We would like to thank the reviewer for providing helpful comments and suggestions for improvement of the paper. We believe that they enhance the scientific value of our contribution. In the following, we discuss the questions and suggestions and give a point by point response.

General comments

A central hypothesis of this study is that the transition from multi-year to first year sea ice in the Arctic ocean affects the distribution, number and intensity of bromine explosion events and thus Arctic tropospheric BrO. The potential mechanism(s) behind should be made a bit clearer in the Introduction and the discussion of results should be made under these assumptions. To be more specific: Multi-year ice that has survived at least one melt cycle has less bulk salinity compared to first year sea ice. The increase in first-year sea ice could potentially favour bromine explosion events by providing more sea salt bromine.

To test this hypothesis, relating first year sea ice area with (excess) tropospheric BrO, as done in Fig. 11 is very useful and the implications of this relation should be stated accordingly. The correlation between first year sea ice area and BrO seems to be significant (by looking at Fig.11, I couldn't find a statistical test for significance) although of course not perfect. How much this relation explains (in terms of variance, trend, shift in geographic distribution) should be stated clearly, together with its limitations. Phrases like "linked in a complex way" (end of abstract and in the Conclusions) without explaining what the "complex way" is hide more than they reveal.

There may be other factors how sea ice age impact on tropospheric BrO e.g. through a reduction in sea ice thickness. So while for the point above the distinction between first year and multi-year ice is most important (and the distinction between say 5 or 6 year old ice is not so important), for other factors the actual age may be critical. Please be specific in the discussion and in the presentation. Example 1: the colour scale in Fig. 9 makes it difficult to clearly identify if the majority of the ice in recent years is first year ice or 2 year old ice. Example 2: the age trend in Fig. 13: Even a relatively small trend may have resulted in a shift from multi-year to first year ice over the years. Try to be specific in the discussion of the implications.

Currently, although the Arctic sea ice extent is decreasing, the more saline first year ice grows at the expense of multiyear ice. This is affecting the number and magnitude of bromine explosions occurring. The criticism is that there is no clear conclusion on the relationship between tropospheric BrO and first year ice conditions. In response your comment, we have rephrased parts of the abstract, the introduction, the main text and the summary, (e.g. the moderate correlation coefficient of 0.32 between the two quantities during polar springs is now given on the abstract). With the help of maps in Fig. 9 and time-series in Fig. 11, we have pointed out that changing sea ice conditions do not explain all the changes of the observed tropospheric BrO. This is because the appearance of bromine explosions depends on other parameters as well (air temperature, wind speeds, cyclone activity).

We have performed a hypothesis test of the significance of the correlation coefficient as you suggested, based on the null hypothesis that there is no correlation between tropospheric BrO

and first year ice. The null hypothesis was rejected, meaning that the correlation between tropospheric BrO and first year ice is significant. The color scale in Fig. 9 was optimized in a way that first year ice is distinguishable from multiyear ice. Values from 1 to 2 are colored blue, while multi-year ice (2 years and above) is denoted with green and other colors. So everything blue in the maps of Figure 9 means that there was at least one first year ice grid box during the polar spring average. Also, in Fig. 13, we tried to explain better the point, which you have made by adding another sub-plot in the figure, which shows the trend of the occurrences of first year ice. We infer from it that the trend of occurrences of first year ice has increased in some areas where also an increase of tropospheric BrO occurred (i.e. east of Greenland), but also decreased in some others (i.e. in the Canadian Archipelago, where BrO also increased). In summary, we explain that sea ice age influences tropospheric BrO but, given that the correlations between first year ice and daily tropospheric BrO are moderate, does not fully account for the changes of the tropospheric BrO column dataset.

Section 3.1 contains a general description of the DOAS method used to retrieve the BrO SCDs. I am not sure if a general description of the DOAS method is needed here, but in its current form there are too many mistakes and omissions to make it useful (specific points below). Please carefully check.

With respect to your comments for the DOAS section, we have followed the specific comments (indicated below):

Specific comments

P1,L11: First sentence of abstract is a bit disconnected here. Would be better in the Introduction. More generally I feel that at many places (in the abstract and elsewhere in the manuscript) "Arctic Amplification" could better be replaced by "Arctic warming" because what matters in this context is the warming, not so much the amplification (although this may be seen as pettifoggery).

