We like to thank the reviewer for his/her review and the useful and constructive remarks. We are also thankful for the opportunity to revise our manuscript in light of the reviewers' comments. We are confident that we addressed all reviewers' comments and trust that the revised manuscript is can now be accepted for publication.

In the following, we give the answers to the anonymous referee #2 regarding the preprint manuscript Warnach et al.: Referee comments are reproduced in black, answers in red. Changes to the text in the revised manuscript are also given in red and are enclosed in quotes ("…").

Warnach et al. presented. A new scheme to retrieve BrO with improved precision and accuracy. This is made possible thanks to a combination of improved DOAS settings and sophisticated bias correction. This is an important study in view of the systematic investigation of volcanic BrO plumes measured by TROPOMI. The paper is well structured and a pleasure to read. The approach is scientifically sound. I recommend publication in AMT after addressing my (minor) comments below.

All line numbers in the following answers refer to the original (preprint) version of the manuscript.

Section 2

-In the section describing TROPOMI, there are sub-sections 2.1.1 and 2.1.2 which are very short. The author could consider removing the sub-sections structure (but keep the text in section 2.1).

We moved the paragraph where the O3 VCD calculation is described into Sect. 2.1.2 as suggested by Reviewer #1. Thus, Sect. 2.1.2 is now longer and we hope that this merits an own subsection. In addition, we feel that it would be better to keep the additional data in a separate section, so that the reader can directly find it.

-Section 2.1.2. Here FRESCO and MICRU are mentioned as available cloud products, but the author does not consider OCRA/ROCINN which is the S5P operational cloud product. Is there a reason for not considering OCRA/ROCINN. For the O3 VCD, why was the operational total ozone column product not used?

We appreciate the valuable comment. The primary reason for choosing FRESCO is that its cloud fraction is directly retrieved on the TROPOMI UV-VIS grid. Ergo no transformation from the SWIR grid was needed for this. The MICRU cloud fraction product was tested and in fact used within the first author's Ph.D project (but is not a readily available product), and we therefore found it relevant to mention it.

We acknowledge that the use of OCRA/ROCINN is another viable option for such kinds of studies as it is readily available on the TROPOMI UV-grid via for example the L2 SO2 product.

As OCRA/ROCINN is the operational product, it is worth to mention this, and we changed line 128 to:

"However also other cloud products are available (a comparison of TROPOMI cloud products can be found in Latsch et al., 2022), such as the operational cloud product (OCRA/ROCINN, Loyola et al., 2018), and could in principle be used."

Concerning the selection of the O3 VCD:

The decision not to utilize the operational total ozone column product in our study was based on two factors. Firstly, from a practical standpoint, we found it more convenient to use the ozone column obtained from the BrO fit, as it was readily available and does not require, for example, additional data download.

Second, in contrast to our fit, the operational total ozone column product does not include the SO2 cross-section in the DOAS fit. The total ozone column fit is therefore more susceptible to spectral interference between SO2 and O3. This susceptibility becomes particularly critical when retrieving trace gases within a volcanic plume characterized by high SO2 columns. Consequently, the ozone columns derived from the total ozone fit will deviate within the volcanic plume. This inaccuracy, in turn, would lead to an incorrect correction term within the plume and an erroneous BrO column estimation

Therefore, considering both practicality and accuracy considerations, we made the decision to rely on the ozone column derived from the BrO fit rather than using the operational total ozone column product.

In order to reflect this in the manuscript, we add the following after line 135 (where we also moved the detailed explanation of the O3 VCD calculation following a comment from reviewer #1) after "The O3 VCD is derived directly from the BrO DOAS fit.":

"We favour this O3 VCD over the operational O3 L2 product (1) because it is more practical and most importantly (2) because in difference to our fit, the operational O3 product does not include SO2 within the DOAS fit and is therefore affected stronger by SO2-O3 spectral interference leading to high inaccuracies within volcanic plumes."

We added the following reference:

Loyola, D. G., García, S. G., Lutz, R., Argyrouli, A., Romahn, F., Spurr, R. J. D., Pedergnana, M., Doicu, A., García, V. M., and Schüssler, O.: The operational cloud retrieval algorithms from TROPOMI on board Sentinel-5 Precursor, Atmospheric Measurement Techniques, 11, 409–427, https://doi.org/10.5194/amt-11-409-2018, 2018.

-Equation 1 does not include the broad-band terms. Please add those to the equation.

