Response to anonymous referee #2

We thank the reviewers for their constructive comments and suggestions which helped us to improve our manuscript. We have addressed their questions as follows:

Seo et al. present first BrO data from TROPOMI, made a number of tests for optimal SCDs retrievals and present a number of interesting case studies. It is an interesting work. The manuscript is clearly written and the methodology is sound. This study should be published in Atmos. Meas. Tech. if the authors successfully address the following (minor) comments:

- The main remark is that section 3.1 on sensitivity test is rather long and ends up with findings that are mostly know. The method of Vogel is meaningful because it was originally applied on synthetic spectra. Here the technique is applied on real spectra, hence no firm conclusions can be drawn on the optimal wavelength range. The interference with O3 and SO2 at short UV is well known and the Ring effect at longer wavelength is not surprising either. The final selection of fitting interval of 333.5-357nm is not very different from other studies and I encourage the authors to cite the past papers.

We agree that the results of the sensitivity study are not very surprising considering results from previous work on synthetic data and other measurements. However, we disagree that the sensitivity tests on real data are less relevant than the synthetic data tests from Vogel et al. While the fact that we do not know the true column is a limitation, the use of real data from different realistic scenarios makes the study much more relevant for real data analysis. Synthetic data are great to investigate the theoretical limitations of a technique like DOAS, but many real world problems are not fully reflected by studies on synthetic data.

Following the suggestions for the reviewer, more references have been included in the text where appropriate.

In section 3.1.1:

Aliwell, S. R., Van Roozendael, M., Johnston, P. V., Richter, A., Wagner, T., Arlander, D. W., Burrows, J. P., Fish, D. J., Jones, R. L., Tornkvist, K. K., Lambert, J. C., Pfeilsticker, K., and Pundt, I.: Analysis for BrO in zenith-sky spectra: An intercomparison exercise for analysis improvement, Journal of Geophysical Research-Atmospheres, 107, 10.1029/2001jd000329, 2002.

Theys, N., Van Roozendael, M., Hendrick, F., Yang, X., De Smedt, I., Richter, A., Begoin, M., Errera, Q., Johnston, P. V., Kreher, K., and De Maziere, M.: Global observations of tropospheric BrO columns using GOME-2 satellite data, Atmospheric Chemistry and Physics, 11, 1791-1811, 10.5194/acp-11-1791-2011, 2011.

In section 3.1.3:

Theys, N., Van Roozendael, M., Dils, B., Hendrick, F., Hao, N., and De Mazière, M.: First satellite detection of volcanic bromine monoxide emission after the Kasatochi eruption, Geophysical Research Letters, 36.3, 10.1029/2008gl036552, 2009.

In section 3.1.5:

Burrows, J. P., Platt, U., and Borrell, P.: The remote sensing of tropospheric composition

from space, Springer Science & Business Media, 2011.

Theys, N., Van Roozendael, M., Hendrick, F., Yang, X., De Smedt, I., Richter, A., Begoin, M., Errera, Q., Johnston, P. V., Kreher, K., and De Maziere, M.: Global observations of tropospheric BrO columns using GOME-2 satellite data, Atmospheric Chemistry and Physics, 11, 1791-1811, 10.5194/acp-11-1791-2011, 2011.

In section 3.1.1, it is written that additional cross-sections for ozone (Pukite et al., 2010) could improve the fits at shorter wavelengths. I propose to test this (simple) approach as it might further stabilize the retrievals.

As you suggested, we performed additional sensitivity test for the polar BrO measurement scenario by adding 2 Pukite pseudo cross sections of O3 at 223 K to the standard DOAS setting. The sensitivity test results show a clear improvement for the fit in particular in the shorter wavelength range. We have added test results and their analysis in Appendix A:

These unphysical negative SCDs and high fitting RMS values may be attributed to interferences of other absorbers, which have strong absorption structures at shorter wavelengths, in particular, O3 which has a maximum at high latitudes in the spring season (Monks, 2000). This can potentially be improved by introducing additional ozone cross-sections, which attempt to account for effects arising from changes in the light path with wavelength (Pukite et al., 2010) (see Appendix A).