We removed the first sentence. We have also replaced in all places (except in the introduction where we describe the term “Arctic Amplification”) the term “Arctic Amplification” with “Arctic warming”.

P1, L15: “e.g. Hg”: any other metals?

Bromine molecules can also react with lead, forming $PbBr_2$. However $PbBr_2$ is insoluble.

P1, L15: “22 year”: suggestion: name range of years already here

Rephrased to: “22 year (1996 to 2017)”

P1,L19/20: “magnitude of BrO: : :of about 1.5%/year”: what exactly increases with 1.5%/year? Tropospheric columns? Please be specific.

Rephrased to: “We determined an increasing trend of about 1.5% of the tropospheric BrO VCDs per year during polar spring”.

P1, L25: It is true that the understanding of Arctic Amplification is inadequate, but a few citations on Arctic Amplification may be useful. E.g. Pithan and Mauritsen, Nature Geosc., 2014.

Added two more references (Pithan and Mauritsen, 2014, Stjern et al, 2019), which discuss the response of Arctic Amplification to individual climate drivers and to temperature feedbacks.

P1, L24/25: "loss of sea ice" and "reduction of ice extent" are of course not independent. Maybe say loss of ice resulting in reduction of ice extent, thickness and reduced fraction of multi-year ice?

Corrected the sentence to: “… are the loss of ice, resulting in reduction of ice extent, thickness and a reduced fraction of multi-year ice (Stroeve et al., 2012) and the increasing rate of loss of the Greenland ice cap (Mouginot et al., 2019)”.

P2, L10: “over 30 years ago”

Added

P2, L12: introduce “O3” when first used as “ozone (O3)”

Done

P2, L13: I think this point should be made a bit clearer: O3 and OH are decreasing, but bromine radicals instead could act as oxidising agents. Are there references how the oxidising capacity overall changes?

Indeed, on short timescales, bromine radicals can contribute to the formation of OH. However, on longer timescales, the depletion of ozone caused by bromine compounds reduces the production of OH. Added a reference (Stone et al, 2018), which suggests that the long-term effect of bromine radicals prevails. Also, added one reference on the global tropospheric OH distribution (Lelieveld et al., 2016), which suggests that the secondary sources of OH (i.e. recycling in radical reaction chains), play a more important role in global OH distribution.

P2, L21: wicket sentence

Rephrased to: “However, there is the general consensus that the potential sources of BrO plumes are (a) rich in sea salts and relatively cold (conditions occurring in potential frost flowers regions; Rankin et al., 2002; Kaleschke et al., 2004; Sander et al., 2006), (b) surfaces covered with liquid or frozen brine (Sander et al., 2006), (c) associated with blowing snow (Yang et al., 2008; Blechschmidt et al., 2016; Frey et al., 2019), (d) surface snow packs (Pratt et al., 2013; Peterson et al., 2018) and young salty sea ice regions (Wagner et al., 2001; Simpson et al., 2007; Peterson et al., 2016).”

P3, R4,R8: should that be "->" instead of "="?

Changed to ⇌

P3, L19: Closing bracket missing.

Added

P3, L19: This idea needs a bit more explanation: Transport of BrO plumes over large distance by deposition and reactivation due to release from snow pack and blowing snow?

Rephrased to: "It was shown that BrO plumes can be transported far from their initial formation areas, as high wind speeds associated with cyclones (Begoin et al., 2010; Zhao et al., 2015; Blechschmidt et al., 2016) can transfer them together with blowing snow (Giordano et al., 2018)".

P3, L24: "Polar Regions" -> "polar regions" (at many places in the manuscript)

Replaced by "polar regions" as suggested

P3, L24: "hostile" for what or whom? In spite of difficulties numerous studies have performed in-situ measurements or ground-based DOAS measurements in the Arctic. Satellite measurements are not "unique".

Changed the sentences containing the problematic words to: "The polar regions are some of the most remote places on the planet. Consequently, satellite remote sensing is a suitable method to study bromine chemistry in the Arctic."