We thank the reviewer for alerting us of this omission. Indeed, we forgot to include the Polynomial term fitted in the DOAS algorithm to account for broad band absorption structures. We added the polynomial term "P(lambda)" in eq. 1 and added after eq. 1 prior to line 144:

"where sigma is the absorption cross-section and c the concentration of the trace gas i, while the polynomial term P(lambda) accounts for broad-band absorption and scattering processes, e. g. Rayleigh and Mie scattering."

-Equation 3 is incorrect as it should in principle imply the cosine of the angles (SZA, VZA). Please update the equation.

We thank the reviewer for noticing this small but significant omission and added the cosine to both parameters. The denominators now read "cos(SZA)" and "cos(VZA)".

-Section 2.3: it is mentioned that the full zonal band (from 20S to20N) is used as reference region. However, the author is not really justifying its choice. This is a very large region which covers many volcanoes (with potential contamination by strong plumes SO2 and BrO, and to some extend HCHO). Please clarify why it is an advantage to use such an extended reference region compared to a smaller region (e.g., in the equatorial Pacific).

As this comment overlaps with the General comment 1 (GC1) from referee #1, we deemed it beneficial for both referee comments to be address by a single answer, which encompasses all aspects of this topic.

We thank the reviewer for these positive remarks. Indeed, our approach is novel and this warrants some justification, as up to now, the region used to calculate the earthshine reference was chosen over the pacific only (Theys et al., 2017, Seo et al., 2018). There the assumption that no volcanic plume can be assumed is valid in almost all cases.

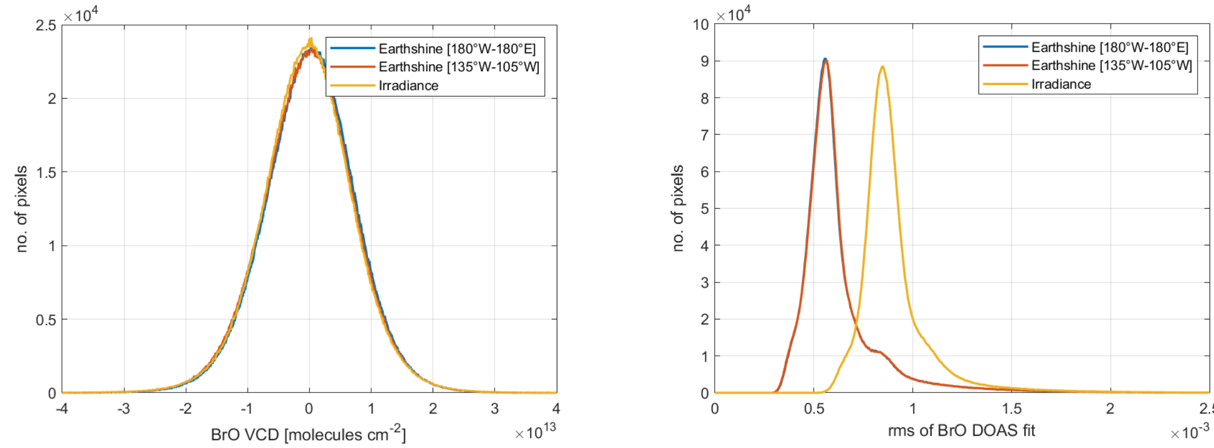
We chose a new approach using a reference spectrum stretching over the complete equatorial region for two reasons:

Firstly, and importantly, we chose a band stretching over the complete equatorial region to better account for variations in the spectral response over the day. We found that there are weak stripe features introduced over the day. Therefore, using a pacific reference spectrum, we observed cross-track stripes at locations at a different longitude (e.g. Europe). These vanished when using a reference spectrum obtained from the same orbit (e.g. over equatorial Africa). We found that using a reference spectrum of the complete equatorial band reduces this stripe pattern while best representing the equatorial reference spectrum of the complete day.

Secondly, using the complete band, (1) enhances the statistics for each detector and (2) ensures that each across-track detector has (nearly) the same number of pixels used for the calculation. This minimizes possible inconsistencies between the across-track detectors.