Appendix A. Improvement of the BrO retrieval with the Pukite Taylor series approach

To investigate the possibility of a DOAS fit improvement for the polar BrO measurement scenario by applying the Taylor series approach (reference to Pukite), we performed an additional sensitivity test. The test was conducted in the same way and with the same measurement scenario as described in section 3.1.1, but two pseudo cross sections of O_3 at 223 K ($\lambda\sigma_{O3}$ and σ_{O3}^2) were added to the standard DOAS settings. The reason for choosing the lower temperature O_3 cross section is that this temperature is closer to the polar lower stratospheric temperature in spring. These two fitting parameters are terms derived by a Taylor series expansion to account for the wavelength dependency of the SCD which results from changes in light path distribution with wavelength and absorption strength (Pukite et al., 2010). Pukite et al. (2010) demonstrated that the application of the Taylor series approach to strong absorber O_3 leads to an improvement for the fit of the weaker absorber BrO in the UV range of limb measurements.

Fig. A1 shows BrO retrieval results obtained with the DOAS settings including the Taylor series approach for the TROPOMI polar BrO measurement scenario. Compared with the standard retrieval results (Fig. 2 in section 3.1.1), BrO retrieval results applying the Taylor series approach show reduced fitting RMS values across the whole retrieval wavelength range (see Fig. A2, right plot). In particular, fitting results at wavelength ranges with a start limit between 323-327.6 nm where negative BrO SCDs and high fitting errors occurred due to strong O_3 interference are significantly improved as BrO SCDs increased by ~1.4×10^{14} molec cm^{-2} and fitting errors decreased by ~32 %. Also, the abrupt changes of BrO SCDs around 333 nm of start wavelength and wavelength range with start limits of 335-337.6 nm and end limits of 349-353.6 nm are moderated by use of the Taylor series expansion for O_3. These sensitivity test results using TROPOMI nadir measurements clearly demonstrate that introducing the Taylor series approach for O_3 results in an improvement of the DOAS fit. However, as is also clear from Figure A1, not all of the problems at low wavelengths apparent in Figure 2 are solved by including the Pukite terms.

For the fitting window selected in this study (334.6-358 nm), the application of the Taylor series approach for O_3 does not significantly affect BrO retrieval results compared with the standard DOAS retrieval. However, as can be seen from Fig. A2, effects of the Taylor series expansion for O_3 on the BrO SCD retrieval vary depending on the retrieval wavelength interval. The strength of absorption and the slant path of scattered light in the atmosphere vary considerably with wavelength, and thus the degree of improvement by the Taylor series approach for O_3 in the BrO retrieval is also different depending on the fitting wavelength range. Therefore, it is necessary to evaluate the improvement of the SCD retrieval by the Taylor series approach with respect to standard DOAS retrieval according to the fitting window selected. Moreover, we showed only sensitivity test results applying the Taylor series expansion of the lower temperature O_3 cross section to TROPOMI polar BrO measurements in this section, but note that the effect of the Taylor series approach may be different for different trace gas cross sections, temperature, and measurement scenarios.