P3, L33: It is good to provide context by citing previous studies, but better cite what has been learned rather only what has been done.

We have rephrased the whole section, now mentioning also the findings of each publication: "The relationship between BrO release and young sea ice was also discussed (Wagner et al., 2001), where it was indicated that large BrO concentrations are found over or near sea ice on the Caspian Sea. Van Roozendael et al. (2004) compared SCIAMACHY observations of Arctic BrO to GOME data, showing satisfactory agreement between the two different sensors. Theys et al. (2011) compared tropospheric BrO columns derived from GOME-2A to a chemical transport model, showing consistency with the release mechanisms of bromine. Sihler et al. (2012) compared GOME-2 BrO columns to ground-based measurements in the Arctic, demonstrating good agreement between the retrievals. Seo et al. (2019a) presented the first BrO retrievals from the TROPOMI instrument, showing high-resolution bromine explosion cases with low fitting errors".

P4, L2: "them" = "BrO explosion events" ?

Replaced as suggested

P4, L10: at some stage you should mention that first year sea ice is more saline than multi-year ice

Rephrased the sentence to: “Changes in meteorological parameters, e.g. increasing air temperature (Serreze and Barry, 2011), decreasing mean sea level pressure over northeastern America and increasing pressure over Eurasia (Ogawa et al., 2018; McCusker et al., 2016), increase in cyclone frequency and intensity (Akperov et al., 2019), stronger surface winds (Mioduszewski et al., 2018) and changes in sea ice conditions (e.g. reduced sea ice extent; Stroeve et al., 2012), increased first year sea ice fraction (and consequently salinity), and therefore decreased sea ice thickness (Richter-Menge et al., 2017) occur due to Arctic warming.”. Also included one sentence in the introduction: “The reduced multi-year ice is being replaced by first year ice, which is in addition more saline. (Galley et al., 2016)” (P1, L30).

P4, L26: remove word “results”

Removed

P5, L9: This statement is too general. There were other satellite instruments before GOME, depending on what you mean by "many" and "key trace gases"

Rephrased the sentence to: “GOME was the first satellite instrument, which was able to measure key tropospheric gases which have weaker absorption lines than ozone: examples are NO2, BrO, HCHO and SO2”.

P5, L16: what is the difference between "near IR" and "short wave IR"?

Although the spectral limits of the terms near IR and short wave IR are not SI standardized, near-infrared radiation usually implies a spectral wavelength region from 0.75 to 1.4 μm, while short-wavelength infrared implies the spectral region from 1.4 to 3 μm. Shortwave IR is sometimes defined as 0.76 to 2 μm. The sentence has been changed to: “... allowing the observation of many trace gases in the near infrared (0.75 to 1.4 μm) and short wave infrared (1.4 to 3 μm) spectral wavelength regions”.

P6, L5: throughput mentioned twice

Corrected

P6, L9: This sentence seems odd. "Long changing polymers" -> “long chained polymers"?

Changed to “long chain polymers”

P7, Eq.1: In eq.(1) the concentration of the gas j is missing.

We define the concentration in this study as the number density of the trace gas j with concentration ρ (in molec cm^{-3}). This, when multiplied by the length of the light path s (in

meters), gives the column amount or column (in molec cm^{-2}) of the trace gas j. We have rewritten Equation 1 as follows:

$$I = I_o\, e^{-\int \sum_{j=1}^{J} \{\sigma_j(\lambda)\rho_j\} ds}$$

Also, we have rephrased the text below to: "where I is the measured intensity of the electromagnetic radiation, I_o is the initial intensity, J is the total number of absorbing trace gases, j denotes a particular trace gas (e.g. BrO), $\sigma(\lambda)$ is the cross section of the absorber at wavelength λ, ρ the concentration of the trace gas".

P7, L14: really ABSORPTION cross sections or SCATTERING cross sections?

Changed to scattering cross sections

P7, L31: is a "four degree" polynomial a polynomial of fourth-order, i.e. with five degrees of freedom? Please be specific and consistent to avoid confusion.

Changed to fourth-order polynomial

P8, Eq.5: There is something wrong with eq. (5). The sum should be under the root. And not on the LHS.