Nevertheless, it has to be ensured that influence of potential sources do not out-weight these advantages. Therefore, for comparison with the previous methods, we also applied a pacific earthshine reference spectrum fit calculated using (135-105°W, +-20°N). We included the results for October 1 in figure 2, and changed its caption to:

(a) (b)



"Figure 2: (a) Distribution of the BrO VCD and (b) the rms uncertainty of the DOAS fit for the complete equatorial region [+-20°N, +-180°E] on 1 October 2018 employing three different reference spectra: An earthshine spectrum calculated using the complete equatorial region [+-20°N, +-180°E] (blue), an earthshine spectrum calculated using the equatorial pacific region [+-20°N, 135°W-105°W] (red), as well as an irradiance spectrum (yellow). For comparability with the earthshine results, the median BrO VCD (corresponding to the median stratospheric column) is subtracted for the Irradiance BrO VCDs."

We moved lines 191 – 196 after line 185, and changed them to:

> "However, an earthshine spectrum using the complete equatorial latitude band might include influences from volcanoes as well as biogenic or anthropogenic influences. A comparison between the use of the new expanded area earthshine spectrum calculated from the complete equatorial region [+-20°N, +-180°E], the earthshine spectrum from the pacific equatorial region only [+-20°N, 105°-135°W], as well as using an irradiance spectrum is shown in Fig. 2 for measurements over the equatorial region [+-20°N, +-180°E] on 1 October 2018. It can be seen that the retrieved VCD distribution shows no difference or offset between all three fits (here the stratospheric influence in the irradiance data is eliminated for comparison by subtracting the median BrO VCD). The fit root-mean-square (RMS), however, is about 25% lower (at roughly 6x 10^{-3}) for both earthshine fits compared to the irradiance fit. This RMS distribution is in very good agreement with RMS reported over a pacific equatorial region by Seo et al. (2019, Fig. 11b) who employed a DOAS earthshine fit based on a large pacific equator region (+-30°N, 150-240°E) independently from the fit presented in this study."

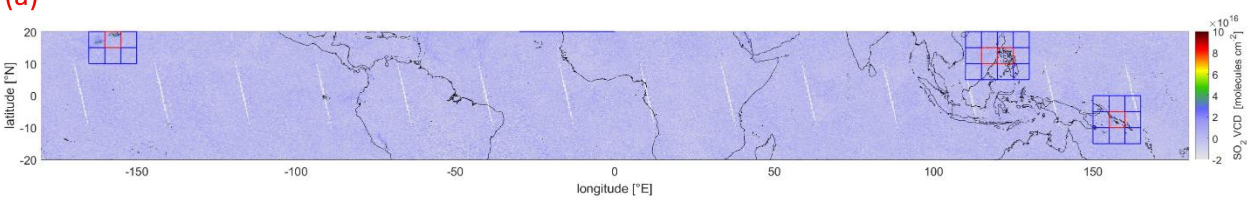
We added a section at the beginning of the appendix (Appendix A) to investigate the strength of a potential contamination of volcanic plumes:

> "In order to quantify the influence of the presence of volcanic plumes within the equatorial reference spectrum region onto the BrO VCDs, we selected two example days: 2 October 2021, where only several, small plumes are present (cf. Fig. A1a, red areas), representative of normal conditions, and 30 July 2018, where a very large plume stretched over a large

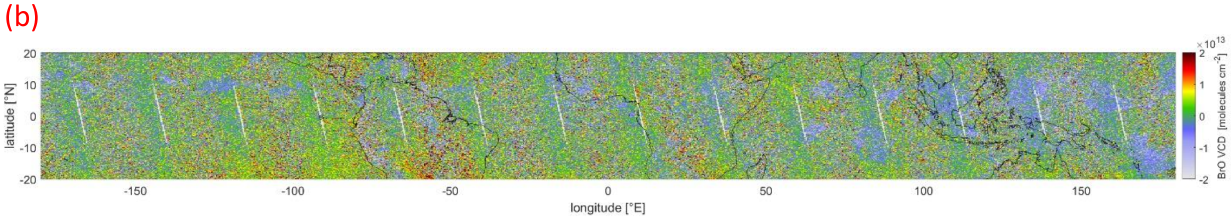
portion of the equatorial region (cf. Fig. A2a, red areas), representative of exceptionally strong volcanic activity within the equatorial region. For both days we identified areas affected by a volcanic plume based on the SO2 signal (as done in Warnach, 2022, cf. Sect. 5.2). Lastly, we calculated the mean BrO VCD within the equatorial region independently for each across-track detector both including and excluding the affected volcanic areas. The difference between both should be equivalent to the contamination of the earthshine reference spectrum.
For the 2 October 2020, there is no difference in the BrO and for SO2 only a small difference for a few detectors (cf. Fig. A1c,d). Our interpretation is that typical signals are too weak to exceed the noise of the BrO retrieval (as the signal-to-noise is two orders of magnitudes larger than for SO2) and that typically only a very small fraction of pixels are affected by volcanic plumes. On the 30 July 2018, which is representative for an exceptionally strong volcanic plume, there is only a contamination of the SO2 SCD of 6x10^{15} molecules cm^{-2} is more than one order of magnitude lower than typical volcanic SO2 SCDs (which are on the order of 1x10^{17} molecules cm^{-2}). For BrO, the difference is even smaller and less than 1x10^{11} molecules cm^{-2} which is also at least 1.5 orders of magnitude lower than typical volcanic signals (which are on the order of 1x10^{13} molecules cm^{-2})."