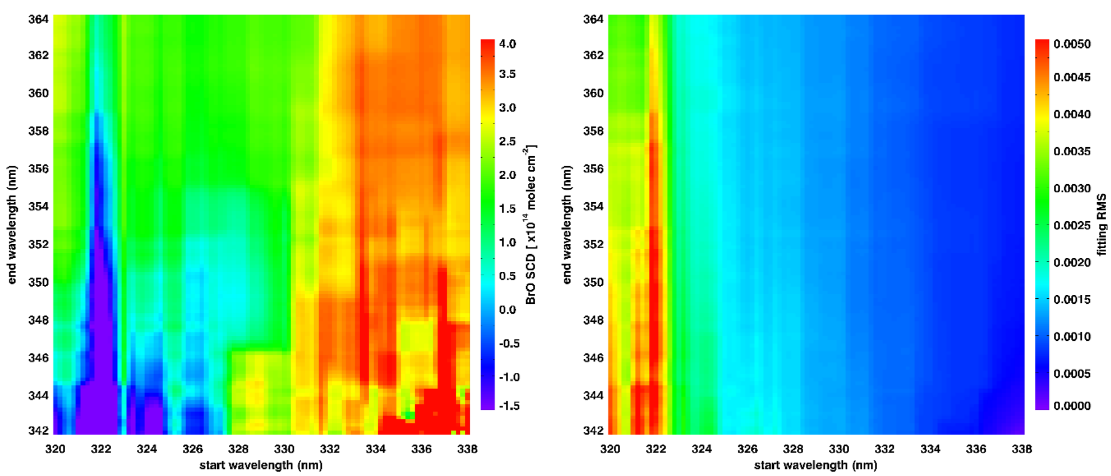

Figure A1. Color coded means of BrO SCDs (left) and fitting RMS values (right) retrieved when including the Taylor series approach for O3 in the DOAS analysis.

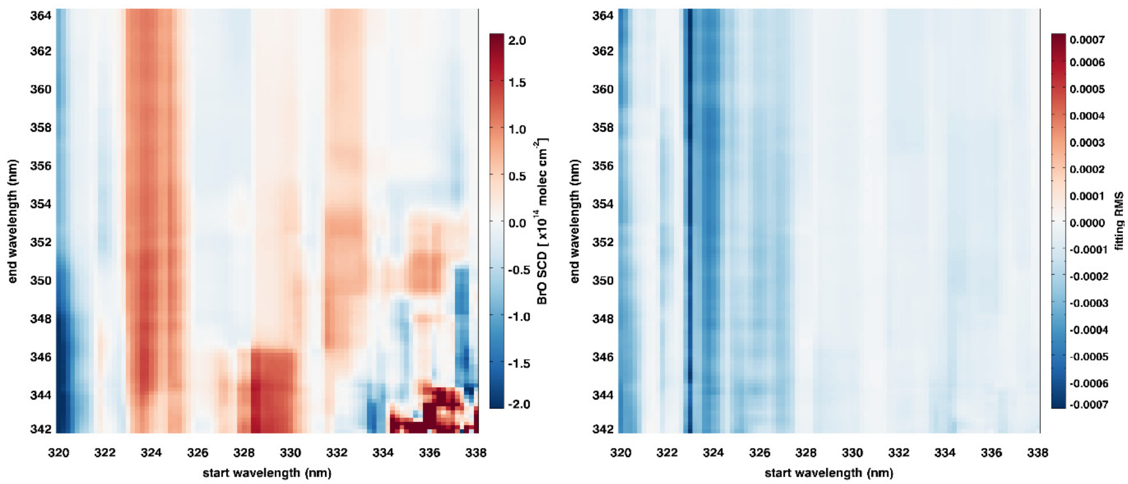

Figure A2. Color coded means of differences for BrO SCDs (left) and fitting RMS values (right) between analyses including the Taylor series approach (see Fig. A1) and the standard DOAS (see Fig.2 in section 3.1.1).

References are missing: on page 1, studies on the presence of global BrO background in the free-troposphere should be listed. References for BrO observations by CIMS and long-path DOAS should be added. An important paper on MAXDOAS BrO in polar region is also: Frie? U., H. Sihler, R. Sander, D. Pöhler, S. Yilmaz, and U. Platt (2011), The vertical distribution of BrO and aerosols in the Arctic: Measurements by active and passive differential optical absorption spectroscopy, J. Geophys. Res., 116, D00R04, doi:10.1029/2011JD015938. Finally the work of Theys et al., ACP, 2011 is absent throughout the manuscript and should be added.

As you suggested, references have been added in the revised manuscript:

P1 L26 (in the revised manuscript): “BrO observations have been carried out by in-situ chemical ionization mass spectrometry (CIMS) (Liao et al., 2011; Choi et al., 2012), ground-based differential optical absorption spectroscopy (DOAS) measurements such as long-path DOAS (LP-DOAS) (Hönninger et al., 2004; Liao et al., 2011; Stutz et al., 2011) and multi-axis DOAS (MAX-DOAS) (Hönninger et al., 2004; Frieß et al., 2011; Zhao et al., 2016).”