Corrected

P8, L16: Latitudes and longitudes need to be specified more consistently. E.g. "- 180_E" should be "180_W" and "-50.0_S" should be "50_S". If you use east and west longitudes please avoid "235_E to 270_E" and use instead "90_W to 125_W".

Changed to "the Arctic from 70.0^o N to 85.0^o N latitude, 180^o W to 180^o E longitudes) and for the Pacific reference region (50.0^o S to 10.0^o N latitude and 90.0^o W to 125.0^o W longitude).".

P10, L1: Please mention this drift already in the instrument overview in Section 2.

Moved to section 2.4

P11, L2: This first sentence does not make much sense and is redundant.

The sentence has been changed to: "NO_2 and O_3 columns from satellite retrievals and tropopause height from meteorological reanalysis data are used for deriving the tropospheric BrO component from the retrieval (stratospheric separation)."

P13, L13: what is the meaning of the word "high" here?

Removed from the sentence

P13, L15: you can remove "due to Arctic Amplification" here. See also my general comment on Arctic Amplification.

Removed

P14, L10: There are also real stratospheric BrO trends due to changes in anthropogenic emissions!

The sentence has been changed to: "The stratospheric BrO VCDs show a small upward trend from 1995 to 2001 and a slight decrease afterwards, which are in agreement with measurements of stratospheric BrO from Harestua station (Hendrick et al., 2008)."

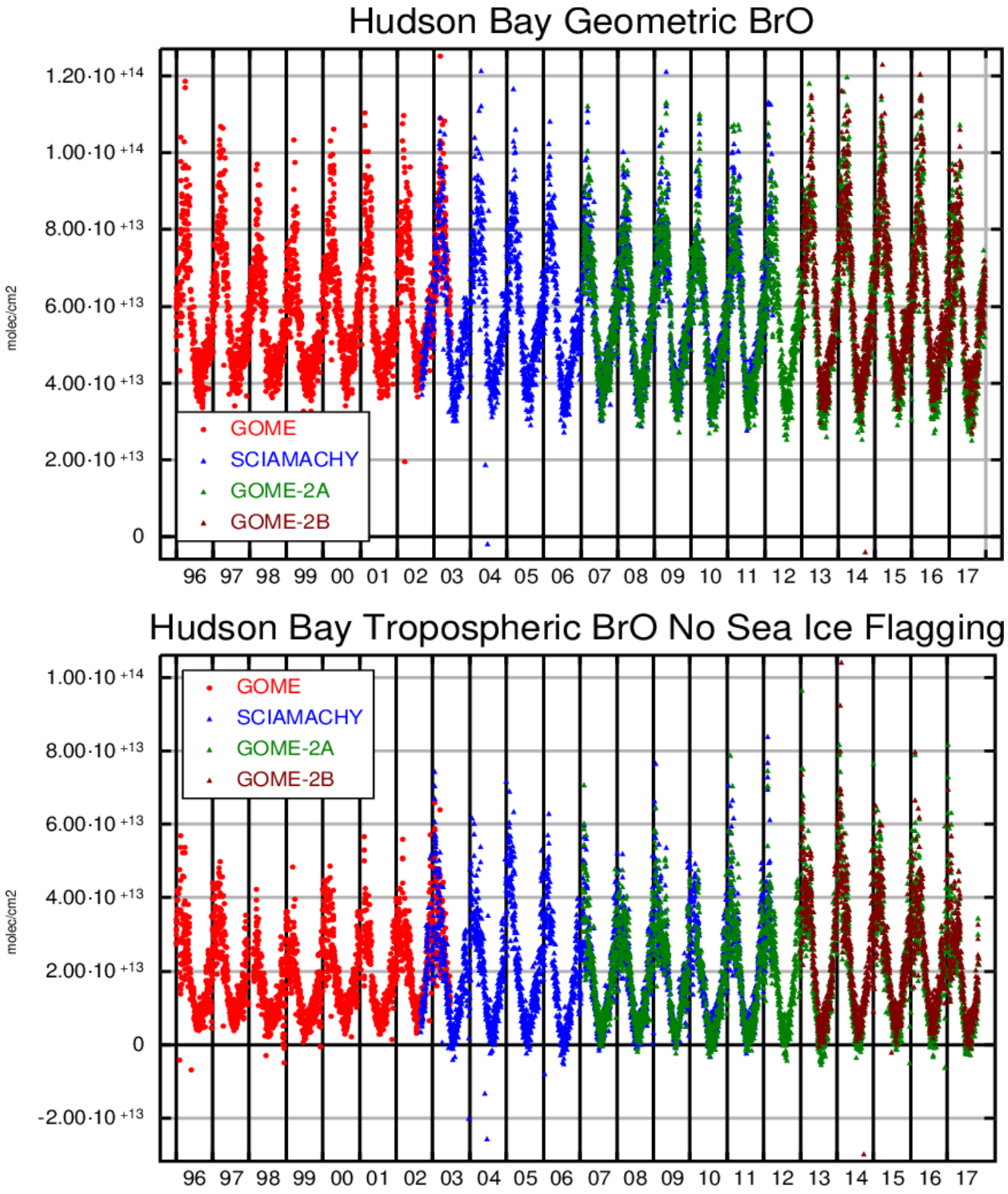
P15, L1: I don't think you need to explain that temperatures in summer are higher because of increased solar insolation (it does not matter in this context), but you could specify in which month the maximum temperature and in which month the sea ice minimum are reached.
Changed to: "During July and August, the temperature reaches its maximum. In September, the minimum sea ice extent is observed."