We added two plot for the 30 July 2018 and 2 October 2020 to showcase the differences:

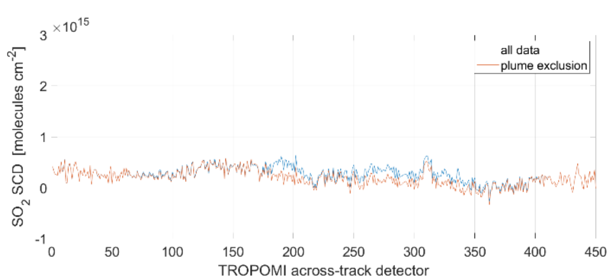
(a)



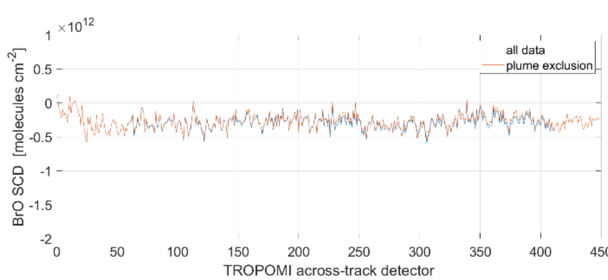
(b)



(c)

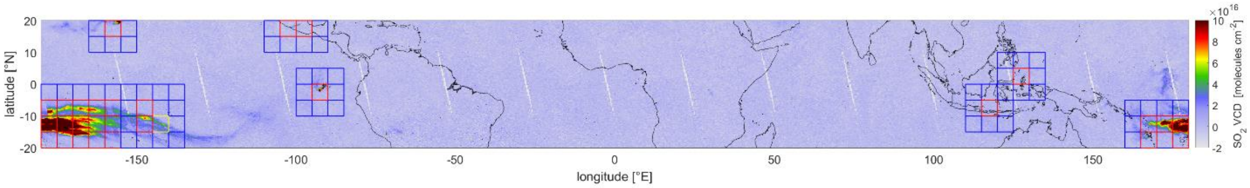


(d)

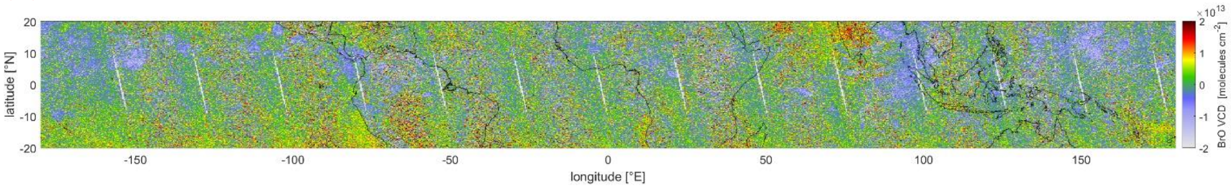


"Figure A1: (a) Map of the SO_2 VCD for the equatorial region on 2 October 2021. The areas of enhanced volcanic signals are marked in red squares. (b) Map of the BrO VCD for the equatorial region on 2 October 2021. (c) Mean SO_2 VCD for each across-track detector considering all pixel (blue) and excluding pixel with volcanic gas columns (red). (d) Same plot for the across-track dependent mean BrO VCD."