P2 L14: “The higher spatial resolution data of GOME-2 and OMI have been successfully used to monitor daily global distribution (Theys et al., 2011) ~

P6 L29: “a spectral cross correlation between BrO and HCHO were also identified in Theys et al. (2011) and Vogel et al. (2013)”

Liao, J., Sihler, H., Huey, L., Neuman, J., Tanner, D., Friess, U., Platt, U., Flocke, F. M., Orlando, J. J., Shepson, P. B., Beine, H. J., Weinheimer, A. J., Sjostedt, S. J., Nowak, J. B., Knapp, D. J., Staebler, R. M., Zheng, W., Sander, R., Hall, S. R., and Ullmann, K.: 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, 2011.

Stutz, J., Thomas, J. L., Hurlock, S. C., Schneider, M., von Glasow, R., Piot, M., Gorham, K., Burkhart, J. F., Ziemba, L., Dibb, J. E., and Lefer, B. L.: Longpath DOAS observations of surface BrO at Summit, Greenland, Atmospheric Chemistry and Physics, 11, 9899-9910, 10.5194/acp-11-9899-2011, 2011.

Hönninger, G., Leser, H., Sebasti´an, O., and Platt, U.: Ground-based measurements of halogen oxides at the Hudson Bay by active longpath DOAS and passive MAX-DOAS, Geophys. Res. Lett., 31, L04111, doi:10.1029/2003GL018982, 2004.

Frieß, U., H. Sihler, R. Sander, D. Pöhler, S. Yilmaz, and U. Platt (2011), The vertical distribution of BrO and aerosols in the Arctic: Measurements by active and passive differential optical absorption spectroscopy, J. Geophys. Res., 116, D00R04, doi:10.1029/2011JD015938.

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, Journal of Geophysical Research-Atmospheres, 121, 457-477, 10.1002/2015jd023711, 2016.

Theys, N., Van Roozendael, M., Hendrick, F., Yang, X., De Smedt, I., Richter, A., Begoin, M., Errera, Q., Johnston, P. V., Kreher, K., and De Maziere, M.: Global observations of tropospheric BrO columns using GOME-2 satellite data, Atmospheric Chemistry and Physics, 11, 1791-1811, 10.5194/acp-11-1791-2011, 2011.

Needs for clarifications:

Section 2: band 3 is not starting at 320 nm.

According to the latest version of TROPOMI L01b ATBD and IODS, the spectral performance range of band 3 is specified as 320-405 nm. While there is data at shorter wavelengths, it is in the overlapping regions between band 2 and band 3.

[1] Table 1 of P 22, ATBD; Algorithm theoretical basis document for the TROPOMI L01b data processor; source: KNMI, ref: S5P-KNMI-L01B-0009-SD; issue: 8.0.0; date: 2017-06-01;

url: https://sentinel.esa.int/documents/247904/2476257/Sentinel-5P-TROPOMI-Level-1B-ATBD

[2] Table 1 of P 15, IODS; Input/output data specification for the TROPOMI L01b data processor; source: KNMI; ref: S5P-KNMI-L01B-0012-SD; issue: 9.0.0; date: 2018-04-01;

url: https://sentinel.esa.int/documents/247904/3119978/Sentinel-5P-Level-01B-input-output-data-specification

Veefkind et al. (2012) specified the spectral range of band 3 as 310-405 nm, but this paper was written before the launch of TROPOMI and contains outdated information that is different from the current state (ex. the current spatial sampling for UV/vis band is 3.5x7 km^2, not 7x7 km^2; the current spectral performance range of band 3 is 320-405 nm, not 310-405 nm).

Page 3 l27: it is not only noise that results from interferences but also important are biases. Please clarify.

The sentence has been changed to as (P3 L29 in the revised manuscript):

"In general, larger fitting windows can improve the quality of DOAS retrievals by using more spectral points, but at the same time, they can increase the noise and bias resulting from interfering signals with other absorbers and wavelength dependent light path lengths."

Page3 l30-31 is unclear. Please rephrase.

The sentence has been rephrased as (P4 L4 in the revised manuscript):

"Thus, finding a compromise for a fitting window that avoids the disadvantages as well as making the best use of the advantages from the retrieval wavelength interval is important to yield the best quality DOAS retrieval result."