P15, L7: Looking at the spatial distribution may tell if there are (few and/or small) areas of bromine explosion in September or only a gradual increase in the background. Would it make sense to include somewhere also maps with BrO in autumn?

During September, the sea ice coverage is very low, so it would not make sense to include September maps of tropospheric BrO in the manuscript. However, we have checked September maps of tropospheric BrO and concluded that the small increase observed is mainly a background increase, as we cannot see clearly localized BrO explosion events.

P22, L4: "in the early years, most of the BrO is found in the region of the Barents and Kara Sea": I am confused. Richter et al. (1998) show largest BrO enhancements in March 1997 over the Canadian Archipelago, Hudson Bay and north-west of Greenland.

Richter et al showed specific days of total BrO vertical column densities. Although the Hudson Bay is a well known BrO hotspot, we decided to perform our analysis only over latitudes > 70^0 N, as the proportion of sea ice covered areas and the fraction of pixels for which tropospheric BrO VCDs can be retrieved is larger there. Retrieved tropospheric BrO columns are more accurate for latitudes > 70^0 N, as the tropospheric air mass factor that we use considers a 0.9 surface reflectivity.
We have analyzed retrievals over Hudson Bay, and we are attaching figures of geometric and tropospheric BrO VCDs (similar to Figure 3 of the paper, with a land mask applied):



We see a less pronounced trend for BrO VCDs over the Hudson Bay, in comparison with the Arctic region, as defined in the manuscript (latitudes > 70^{o})

P24, Fig11: I don't fully understand what are the points shown in the right hand side panels? Related to this: what is the correlation coefficients for the data shown in the time series on the

left hand side? The same as given in the right hand side plots? Did you check if the correlations are significant? (By eye, the correlation between Arctic MAM mean BrO and 1st year ice extent in Fig. 11a seems significant.)

Every point in the right panel scatter plots represents one polar spring day (the average tropospheric BrO VCD for this day is plotted against first year sea ice extent in the top right panel, the area with tropospheric BrO VCD above the threshold is plotted against first year sea ice extent in the bottom right). The time-series on the left side are the polar spring averages of the scatter plots. The correlation is positive but moderate for the top right scatter plot. We have performed a hypothesis test to verify the significance of the correlation coefficient and found that the correlation is significant (p value lower than 0.05). We added this information to the text of the paper. Also, we have calculated the correlation coefficient of the annual time-series (the blue and red curve shown in the left sub-figures). The correlation coefficient between polar spring averages of tropospheric BrO and first year ice extent corresponding to the top left sub-figure is 0.62, while the correlation between areas of polar spring averages of tropospheric BrO VCDs > 7 x 10^{13} molec cm^{-2} and first year ice is 0.46. However, we believe that the correlation coefficients obtained from the daily scatter plots are more meaningful and represent the actual relationship of tropospheric BrO and first year ice extent more accurately.