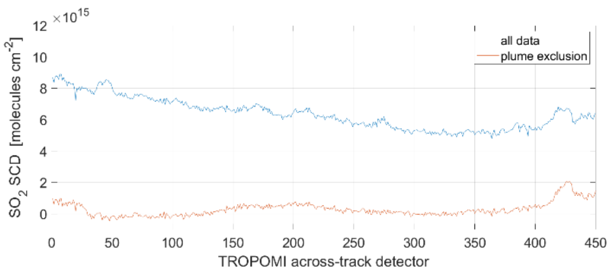
(a)



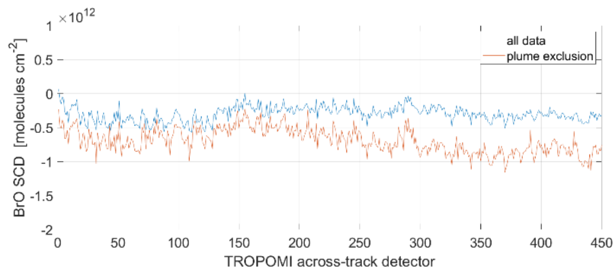
(b)



(c)



(d)



"Figure A2: (a) Map of the SO_2 VCD for the equatorial region on 30 July 2018. The areas of enhanced volcanic signals are marked in red squares. (b) Map of the BrO VCD for the equatorial region on 30 July 2018. (c) Mean SO_2 VCD for each across-track detector considering all pixels (blue) and excluding pixels with volcanic gas columns (red). (d) Same plot for the across-track dependent mean BrO VCD."

In the main body of the manuscript, we added the following text after lines 191-196 (which are modified already above):

„In order to ensure that the inclusion of volcanic plumes within the reference spectrum will not introduce a noticeable contamination into the reference spectrum, we investigated the difference between including and excluding volcanic areas onto the retrieved mean SCD over the equator. This is done for two days in the appendix (cf. Sect. A): 2 October 2021, where only several, small plumes are present (cf. Fig. A1a, red areas), representative of normal conditions, and 30 July 2018, where a very large plume stretched over a large portion of the equatorial region (cf. Fig. A2a, red areas), representative of exceptionally strong volcanic activity within the equatorial region. For the normal conditions on 2 October 2021, excluding the volcanic areas only leads to negligible changes in the SO_2 SCD (cf. Fig. A1c) and no detectable changes in the BrO SCD (cf. Fig. A1d). For the exceptional conditions on 30 July 2018, there is a difference of several 10^{11} molecules cm^{-2} visible between including and excluding the volcanic areas for BrO SCDs (cf. Fig. A2d). However, this is 1.5 orders of magnitude below typical volcanic BrO columns ($1x10^{13}$ molecules cm^{-2}) and therefore negligible. The same is the case for SO_2 SCD, where it is more pronounced, but still 1 order of magnitude below typical volcanic columns of $1x10^{17}$ molecules cm^{-2} (cf. Fig. A2c). Furthermore, the large plume on the 30 July 2018 stretches also over the pacific area typically used as a pacific reference region (e.g. 120°-160°W, as used for the operational SO2 product, Theys et al., 2017, or even more affected using 150°E – 120°W, Seo et al., 2019). Thus, in this exceptional case using a pacific reference sector will also not be free of volcanic influence. To the contrary, in this case the influence is most likely stronger using a pacific reference area, as the plume affects a relatively larger portion of pixels within the reference area compared to our reference area which spans the complete equatorial band. It should further be noted that a constant offset expanding over all across-track detectors would be removed efficiently by our background correction algorithm and would therefore be irrelevant to our approach."

Section 4

-Section 4.1. on the effect of clouds. From Fig4, it is not clear to me whether the observed effect of clouds is an artefact or not. For large CF and elevated clouds, the BrO VCDs are lower which is compatible with a possible cloud shielding of the tropospheric BrO column. I am not sure why this should be corrected.

We thank the reviewer for this comment. We agree, that we have maybe not sufficiently explained why these structures of non-volcanic origin should be removed. The possible cloud shielding constitutes a "background offset" which – while interesting for other applications – is a source of uncertainty for the estimation of the volcanic BrO column and therefore not desirable in this study. This is especially relevant as cloud edges can form a distinct gradient in BrO. Any gradient in the BrO background overlapping with the volcanic plume dispersion pattern can lead to a systematically false gradient in the estimation of the volcanic BrO column.

We add the following sentences to the end of section 3.1 (after line 234):

"As all gradients in the BrO column of non-volcanic origin constitute a potential systematic error source, they ideally are removed via a background correction (e.g. via a spatial polynomial, Hörmann et al., 2013). In order to further improve the accuracy of the BrO retrieval, we investigate a more sophisticated correction scheme in Sect. 4.2."