Table 1: for volcanic plume, the number of pixels is 1748. Is there not a mistake? It seems a lot, especially that it is mentioned in section 3.1.2 that it a 'small-scale BrO plume'. Please check.

We have checked the number of s5p satellite pixel used for each scenario test. The number of pixels used in volcanic BrO sensitivity test is 1748 and there was no mistake (You can identify the domain of the volcanic plume scenario in Figure 6). However, for the salt marsh sensitivity test, 137 pixels instead of 113 were used and this has been corrected in Table 1.

Page 7, l16: It is speculated about the impact of the Ring effect due aerosol loads and cloud formation after the eruption. Is there any indication about this? Is the TROPOMI AAI product suggesting the presence of aerosols?

As you suggested, we have added this figure in section 3.1.3:

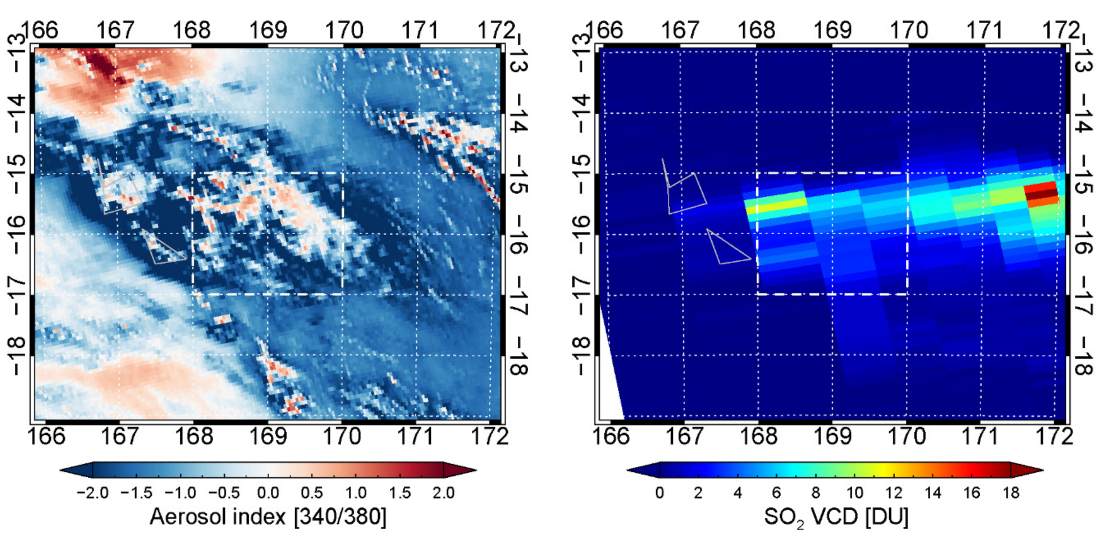

Figure 6. TROPOMI UV aerosol index (340 nm/380 nm) from the operational Level 2 product and OMI SO_2 vertical columns [DU] from the column amount SO_2 TRM (mid-troposphere) of the operational OMSO2 product for a volcanic BrO measurement scenario. The domain used for the sensitivity test is indicated by a gray dashed box.

Section 3.2: a figure illustrating the offset correction would be nice. The approach is not very clear from the text (from l18).

As you suggested, we have added this figure in section 3.2:

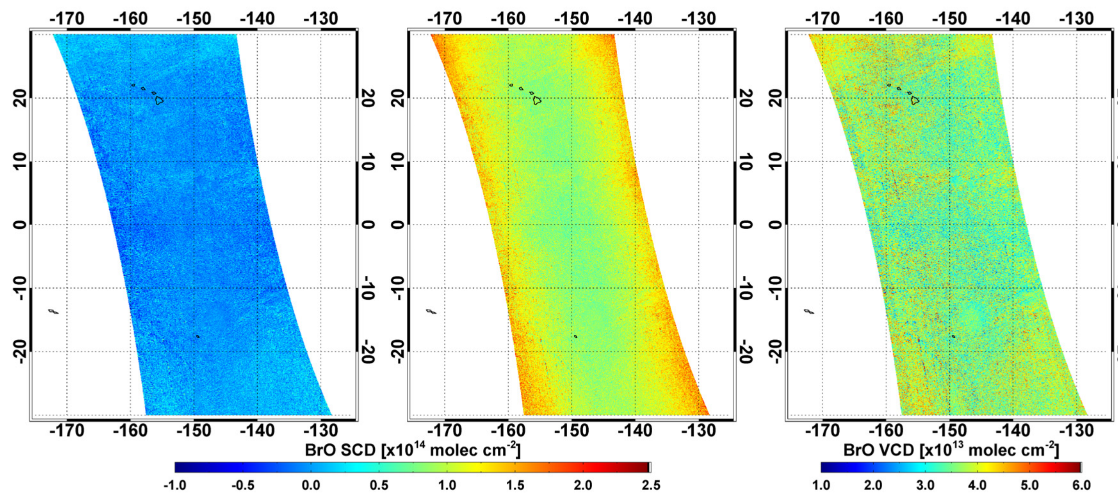

Figure 10. Illustration showing destriping and offset correction steps described in section 3.2 using TROPOMI orbit 2207 on Mar 17 2018. BrO SCDs retrieved by daily row-dependent mean radiances in the Pacific reference sector as background spectrum for the across-track correction (left), offset-corrected BrO SCDs treated by applying the normalization approach including the VZA dependency on the BrO SCDs (middle), BrO VCDs computed by dividing the offset-corrected BrO SCDs by geometric AMFs (right).

P12, l28: The shift of OMI compared to other satellites is attributed to the ‘relatively high measurement noise’. It is unclear. Fitting residuals from OMI are fine.

Compared with GOME-2B and TROPOMI launched in 2012 and 2017, OMI launched in 2004 shows the most severe degradation among the three satellites. The quality of OMI Level 1b radiance data is affected by the row anomaly, which causes errors in the BrO retrieval from the L1b spectra. Although OMI pixels affected by row anomaly were not used in the intercomparison study, the lower spectral stability of OMI compared to the other two satellites leads to systematic errors and biases in the BrO retrieval, which can be confirmed by a slightly positive biased BrO distribution in the Pacific background.

We have revised the sentence as (P15 L22 in the revised manuscript):

“The latter is attributed to be a consequence of systematic biases caused by the relatively lower quality of Level 1b radiance due to the instrument degradation.”

Figures 9 and 10 are difficult to read. Coastlines are not always visible. It would be good to improve the figures.

Figures have been modified to high resolution with adding texts denoting the region in the revised manuscript.

Figure 11 in the revised manuscript:

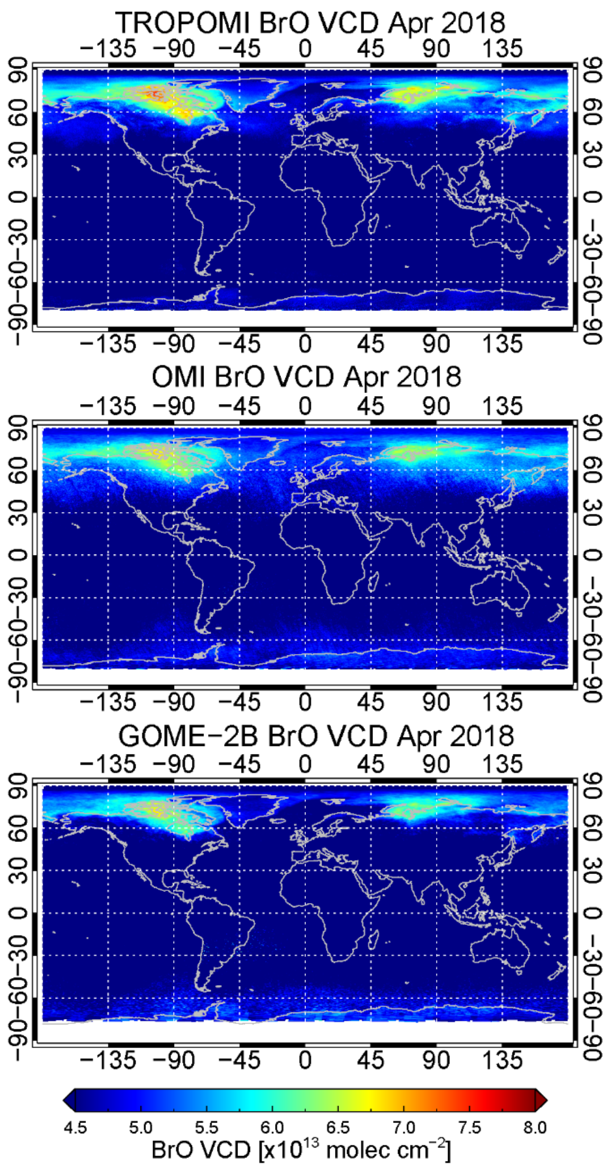

Figure 12 in the revised manuscript:

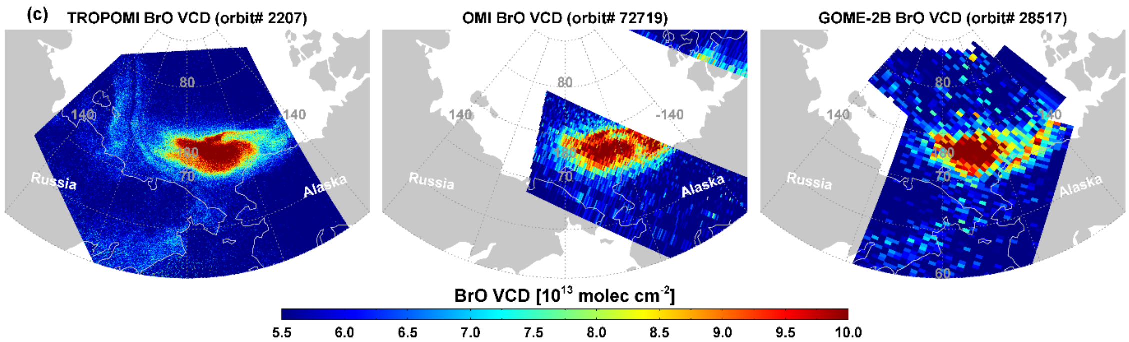

Figure 11: the MODIS pictures are shown but not really commented. What should be seen?

We have revised and added more text in the Section 4.3.1 as:

"Another example of a BrO explosion event case is shown in Figure 14. A relatively narrow and long shape of enhanced BrO over the Beaufort Sea can be found in all three satellite maps. As discussed for the previous example, TROPOMI data with the high spatial resolution of 3.5x7 km^2 yield a more detailed view of the BrO explosion event compared to OMI and GOME-2B. The enhanced BrO plumes appear around open leads and sea ice cracks shown as slightly darker areas in the matching MODIS image (arrows pointing at examples). In particular, the elevated BrO around the Banks Island and the eastern Beaufort Sea (-140~-120 °E, 70~77 °N) could be significantly linked to open leads since frost flowers and sea salt aerosols which act as the source of reactive bromine can be formed in such areas (Simpson et al., 2007). Also, opening of sea ice leads can locally create enhanced vertical mixing and uplifting of bromine sources. However, the analysis of the long enhanced BrO plume from the coast of Alaska towards north should be cautious. The MODIS image composed of the 7-2-1 bands can distinguish clouds (as white) from the sea ice (as sky blue), and this image shows that the shape of the enhanced BrO plume is similar to that of clouds. Convective clouds can be formed around open leads due to the supply of water vapor and enhanced vertical mixing, but computed BrO enhancement over clouds may have an error because of the use of AMFs which do not consider the effects of clouds. In spite of this uncertainty, the enhancement of vertical columns by up to 4×10^{13} molec cm^{-2} compared to the surrounding values indicates that open leads could be associated to the BrO enhancement. Small-scale BrO explosion events around open leads or polynyas can be better investigated with the high spatial resolution TROPOMI data and will be the topic of a follow-up study."

Conclusions, P19, l9: please add the obtained values for the slopes of the regression lines, in addition to the correlation coefficients.

We have added slope values of the regression lines as:

"TROPOMI BrO retrievals show good agreements with OMI and GOME-2B BrO columns with high correlation coefficients (slopes of the regression lines) of 0.84 (0.89) and 0.84 (0.72) for enhanced BrO plumes in Arctic sea ice region, respectively."