Response to anonymous referee #2

We thank the reviewer for his/her constructive comments and suggestions, which helped us to improve our manuscript. We have addressed the questions as follows:

General comments:

This manuscript reports a statistic analysis of polar total column BrO and its corresponding meteorological parameters (including temperature, surface pressure, wind seed, direction and tropopause height), using 10-year GOME-2 BrO and meteorological data. A further Spearman rank correlation analysis was applied to assess the dependence of total column BrO on various meteorological parameters. Some interesting results regarding the spatial distribution of enhanced BrO and its relation to the above meteorological parameters are reported which is welcome. The authors also attempted to link the relationship to relevant physical/chemical mechanism(s) being proposed previously. This study indeed adds some new knowledge to help a better understanding of the underlying processes involved. However, I found their attempt to evaluate some relevant mechanisms is dangerous and inappropriate. This is because the bromine source is a complex function of multiple (rather than one) factors and the relations between them are largely non-linear, rather than linear. For example, the SSA production from blowing snow is a complex function of wind speed, temperature and relative humidity (Yang et al., 2019), more specifically, the SSA production (and bromine flux) is actually proportional to the sublimation flux of blowing snow, rather than individual parameters (winds, temperature and RH). Secondly, these meteorological parameters examined are not independent, instead they are deeply correlated (also pointed by the authors). For example, they are deeply correlated during a storm system. For this reason, I suggest the authors add a table to show the cross-correlation among these meteorological parameters during enhanced BrO cases and discuss their implications to deriving your conclusions. Thirdly, it is not correct to assume that a higher correlation coefficient between two factors than others necessary means this link is stronger than other links. Unfortunately, this assumption seems being used in this manuscript to make some conclusions. For the above reasons, I suggest a major revision before recommend it to publish in ACP.

We agree with the reviewer's comments that the mechanisms associated with bromine release are a function of multiple factors and that the meteorological factors examined in this study are not independent but correlated. To provide additional information on this, cross-correlations between the relevant meteorological parameters during the enhancement of total BrO VCD have been investigated and analyzed. The results were added in section 4.3 of the revised manuscript as suggested. Also, time-lagging effects on the relationship between meteorological parameters and total BrO VCD have been investigated. These additional test results and their analysis are described in section 4.3 (P16 L514) as follows (blue text):

"The relationship between individual meteorological parameters and the total BrO vertical column investigated above illustrates how each meteorological parameter is linked to BrO variations in terms of temporal and spatial distribution. However, since meteorological parameters are not independent of each other and vary systematically in general, cross-relationships between meteorological parameters affecting directly or indirectly BrO variations should also be considered. For example, Yang et al. (2019) showed that the sea salt aerosol (SSA) production affecting the enhancement of BrO at the tropospheric level is proportional to the sublimation flux of blowing snow which is affected by various meteorological parameters including surface wind speed, temperature and relative humidity. Also, Zhao et al. (2015) and Blechschmidt et al. (2016) showed that large-scale enhanced BrO plumes over the Beaufort Sea are associated with weather systems which change the various relevant meteorological parameters together. They also demonstrated that the size and lifetime of BrO plumes depend on the development stage of the weather system. Therefore, cross-correlations between meteorological parameters for those data having enhanced total BrO columns were investigated (see Table 4). During the occurrence of enhanced total BrO, sea level pressure has a negative correlation with surface level temperature and wind speed, while it has a positive correlation with tropopause height. For example, the development of a low pressure system during the enhancement of BrO columns may correlate with a decrease in tropopause height as well as an increases in surface level air temperature and wind speed. Although the correlation coefficients found are not large, results show that sea level pressure is linked with both surface level meteorological conditions and the tropopause height which can account for stratospheric dynamics. Indeed, pressure systems which usually evolve due to interactions of temperature differences in the atmosphere derive directly the airflow motion within the troposphere and also may affect the tropopause height in relation to the convergence or divergence of air masses. It is also interesting to note from Table 4 that the tropopause height has insignificant correlations with surface level meteorological parameters during the BrO enhancements, except for the air temperature in the Arctic, which is predictable since the tropopause height is a factor more closely related to stratospheric dynamics compared to the surface level weather system.

