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 #1 regarding the preprint manuscript Warnach et al.: Referee comments are reproduced in black, the author answers in red. Changes to the text in the revised manuscript are also given in red and are enclosed in quotes ("…").

All line numbers in this answer refer to the preprint version of the manuscript.

General comments

I have read with interest the manuscript entitled "A new accurate retrieval of bromine monoxide inside minor volcanic plumes from Sentinel-5 Precursor/TROPOMI" submitted to the EGUsphere/AMT by S. Warnach and collaborators. The study presents a thorough empirical and quantitative investigation of the choice of different retrieval parameters to obtain column densities of BrO in volcanic plumes, which can be applied for global observations with the TROPOMI satellite instrument. In essence, the study investigates the systematic effects of the choice of different spectral intervals and the interference of HCHO, and, most notably, it presents an empirical background correction scheme using latitude-dependent information on O3 and cloud height and fraction parameters derived from independent algorithms applied to TROPOMI observations. Most of these ideas are present in previous studies, and for this reason, in my opinion the main merit of the study does not lie on its originality, but rather on the meticulous analysis to find optimal retrieval parameters and to quantify the expected uncertainty for results obtained at different regions of the world.

The background correction scheme seems in a way similar to the DOAS algorithm, in the sense of separating "structured" information from "smooth" information, but it does so in the spatial domain instead of the spectral domain. The rationale behind is that the choice of an imperfect background, here proposed as the average radiance at a latitude band around the globe, leads to a bias that vary in space smoothly in relation to the strong variation caused by a volcanic plume. The further exploration of the co-location of volcanic SO2 leads to an even more accurate representation of the background and retrieval of the volcanic signal.

GC1) It would be interesting to see a discussion on why the region selected for the background: a band around the equator, which includes many potential sources of volcanic or biogenic interference, does not lead to more noisy results. Is this just an effect of reducing random noise by averaging more pixels? Were the examples presented representative of general conditions? A map of uncorrected SO2 columns in the band used for the background presented in the examples could give a visual representation on how "clean" this background was.

As this comment overlaps with the comment to Section 2.3 from referee #2, 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.

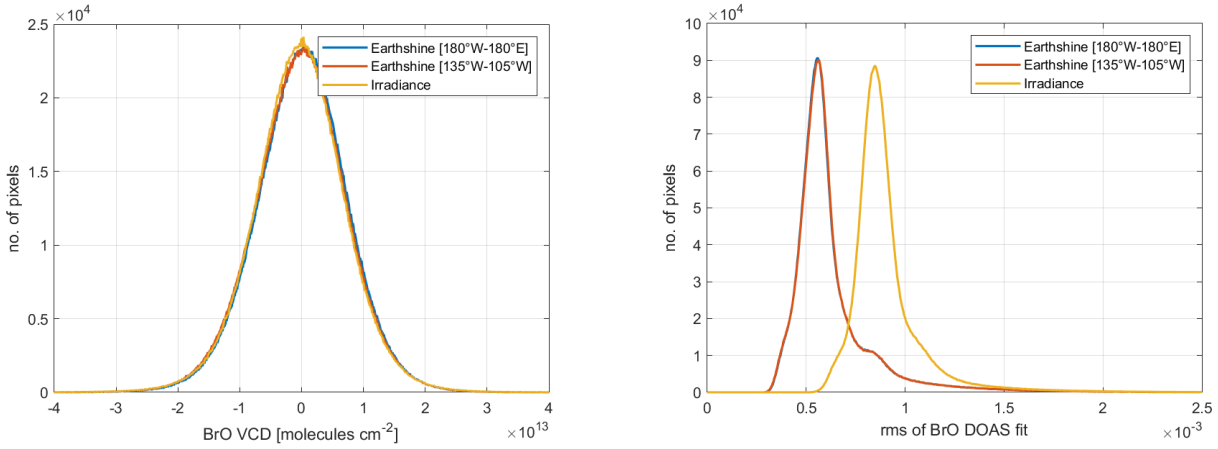
We chose a new approach using a reference spectrum stretching over the complete equatorial region for two reasons:

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."

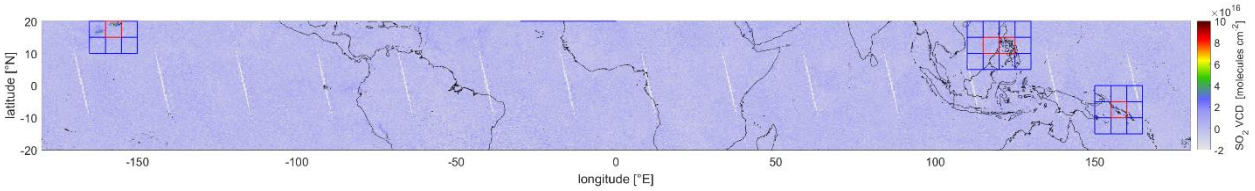
We added a section at the beginning of the appendix (Appendix A) to investigate the strength of a potential contamination of volcanic plumes:

> "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}

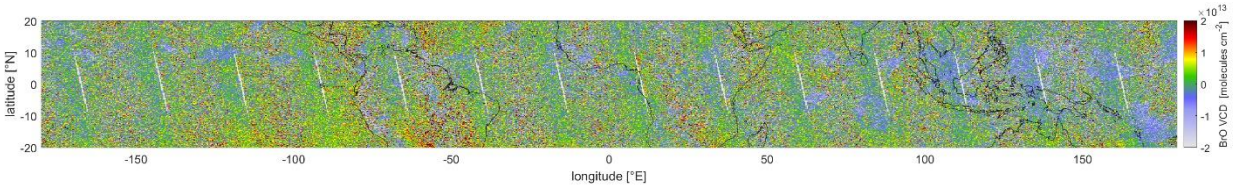
molecules cm^{-2} which is also at least 1.5 orders of magnitude lower than typical volcanic signals (which are on the order of $1x10^{13}$ molecules cm^{-2})."

