank the reviewer for this suggestion which helped to better understand the evolution of this event! We checked the aerosol profiles and found enhanced aerosols only on April, 1st,. April 2nd and 3rd did not show any signs of enhanced aerosols in the profiles.

After April 1st, a new layer of ice seemed to form in Kingsbay when looking at the camera images from Zeppelin Mountain, which might be contributing to the ozone depletion on the second and third, instead of the blowing snow as initially assumed. Therefore, ozone depletion from local Br is likely to happen on April 2nd and 3rd. The paragraph has been rewritten accordingly.

Figure 8: BrO does not seem to be present over the entire altitude range where ozone depletion is observed. Can you elaborate on the reasons for this discrepancy?

The main reason lies in the details of the BrO retrieval, which has been modified as described below:

Figure 8 shows slightly different vertical profiles of BrO compared to the initially published manuscript as we realized that profiles with test retrieval settings have been shown rather than the commonly used settings. The new profiles are slightly lifted to higher altitudes compared to the results from the previous manuscript and a double peak appears for the first shown day. As the sensitivity for higher altitudes is limited, the exact altitude for the maximum concentration cannot be completely constrained within a MAX-DOAS profile retrieval (see also comment below). The double peak indicates that BrO can be found in higher concentrations for almost the entire altitude range where also lower ozone values can be seen. Note that it is not possible to retrieve box-like features or sharp edges from MAX-DOAS measurements with optimal estimation based inversion algorithms due to a priori smoothing effects.

The vertical sensitivity of MAX-DOAS profile retrievals is highest for lower altitudes and decreases strongly for altitudes larger than 2-3km. However, elevated trace gas layers can still be retrieved when this layer is the dominant trace gas concentration - no shielding effect of larger near surface concentrations are present.

Technical Corrections

If no further comment has been written, it should be considered as ‘done’.

P2, R1-R6: Chemical formulas should not be in italic

P4, L103: On can either discuss a case study or observe a case, but observing a case study does not make much sense (this would at the very most be meta-science).

P4, L110: I suggest to rewrite this sentence as follows: “The vertical resolved ozone sonde profiles allow to study the altitude distribution of ODEs in the boundary layer”

P4, L113: Add “described below” to the end of the sentence since the threshold values are not defined yet.

P4, L116: Insert “does” before “not necessarily”.

P5, L118: Replace “enables” with “provides”.

not changed → two times provide then in two consecutive sentences

P5, L122: It should be stated that the threshold value applies to the ozone VMR.

P5, L125: “The background level of ozone in the boundary layer is normally around 40 ppb”.

P5, L126: This sentence can be deleted since the application of the threshold value is already explained at the beginning of the paragraph.

P7, L136: Please explain the acronym/abbreviation “AWIPEV”

removed AWIPEV

P7, L140: “sun light” -> “sunlight”

P8, L194: Replace “have” with “achieve”

Section 2.7: “Time point” is not a correct English term, I suppose you mean point in time or time of measurement.

P9, L220: “where” -> “when”

P21, L439: “the same pattern” -> “similar patterns”

P21, L452: “extend” -> “extent”

References

Benavent, N., Mahajan, A. S., Li, Q., Cuevas, C. A., Schmale, J., Angot, H., Jokinen, T., Quéléver, L. L. J., Blechschmidt, A.-M., Zilker, B., Richter, A., Serna, J. A., Garcia-Nieto, D., Fernandez, R. P., Skov, H., Dumitrascu, A., Simões Pereira, P., Abrahamsson, K., Bucci, S., Duetsch, M., Stohl, A., Beck, I., Laurila, T., Blomquist, B., Howard, D., Archer, S. D., Bariteau, L., Helmig, D., Hueber, J., Jacobi, H.-W., Posman, K., Dada, L., Daellenbach, K. R., and Saiz-Lopez, A.: Substantial contribution of iodine to Arctic ozone destruction, Nature Geoscience, 15, 770–773, https://doi.org/10.1038/s41561-022-01018-w, 2022.