All analysis so far was correlating meteorological parameters and BrO enhancements for the same time step. In order to investigate possible time-lagging effects of meteorological conditions on total BrO VCDs, correlations between total BrO VCD and meteorological parameters were performed with several days lag for each grid cell. In general, signs of correlations between meteorological parameters and total BrO VCDs are not changed with ± 2 days time-lags and the most pronounced correlation for the tropopause height and surface air temperature appears without the time lag (see Fig.21 and 22). In particular, the tropopause height shows clearly the strongest negative correlation with total BrO VCD in both the Arctic and Antarctic when there is no time-lag and has a gradient where the correlation coefficient weakens with the time-lags. This may indicate that the effect of stratospheric contribution on total BrO VCD according to the change in the tropopause height is immediate and the effect of time-lagging is not significant compared to other factors. However, total BrO VCD shows slightly larger negative correlations with the sea level pressure in terms of negative time-lags. Although changes in the correlation coefficients are small, the analysis result indicates that the current total BrO VCDs are more related to the atmospheric pressure systems of the previous days than those of the next days. Also, surface wind speed shows positive correlations with total BrO VCD, but it can be confirmed that as the time-lag increases, the relationship weakens with a wide correlation coefficient range.

Table 4. Cross-correlations of meteorological parameters for the enhanced total BrO cases in the Arctic and Antarctic sea ice region.

	Arctic				**Antarctic**			
	Sea level pressure	Temperature	Wind speed	Tropopause height	Sea level pressure	Temperature	Wind speed	Tropopause height
Sea level pressure	1	-0.186	-0.228	0.186	1	-0.163	-0.195	0.173
Temperature		1	0.183	0.249		1	0.158	-0.009
Wind speed			1	-0.013			1	-0.05
Tropopause height				1				1