P24, L11: the phrase "fresh" ice is misleading, as first year ice has a higher salinity, i.e.is not "fresh"

Changed to “and more first or young sea ice is formed each winter period”

P24, L15: What is the correlation coefficient? (See comment above.) A rigorous statistical analysis is more useful than the “anecdotal evidence” given in the following sentences.

The correlation coefficients for the two scatter plots are presented inside the plots (denoted with r). The results of the significance test has been added to the text.

P25, L4-7: I find these sentences confusing. My impression from Fig. 11a is that there is a positive correlation between first year sea ice and BrO, although this is clearly not the only factor. But then you say this agrees with Choi et al. who found even a negative correlation? I would say this is in contrast to Choi et al. and part of this difference may be attributed to a possible degradation of the OMI instrument?

Although the correlation between tropospheric BrO and first year ice is positive from the scatter plot, we see that it is not strong, as many days with high tropospheric BrO columns occur with moderate first year ice extent (around 4.5x10^{6} km^{2}). From the time-series, we see in many years an opposite evolution (one quantity increases, while the other decreases, for example 2008 and 2015). The text was rephrased, especially in the section where we compare to Choi et al, to: “This finding is in contrast to the results by Choi et al. (2018), where an analysis of BrO VCD retrieved from OMI was performed. They found a correlation coefficient of -0.32 between first year ice extent and tropospheric bromine explosion frequency. The negative sign is attributed to a decrease of tropospheric BrO VCDs over the latter years of the trend analysis. The differences

to Choi et al. (2018) may be explained by a degradation of the OMI instrument (Kroon et al., 2011).".

P25, L22: Please specify the value of the autocorrelation used. What exactly is the meaning of the "period M"? Generally I am not convinced that the trend model with harmonics is an ideal choice as you don't have data during winter. For calculating trends in individual months as in Table 4 the harmonics are not needed at all.

An autocorrelation value of 0.2 was used. The period M is the 22 years of the entire dataset. The reason why we used this approach is that, although the winter columns are missing, we still see a profound seasonality in our dataset. However, when we calculated the monthly trends in table 4, this approach was not used (added in the text).

P25, Table 4: It is a bit unusual to have the units in a column, instead of in the header.

Added the unit information to the title and in the header of the figure

P28, L10: First paragraph of Summary is redundant, largely repeats introduction

Rephrased – removed most parts of the first paragraph

P29, L7: did you show that the correlation in Fig. 11a is not significant?

We have performed a hypothesis test on the significance of the correlation coefficient. The correlation is significant and we have added it in the text.

P29, L18: This statement on temperatures in 2016 is too vague. You could mention here that you have not considered changes in temperature and this may be another factor affecting tropospheric BrO to be investigated in future studies.

Rephrased to "The appearance of plumes of tropospheric BrO VCD and their intensity are influenced by several meteorological drivers (air temperature, sea level pressure, wind speeds and cyclones) and the amounts of blowing snow (Blechschmidt et al., 2016; Seo et al., 2019b). Further investigations are required to understand the evolution of tropospheric BrO and its dependence on these drivers of tropospheric BrO release."

P30, Author contributions: Sounds a bit strange that not all authors are named for their contributions here (not even for "providing insight and knowledge").

Rephrased to: "I. Bougoudis undertook the retrieval of BrO from the different satellite instruments, collected and processed the sea ice age data, performed the analysis and prepared the paper. This study was initiated by J.P. Burrows and A.-M. Blechschmidt. The research presented was supervised by A.-M. Blechschmidt, A. Richter and J.P. Burrows. A.-M. Blechschmidt and N. Theys provided the stratospheric separation. A.-M. Blechschmidt, A. Richter, S. Seo, N. Theys and J.P. Burrows provided input with respect to BrO issues of relevance. A. Richter developed software which was used for processing and analysing the BrO

data. A. Rinke provided input on sea ice and trend analyses. All authors contributed to the writing of the paper.”.

P31: Barrie and Platt listed twice

Corrected

P32: Claas listed twice.

Corrected.

P32, L23: something is missing here

Inserted the missing reference

P33: Fickert listed twice

Corrected