

We thank the reviewer for providing the helpful and critical comments and suggestions about the paper. We believe that they enhance the scientific value and accuracy of the manuscript. In the following, we discuss the questions and suggestions and provide point by point responses.

General comment

"The paper is well written overall. However, there lacks clear take home messages, i.e. how does this work significantly advance our knowledge. I would like to come away with more than the relationship is 'complex'. I would like quantification of the first year sea-ice extent and Arctic amplification on BrO explosion events as these have been nicely explored in this paper, but the clarity of conclusions drawn reduces the potential significance and usefulness of this work. Below I detail edits throughout the paper with my major concern being around the significance of this work, and how it could be used to advance model, observation comparisons going forward, so that predictions of the implications for the oxidative capacity of the Arctic (and therefore also the Antarctic) can be explored in climate simulations".

We have re-written the abstract, introduction and conclusion sections, in order to clearly state the importance of the current study and removed the general statement that the relation between tropospheric BrO VCDs and sea ice is complex. We observe changes in tropospheric BrO plumes in recent years. These changes can be linked non-linearly to changes in sea ice age and extent that are also occurring. This is now pointed out in the manuscript. We provide the correlation coefficient between first year ice extent and tropospheric BrO in the abstract, in order to clarify and quantify the relation from the beginning. Furthermore, we have added a significance test between the two quantities, in order to clarify, that although their correlation (based on the magnitude of the correlation coefficient) is moderate, it is significant. Also, we have included another subplot in Figure 13, where we show the trend of first year ice occurrences. In this way, our conclusions on the impact of first year ice on the recent increased formations of tropospheric BrO plumes are more solid. In the conclusion, we have erased the repetitions and kept only the take-home messages: The 1.5% increase of tropospheric BrO during polar springs and its moderate correlation of 0.32 to the first year ice extent evolution. Also, we discuss potential future significance and usage of work. The dataset that we retrieved can be integrated in chemical transport models, in order to be used as validation of simulations on the impact of bromine explosions on O₃ loss and potential OH changes.

With respect to your detailed edits, we have followed the specific comments (indicated below):

Specific comments

Page 1, line 12: Sentence needs revising: Every polar spring BrO explosions occur, which are a series of chemical reactions that release bromine molecules to the troposphere over sea ice covered regions.

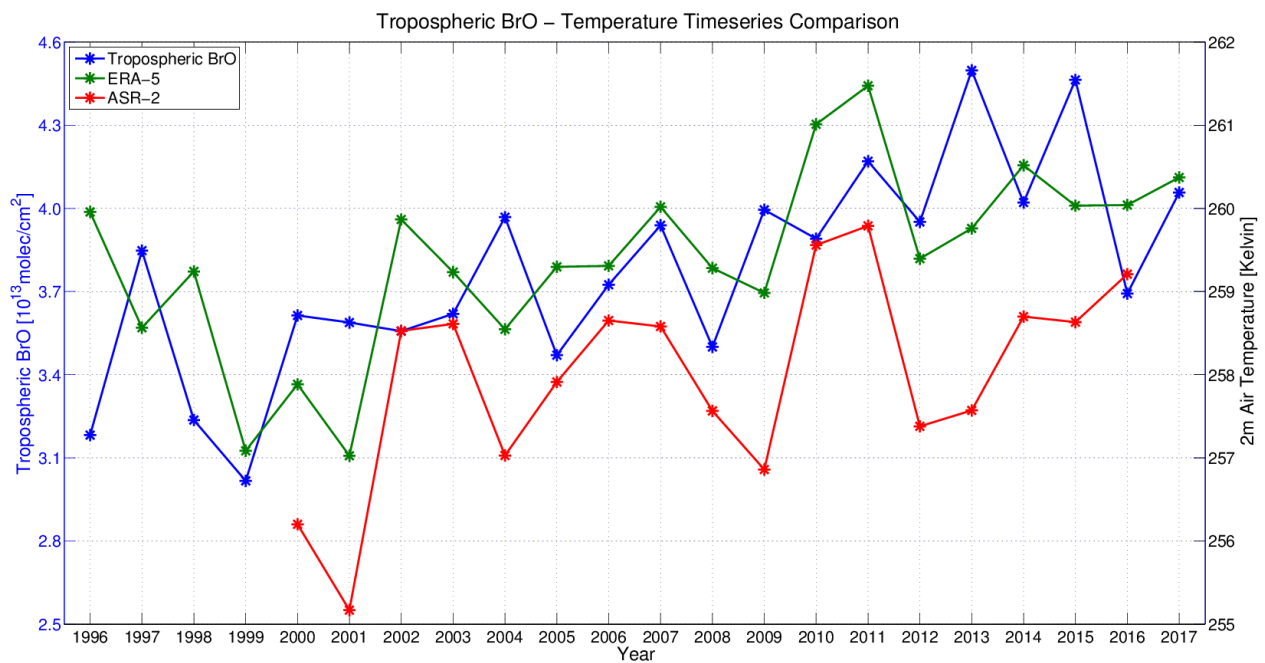
Thank you for pointing to this sentence. We rephrased the sentence to “. Every polar spring, phenomena called bromine explosions occur over sea ice. These bromine explosions comprise photochemical heterogeneous chain reactions that release bromine molecules, Br₂, to the troposphere and lead to tropospheric plumes of bromine monoxide, BrO.”