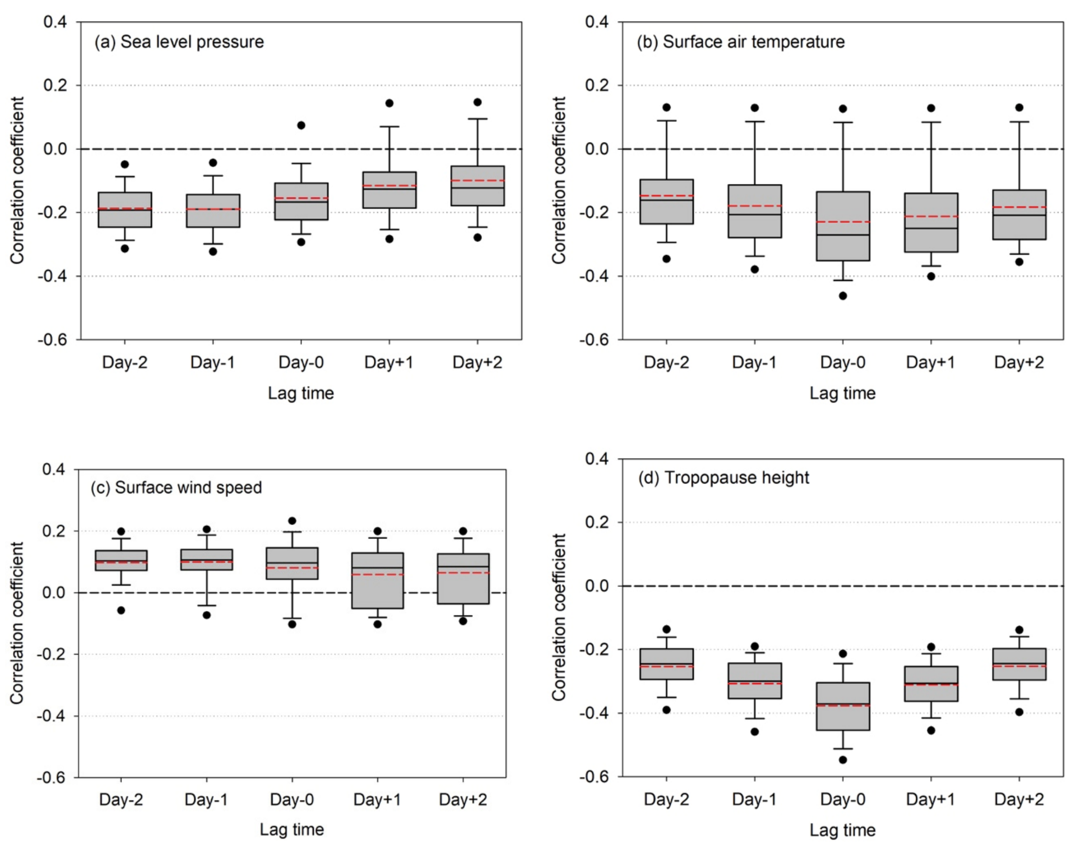

Figure 21. Box and whisker plots of the time-lagged correlation between the total BrO VCD and each meteorological parameter (a) sea level pressure, (b) surface air temperature, (c) wind speed at 10 m, and (d) tropopause height in the Arctic sea ice region. Time-lagged correlation coefficients performed in all reference grid cells with different time-lags (-2, -1, 0, +1, and +2 days) are summarized. The 5th, 10th, 25th, 50th (median), 75th, 90th and 95th percentiles of correlation coefficients are represented by the bottom filled circle, the lower whisker, horizontal lines of the box, the upper whisker, and the upper filled circle, respectively, and the mean value is shown as a red dashed line.