To also add some more information on the origin of the cloud-related BrO patterns, we added the following sentence prior to the aforementioned sentences:

"The reason for this cloud effect is not fully clear. The effect could be a spectroscopic artefact (e.g. via the Ring Spectrum) or a true shielding effect of a potential tropospheric BrO background column."

-Fig5: it is stated that:" the sign of the relation is inverted and high cloud fraction results in elevated BrO VCDs" but without the information on cloud height it is difficult to know whether there is a significant difference as for the effect of clouds, compared to the tropics. Please add the cloud height map and expand the discussion.

We thank the reviewer for noticing this. We added the FRESCO cloud height to fig. 5 and rearranged the subplots, the caption now reads:

> "Maps of the (a) FRESCO cloud fraction, (b) FRESCO cloud height, (c) retrieved O3 VCD, (d) BrO VCD from fit SR 323 – 360 nm without any correction and (e) BrO VCD from fit SR 323 – 360 nm after applying the correction scheme. Data taken over the northern high latitude region [40° – 60° N, 110° – 50° W] on 1 October 2018."

After re-analyzing the situation, we agree that it is difficult to separate the effect of cloud fraction and O3 for low clouds in this figure. As this distinction is not necessary for the interpretation, we removed the following line 265: "These coincide with cloud cover (indicated by the cloud fraction). In contrast to the equatorial region, the sign of the relation is inverted and high cloud fraction results in elevated BrO VCDs." And replaced it with:

> "These coincide with cloud cover (indicated by the cloud fraction) and show a positive BrO signal for high cloud cover."

Furthermore, we replaced lines 284-288 "While the general […] independent of cloud fraction. This dependency" with the following:

> "While the general CH dependency of the BrO VCD (higher clouds, lower BrO VCD) is prevailing for high latitudes, the CF dependency appears to be changing its sign, so that a lower cloud fraction leads to a lower BrO VCD for 70° N – 90° N. However, there is an overlaying BrO signal for cloud heights between 1000 and 5000 m, independent of cloud fraction in this region, which complicates the interpretation. This feature"

From FigA1, it seems that the 323-360nm range is the one with the strongest cloud impact. I find it hard to justify that this range is the one retained for the final SCD retrievals.

While this is true and would mean that the 323-360nm fit range is not the best, after applying the cloud-ozone correction, this impact is removed and all fit ranges show a similarly negligible cloud impact. Therefore, regarding the cloud impact, there is no "better" or "worse" fit range after the correction is applied and therefore not considered for the fit selection. This is addressed in the text already in line 500: "The systematic influence of clouds and the stratospheric background can be well corrected by the correction scheme described in Sect 4.1 for all eight fits. Thus, there is no preference for a fit range or fit setting."

Section 4.2.

-for low CF, the retrieved cloud height is uncertain, not to say ill-defined. How do you manage this in your correction?

We thank the reviewer for this remark, and agree completely that this has to be considered carefully. We therefore employed a low order polynomial (order two) for the cloud height to prevent potential inconsistencies to affect the correction term.

-Regarding the lat-ozone correction, it is mentioned that 'the latitude band can be adjusted for each volcanic plume' (line 351). I guess this is a future implementation wish or is this really what is implemented. Also, it is not clear how frequent the correction parameters are updated. Is this done separately for each calendar day? Please clarify.

We thank the reviewer for raising this question. The latitude band is in fact chosen individually for each volcano already in the present study and this is mentioned in section 6.3. We realize that our wording in line 351 might suggest otherwise. Therefore, we changed line 352 to read:

> "As the latitude band location can be adjusted freely, the latitude band will be chosen individually for each volcano in order to ensure that the volcano is located at the center of the latitude band and not on its edge (e.g. for Mt. Etna located at 37°N, the latitude band 25°-45° N is chosen for the background correction, cf. Sect. 6.3)."

-Figure 11 is an interesting plot but it is not clear from where the model factor of 6.22e12 is coming. Could you elaborate? Also, in the evaluation of the systematic uncertainty of the retrievals (summarized in Table4), the authors assume that a good estimate of these systematic influences can be obtained from the SCD std over many pixels. The validity of this approach is not clear because random uncertainties are still present and contribute to the std (these are not reducing as the square root of the number of pixels, as they do for the estimated mean). Also, systematic uncertainties like the one related to the BrO cross-section uncertainty is not accounted for.