Page 1, line 16: Name the four satellite experiments

Added (GOME, SCIAMACHY, GOME-2A and GOME-2B)

Page 1, line 20: and elsewhere: 1.5% per year – since there is a focus in the abstract on Arctic Amplification – could this response be expressed also per degree of warming experienced?

This is an interesting suggestion. However, the non-linear connection of tropospheric BrO plumes with their driving mechanisms (as also indicated in the paper, the comparisons of tropospheric BrO and sea ice age can be seen in some cases, but not in all), and the deep interactions between the mechanisms themselves, make the specification of the effect of the warming of the air temperature on bromine explosions and the trends appearing in the time-series a difficult task. We therefore prefer not to add a statement such as “BrO increased by 5% per K warming” as this would suggest a linear relationship, which in our opinion does not exist. However, we are attaching here the time-series of tropospheric BrO and 2m Arctic air temperature, from 2 reanalysis datasets (ECMWF ERA-5 (Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., et al. (2020). The ERA5 global reanalysis. Quarterly Journal of the Royal Meteorological Society, 146(730), 1999–2049. <https://doi.org/10.1002/qj.3803>), downloaded from the following website: <https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5>, and WRF Arctic System Reanalysis version 2 (Bromwich, D. H., Wilson, A. B., Bai, L., Liu, Z., Barlage, M., Shih, C.-F., et al. (2018). The Arctic System Reanalysis, Version 2. Bulletin of the American Meteorological Society, 99(4), 805–828. <https://doi.org/10.1175/BAMS-D-16-0215.1>), downloaded from: <https://rda.ucar.edu/datasets/ds631.1/>).



Page 1, line 22: Specify how linked (stating that it is complex is very general)

We have rephrased the text, stating that the link (based on the magnitude of the correlation coefficient) is moderate. “We infer from comparisons and correlations with sea ice age data that the reported changes in the extent and magnitude of tropospheric BrO VCDs are moderately related to the increase of first-year ice extent in the Arctic north of 70° N, with a correlation coefficient of 0.32.”.

Page 1, line 24: Arctic temperature please qualify as surface air or sea-surface temperature?

Changed it to: “surface air temperature”

Page 2, line 6: here and elsewhere please remove hyphen in ecosystem

Removed it throughout the text

Page 2, line 11: 1990 and 1997 don't represent 3 decades worth of studies, a 2010s review paper reference more appropriate here

Added a reference by Saiz-Lopez and von Glasow, 2012

Page 2, line 14: insert but : : : OH are reduced, but the reactions of: : :

Inserted as suggested

Page 2, line 17: the “Bromine explosion” closing speech mark needs format correction

Corrected

Page 2, line 20: citing a 2009 paper when discussing controversy is problematic, as modelling efforts recently have integrated simple and effective parameterisations, based on both frost flower and blowing snow mechanisms i.e. (Falk & Sinnhuber, 2018) or some other more recent reference would be appropriate here to discuss exactly what controversy still exists.

The exact level of impact of each parameter (such as blowing snow, wind speeds, air temperature) on the formation of enhanced BrO plumes is still unknown. This is what we meant with controversy. Added citation to Falk & Sinnhuber (2018), Huang et al. (2020), Seo et al. (2019). We have rephrased the text to: “Although there are studies which try to model BrO plumes from their driving mechanisms (Falk and Sinnhuber, 2018; Seo et al., 2019b; Huang et al., 2020), the exact level of impact of each parameter on the formation of enhanced tropospheric BrO is uncertain.

RI – hv should be hν, A"oi~A ~ ai~A 'lthese should be defined in the text).