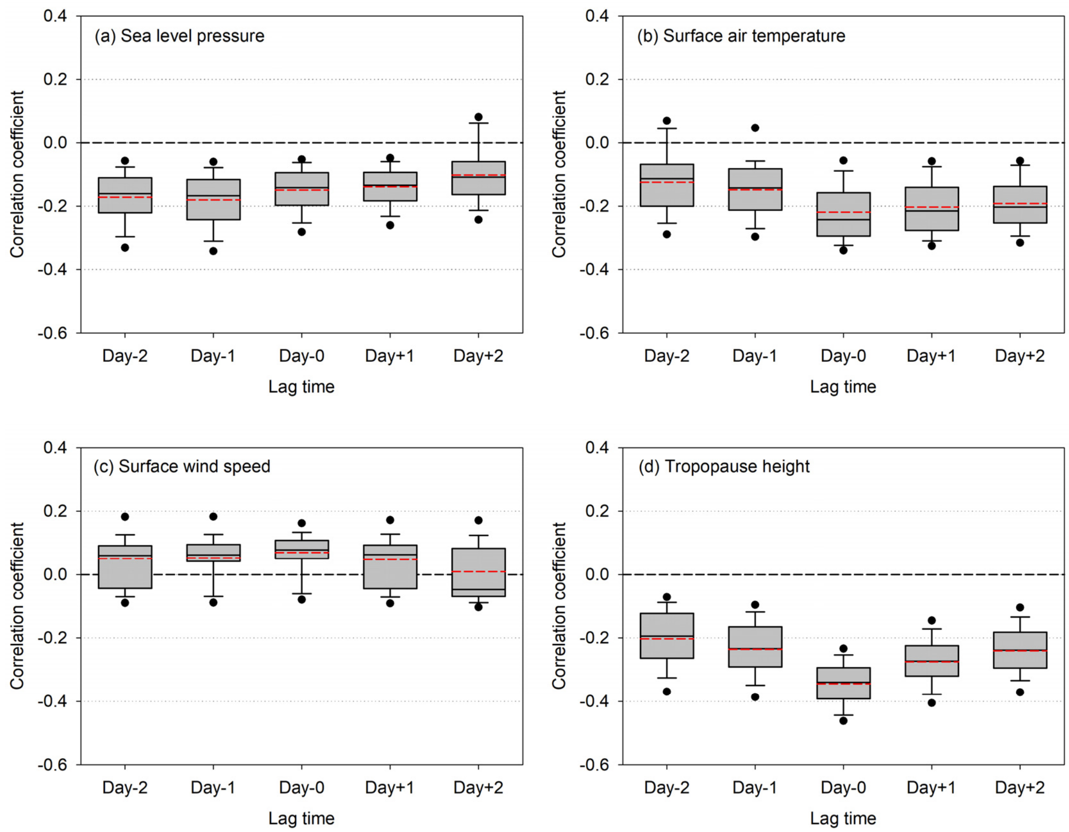

Figure 22. As Fig. 21, but for the Antarctic sea ice region.

Specific comments:

P2L35-36: 'Overall, the BrOx catalytic destruction cycles are significant but not as important as those of ClOx', though this statement is correct, recent modelling-based work indicates the cross-halogen reactions (e.g. between Br and Cl) are important in terms of stratospheric ozone depletion, see recent work by Fraiser et al. (2019).

We have revised the text and added a sentence as suggested (blue text) in section 1 (P2 L34):

"Inorganic bromine is then involved in a variety of stratospheric chemical reactions. The ozone catalytic removal cycles involving bromine atoms, Br, and bromine monoxide (BrO), known as BrOx, have higher chain lengths than those involving chlorine atoms, and chlorine monoxide (ClO), together known as ClOx. Although bromine is more effective at depleting ozone, it cannot be said that BrOx catalytic destruction cycles are more important than those of ClOx since bromine released by man is much less than chlorine. Also, recent studies found that the sensitivity of the ozone depletion to bromine concentration depends on the amount of chlorine present by the chlorine/bromine cross reaction (Yang et al., 2014; Dennison et al., 2019). More details of the current assessment of the role of bromine in the stratosphere are reported in the recent ozone assessment (WMO, 2018)."

Yang, X., Abraham, N. L., Archibald, A. T., Braesicke, P., Keeble, J., Telford, P. J., Warwick, N. J., and Pyle, J. A.: How sensitive is the recovery of stratospheric ozone to changes in concentrations of very short-lived bromocarbons?, Atmos. Chem. Phys., 14, 10431–10438, https://doi.org/10.5194/acp-14-10431-2014, 2014.

Dennison, F., Keeble, J., Morgenstern, O., Zeng, G., Abraham, N. L., and Yang, X.: Improvements to stratospheric chemistry scheme in the UM-UKCA (v10.7) model: solar cycle and heterogeneous reactions, Geosci. Model Dev., 12, 1227–1239, https://doi.org/10.5194/gmd-12-1227-2019, 2019.

P2L43-44: 'An important tropospheric inorganic bromine source is the polar sea ice region (Kaleschke et al., 2004 and references therein)'. It would be better to cite a review paper (e.g. Abbatt et al., 2012) here, as Kaleschke et al.'s paper specifically addressed the importance of frost flowers. Or you can specify various proposed candidates of sea ice sourced bromine from frost flowers (Kaleschke et al. 2004), first-year sea-ice (Simpson et al., 2007b; Pöhler, et al., 2010), sea salt aerosol produced from blowing snow (Yang et al., 2008), stratospheric origin (Salawitch et al., 2010), snowpack photochemistry (e.g. Pratt et al., 2013) and sea spray from open leads (e.g. Peterson et al., 2015). See references shown below.

As suggested, we have revised the text in section 1 (P2 L43) and added references as:

"An important tropospheric bromine source is the polar sea ice region (Abbatt et al., 2012 and references therein). First-year sea-ice (Simpson et al., 2007b; Pöhler, et al., 2010), frost flowers (Kaleschke et al. 2004), sea salt aerosol produced from blowing snow (Yang et al., 2008), snowpack photochemistry (Yang et al., 2008; Pratt et al., 2013), stratospheric origin (Salawitch et al., 2010), and sea spray from open leads (Peterson et al., 2015) have been proposed as major sources of bromine in polar region."