We thank the reviewer for this comment. The factor 6.22×10^{12} originates from the mean std. deviation without any binning (e.g. single pixel base) at the equator and is used in Fig. 11a only to illustrate the difference. In Fig. 11b the respective model curves for each latitude bands are used. We realize this is not clear from the figure caption and therefore added to the caption of 11a: "For illustration purpose only the model curve for the equatorial region [10°S -10°N] is plotted.". Furthermore, we added to the caption of 11b: "For each latitude band an individual model curve based on the respective standard deviation for pixel binning factor of 1 is used."

We thank the reviewer for questioning the validity of our approach to estimate the systematic uncertainties. Indeed, the standard deviation does not depend on sample size. However, in our approach we bin the data spatially and use these "spatial means" as a new sample. This modified sample has then a lower standard deviation, following a 1/sqrt(N) dependency: a binning of 2 pixels results in a sample with ~0.71 times the original stdev (similarly the standard deviation of the TROPOMI pixels at the edge of the swath increase, because the binning size is reduced there). The data plotted in fig. 11a is therefore the standard deviation of different samples, who all have the same mean value but are less affected by random noise for larger binning factors. We realize that this is not well elaborated in the manuscript. We therefore add change line 443 to :

> "The statistical variation can be quantified by a Gaussian fit over the BrO VCD distribution and the statistical variation then estimated by the standard deviation. Statistical fluctuations can be reduced by spatial binning of neighbouring pixels, which reduces the standard deviation of the binned data by 1/square root of the number of pixel binned. Systematic effects, however, can generally not be eliminated by spatial averaging/binning. Thus, ideally, only the systematic effects remain for high binning factors and the deviation from the expected decrease can be used as an estimate for the order of magnitude of systematic effects. In order to estimate the systematic uncertainties, we employ a spatial binning of neighbouring pixels using binning factors of 1 to 100 in both spatial dimensions, corresponding to 1 to 10000 pixels binned respectively. For each binning factor as well as for each latitude band a Gaussian fit is done separately. The resulting standard deviations are plotted in Fig. 11."

The reviewer is correct in pointing out that systematic uncertainties of the BrO cross-sections are not addressed by this. However, a possible error of the assumed BrO cross-section would basically result in an overall scaling factor of the retrieved BrO SCDs, but would not affect the spatial gradients and regional enhancements over background which are the basis of our quantification of BrO emissions.

Typos and suggestions

-p4, l111: ultra-violet**t** -> ultra-violet

We changed it accordingly.

-p4, l112: characteristic**a** -> characteristics

We changed it accordingly.

-Single sub-section like 3.1 is not necessary. Instead I would write just after l225: "To illustrate the fit performance, the global BrO VCD map for 1 October 2018…"

We thank the reviewer for this improvement in readability and changed it accordingly.

-Fig4c: it would be good to only show the cloud heights above a certain CF threshold, to better appreciate low/high clouds.

We acknowledge the notion of the reviewer, but would rather not mask any values, as we do not do this in our correction scheme.

-FigA1: it would be good to have the same color bar limits for all 8 subplots, otherwise it is difficult to compare the results.

We agree with the reviewer completely and thank for this remark. We changed the colorbar limits to [$-1x10^{13}$ $1x10^{13}$] for all plots.

-line 310: '..the for the wavelength..' -> '..the wavelength..'

We changed it accordingly.

-line 445: '..increasing increasing..'->'..increasing..'

We changed it accordingly.

-Table 3 is redundant. I would propose to remove it. Table 1 could better highlight the preferred settings (e.g., with the corresponding text in bold).

We thank this reviewer for this comment and agree that Table 3 is redundant.

We removed Table 3, changed the reference in line 505 to Table 1 and added "considering the wavelength fit range of 323-360 nm and excluding the absorption cross-section of HCHO." in the same line after "The complete overview of the DOAS fit settings is listed in Table 3"

Furthermore, we changed Table 1, so that the "Species" and "Temperature" have a separate column each (as was done in Table 3).

Additionally, we highlighted the chosen fit wavelength range (323-360nm) in bold and added to the caption: "The proposed final wavelength fit range is highlighted in bold." Furthermore, we added to the footnote of HCHO "Not included in the proposed final fit settings."

-line 532: 'the latitudinal background correction is applied' here you mean the ozone-cloud correction, right?

Yes, we changed it to "ozone latitude correction".