We are sorry but we could not figure out exactly what the reviewer wrote. However, we have defined hv as solar radiation in the text. All the other species in the chemical reactions are described in the section below the reactions.

Page 3 line 7: bromine atoms rapidly remove (switch from remove rapidly)

Rephrased as suggested

Page 3 line 20: it's = its

Corrected

Page 3, line 23: favor bromine explosion conditions. Again, there is more recent (and quantitative) modelling efforts.

Here we are mostly presenting studies which indicate the relation of tropospheric BrO to driving mechanisms, not modeling efforts. We added citations from Yang et al, (2020) and Fernandez et al, (2019), regarding recent efforts on BrO modeling.

Page 3, line 17: remove second comma

Removed (Page 4, line 17)

Page 5, line 23: on – was the data from Metop-C useable in theory, if so why wasn't it used in your study?

Metop-C was launched in autumn 2018, but the first data were received during spring of 2019. It therefore does not contribute to the investigated time period (1996 to 2017).

Page 6, Table 1: in GOME-2A line 40 x 40 (remove capitalisation from X)

Removed

Page 6, line 9: and elsewhere – Sun should be capitalised throughout

Changed throughout the paper

Page 9, line 5: insert comma: autumn, when the solar: : :

Inserted

Page 11, line 2 (whole paragraph needs reworking): For the NO₂ and O₃ column satellite retrievals, the tropopause height : : : is used: : .

This has been rephrased to “NO₂ and O₃ columns from satellite retrievals and tropopause height from meteorological reanalysis data are used for extracting the tropospheric BrO component from the retrieval (stratospheric separation). Sea ice data (age and type) obtained by satellite remote sensing was used in order to identify regions with sea ice cover and hence high surface reflectivity, which is required for the retrieval of tropospheric BrO in this study and for data interpretation in relation to bromine sources.”

Page 11 Line 4: reflectivity, which is required: : :

Corrected

Page 13, line 11: change the degree sign from a zero to a circle

Changed throughout the whole text

Figure 5: – please show 1 sigma errors in the VCD retrievals as well as interannual variability with error bars on these plots in order for us to be able to determine agreement significance.

The purpose of Figure 5 is to provide seasonal cycles for geometric, stratospheric and tropospheric BrO time-series, for each instrument. For each sensor we averaged the data over the individual months of the corresponding operation period. Therefore, and since we can see from Figure 3 that for some years we have higher columns than for others (e.g. 2013 to 2017, the GOME-2B operation period compared to 1996 to 2003, the GOME operating period), the agreement between the sensors is hence not the focus of Figure 5 and differences because of the different averaging time periods are expected.

Section 4.2: by selecting only for high BrO over sea-ice areas you are only capturing the genesis and not necessarily determining the implications of the combined oxidative capacity and changes in cyclonic activity. Much of the interesting implications of warming and BrO will happen outside of areas which are over sea-ice. I'm unclear why the only BrO over sea-ice was a necessary criteria?

We chose to work exclusively on sea ice covered regions because the tropospheric air mass factor that we use in the computation of tropospheric columns considers a surface reflectivity of 0.9. As a result, our tropospheric columns can be considered accurate only above bright surfaces. Over dark surfaces such as the ocean, the sensitivity of the satellite measurements to boundary-layer BrO is unfortunately low.