Abbatt, J. P. D., Thomas, J. L., Abrahamsson, K., Boxe, C., Granfors, A., Jones, A. E., King, M.

D., Saiz-Lopez, A., Shepson, P. B., Sodeau, J., Toohey, D. W., Toubin, C., von Glasow, R., Wren, S. N., and Yang, X.: Halogen activation via interactions with environmental ice and snow in the polar lower troposphere and other regions, Atmos. Chem. Phys., 12, 6237–6271, https://doi.org/10.5194/acp-12-6237-2012, 2012.

Simpson, W. R., Carlson, D., Hönninger, G., Douglas, T. A., Sturm, M., Perovich, D., and Platt, U.: First-year sea-ice contact predicts bromine monoxide (BrO) levels at Barrow, Alaska better than potential frost flower contact, Atmos. Chem. Phys., 7, 621–627, https://doi.org/10.5194/acp-7-621-2007, 2007b.

Pöhler, D., Vogel, L., Frieß, U., and Platt, U.: Observation of halogen species in the Amundsen Gulf, Arctic, by active long-path differential optical absorption spectroscopy, P. Natl Acad. Sci., 107, 6582–6587, doi:10.1073/pnas.0912231107, 2010.

Pratt, K. A., Custard, K. D., Shepson, P. B., Thomas, D. A., Pohler, D., General, S., Zielcke, J., Simpson, W. R., Platt, U., Tanner, D. J., Huey, L. G., Carlson, M., and Stirm, B. H.: Photochemical production of molecular bromine in arctic surface snowpacks, Nat. Geosci., 6, 351–356, doi:10.1038/ngeo1779, 2013

P4L119-123: 'In this study, to overcome this snapshot treatment of elevated BrO events during polar spring, and to obtain a more general understanding of the enhancements of total BrO columns, we …' I do not agree that snapshot study is less-advantaged comparing to general statistic study. They both have their individual advantage and disadvantage, they may compensate each other at some point. I suggest a slight change to the tone used in the statement.

As suggested, we changed the tone used in the statement as follows (blue text) in section 1 (P4 L119):

"As mentioned above, many studies on the possible sources of BrO enhancements and the driving meteorological conditions in polar regions have been conducted using ground-based and satellite measurements. Study results clearly indicate that meteorological conditions affect the processes of BrO enhancements in several ways. These previous studies mainly focus on specific case-studies or analysis using relatively short-term datasets. This study aims at adding to this body of knowledge and to obtain a more general and comprehensive understanding of the enhancements of total BrO columns using a consistent long-term dataset. Therefore, we statistically analyse the spatial distribution of occurrence frequency of enhanced total BrO column and its relationship to various meteorological parameters in the Arctic and Antarctic sea ice regions by using a 10 year long-term dataset. In particular, the relationship between total BrO vertical columns retrieved from GOME-2A/2B and meteorological fields including sea level pressure, surface level wind speed and direction, surface air temperature, and tropopause height were investigated."

P15/P18: in P15L476-477 'Surface wind speed is not significantly correlated with total BrO vertical column density in either the Arctic or Antarctic.' A similar statement is also in the summary (P18L557). However, they are contrary to the results shown in Table 3 and Fig. 20, where it shows 'Note that all Spearman's rank correlations are significant ($p<0.001$)'. Is surface wind speed significantly correlated with total BrO vertical column density or not?

In a statistical hypothesis test, p-value and r are indicators of different statistical meanings, finding something that is likely above chance (p-value) or finding meaningful correlations between variables (r). Therefore, having a low correlation coefficient and significant p-value between the surface wind speed and total BrO vertical column density is not a contradictory concept.

To avoid misunderstanding in the interpretation of the analysis results, the sentences (P15 L476-477 and P18 L557) has been changed as follows in the revised manuscript (blue text):

"Surface wind speed has correlation coefficients close to 0 with total BrO vertical column density in both the Arctic and Antarctic, and even for wind speeds above 8 $m \cdot s^{-1}$, which can cause blowing snow, there is no clear relationship between them." This could either be due to stratospheric air dominating the total BrO column or due to the large variability of wind speed conditions under which tropospheric BrO explosion events occur according to previous studies described above.