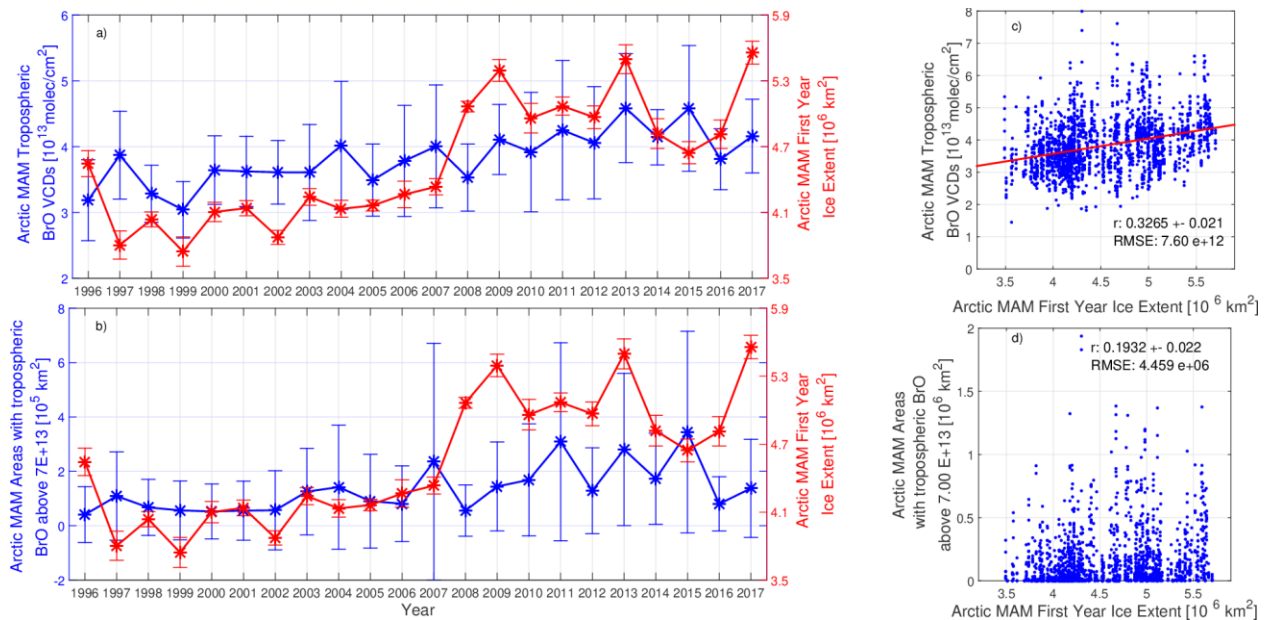
Figure 9: – please try to get onto one page, use horizontal colorbars perhaps?

Figure 9 has been fitted to one page.

Page 22, line 16. And Figure 11: Stating that the relationship between sea-ice area and BrO explosion is complex is unsubstantiated by a useful plot. I would like to see the first year sea-ice area versus average BrO column plot (or BrO explosion area). Figure 11d goes some of the way to providing this but I suggest that an annual number be provided i.e. those produced in 11 a and b (with variability range errorbars for both axes) could be more useful than using every data point above the threshold as is done currently. Figure 11d trend line is arbitrary and should be removed (unless the annual number provides support for a linear relationship). Some metric representation of annual cyclonic activity or amplification amplitude would be valuable too on this plot – i.e. providing an annual cyclonic index in color and some index for Arctic

amplification temperature with size may help to disentangle the story for us. This way we would be able to determine whether a parameterization that would be useful to test models could be derived for this 'complex' relationship.

Removed "complex" in this line and throughout the text (see reply above), being more precise on describing the relationship between tropospheric BrO and sea ice age (for example, adding a sensitivity test). Figure 11a shows the first year sea-ice versus average BrO columns plot (as you suggested). No threshold is applied in this sub-plot. Also, we have removed the trend line from Figure 11d. We have added 1σ uncertainties for the correlation coefficients of the two scatter plots. We calculated the correlation coefficients for the annual time-series of tropospheric BrO and first year ice as well. However, we believe that the ones from the daily scatter plots (as shown in the manuscript) are more informative and describe the relationship between the two quantities more accurately. We have added the variability range error bars for both axes and show the Figure here:



Page 25, line 4: VD -> VCD.

Changed

Figure 11: a-d labels missing.

Added the labels

Page 27, figure 12: How can trends over 1 year be considered significant or reliable with only a few points? I assume the annual cycle is removed for this plot (otherwise like a sine curve, you would get a trend just due to that). I'm really unclear about what figure 12 is showing, given it only is discussed briefly and doesn't add to the papers aims/conclusions. As the minimum time to detect a trend is not provided, I think this plot and the discussion can be removed.

Figure 12 provides information on the variability of the trend of tropospheric BrO VCDs for different starting years (x-axis) and ending years (y-axis). As the value of the calculated trend depends on the averaging time period, the Figure provides a visualization of the consistency of the observed BrO increases and how they changed over time. For almost all time periods over 5 years and more, positive trends occur. As the reviewer correctly pointed out, trends over very short time periods are less significant. The annual cycle for the trends is removed by starting and ending at the same time of the year (i.e. end of March). However, as the reviewer pointed out, trends over short periods are dominated by inter-annual variability.

Figure 13: over what time period is the trend calculated? Provide this in the caption.

Added. The trends are calculated over a 22 year period (1996 to 2017).

Page 29 line 5: the BrO explosion -> Bro explosion events.

Changed as suggested

Page 29: when discussing trends state over what period they are determined.

The information on the time period has been added.