"Sea level pressure and surface level wind speed are negatively and positively correlated with the total BrO vertical column density, respectively, but their correlation coefficients are low and the strengths of the relationships are weak."

P17L533-4 and L552-555, the two statements are actually duplicated, either delete one or move one to section 4.3.2.

As suggested, we have revised sentences (P17 L553-555) as follows in the revised manuscript:

"One remarkable point is that the temperature is negatively correlated with total BrO column in most sea ice regions, but has a positive correlation over the central Arctic, which is the same as the result of the temperature anomaly pattern discussed above. ~~The opposite correlation pattern in the central Arctic where surface air temperature is low might be due to the transport of enhanced BrO plumes from relatively low latitude sea ice regions and low surface temperatures enough to form frost flowers that act as a source of bromine explosion."~~

P27 Table 3: given that the occurrence of blowing snow has a threshold wind speed, e.g. 7~8 m/s, I suggest add an extra row in the Table to show the correlation coefficients for wind speeds > 8 m/s (or >12 m/s).

We have added an extra row in Table 3 for the Spearman rank correlation coefficients of wind speeds > 8m/s as suggested, and revised the text as in section 4.3 (P15 L476-477):

"Surface wind speed has correlation coefficients close to 0 with the total BrO vertical column density in both the Arctic and Antarctic, and even for wind speeds above 8 $m \cdot s^{-1}$, which can cause blowing snow, there is no clear relationship between them."

Table 3. Spearman rank correlation coefficients between total BrO VCDs and four meteorological parameters. The results are shown separately for different months in spring in the Arctic and Antarctic. Note that all Spearman's rank correlation coefficients are significant ($p < 0.001$). Abbreviation: MA (March to April), SO (September to October).

	Arctic			**Antarctic**		
	Mar	**Apr**	**MA**	**Sep**	**Oct**	**SO**
Sea level pressure	-0.032	-0.098	-0.070	-0.228	-0.053	-0.130
Temperature	-0.242	-0.149	-0.180	-0.305	-0.110	-0.193
Wind speed	-0.011	0.067	0.035	0.038	0.043	0.040
($v \geq 8\ m \cdot s^{-1}$)	-0.002	0.026	0.010	0.041	0.035	0.037
Tropopause height	-0.336	-0.298	-0.315	-0.400	-0.307	-0.345

Minors:

P1L26: change 'they release bromine atoms and Br and bromine monoxide (BrO)...' to 'they release bromine atoms, e.g. Br and bromine monoxide (BrO) ...'

This has been corrected in the revised manuscript.

P1L28: Theys et al., 2009b should be 2009a, as Theys et al., 2009a has not been cited.

Thanks for pointing this out. We have corrected all relevant references.

P2L31: change ‘2Br2’ to ‘Br2’

This has been corrected in the revised manuscript.

P2L32: again you cited ‘They et al., 2009b’ before their 2009a paper.

This has been corrected in the revised manuscript. Theys et al., 2009a has been cited before in P1L28.

P3L67: Theys et al., 2009a should be cited before 2009b.

This has been corrected in the revised manuscript.

P4L112: ‘A bromine explosion event linked to cyclone development in the Arctic was investigated by Blechshmidt et al. (2016).’ The same event was also studied by Zhao et al., 2016.

As suggested, we have added the reference Zhao et al., 2016 in the revised manuscript.

“A bromine explosion event linked to cyclone development in the Arctic was investigated by Zhao et al. (2015) and Blechschmidt et al. (2016).”

Zhao, X., Strong, K., Adams, C., Schofield, R., Yang, X., Richter, A., Frieß, U., Blechschmidt, A.-M., and Koo, J.-H.: A case study of a transported bromine explosion event in the Canadian high Arctic, J. Geophys. Res. Atmos., 121, 457–477, doi:10.1002/2015JD023711, 2016.