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

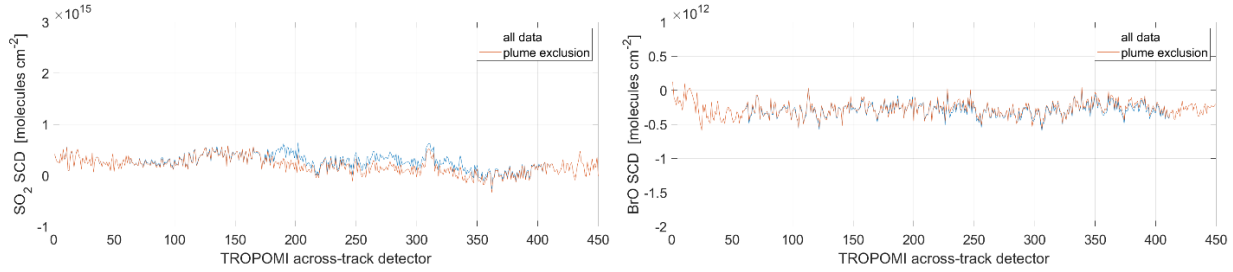
(a)



(b)

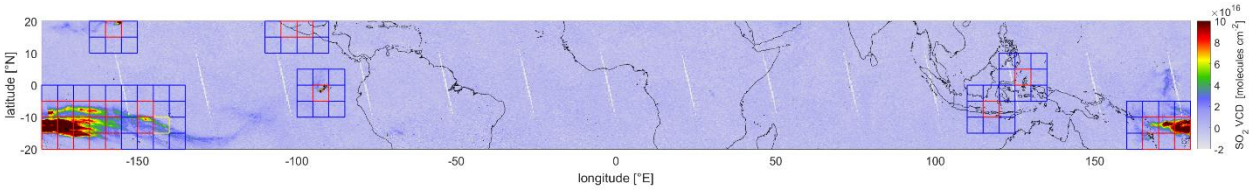


(c) (d)

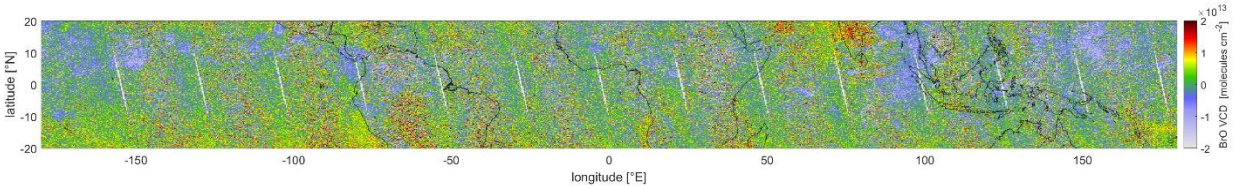


"Figure A1: (a) Map of the SO_2 VCD for the equatorial region on 2 October 2021. The areas of enhanced volcanic signals are marked in red squares. (b) Map of the BrO VCD for the equatorial region on 2 October 2021. (c) Mean SO_2 VCD for each across-track detector considering all pixel (blue) and excluding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

(a)

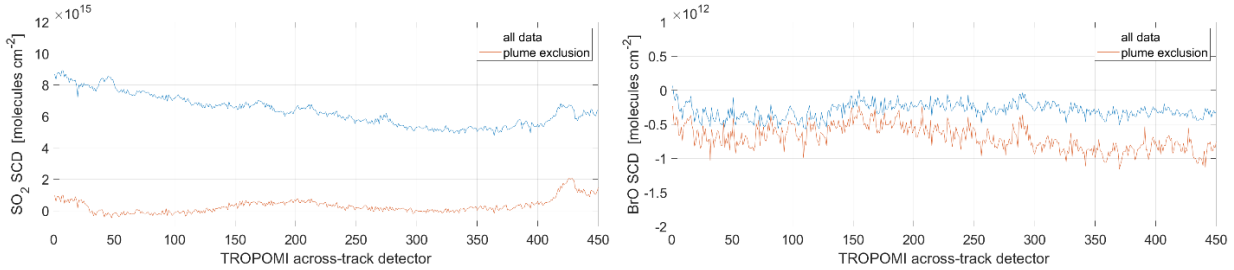


(b)



(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."

Regarding the reviewer's suggestion to include an uncorrected SO2 map of the equatorial region, we include one in the appendix together with the corresponding BrO statistics (cf. Fig. A1a and A2a).

GC2) Because of the importance of the auxiliary information required for the background correction scheme, namely O3 column densities and cloud parameters, it would be good to provide a brief description of how those parameters were derived.

We thank the reviewer for this comment, as indeed a detailed explanation of neither the O_3 VCD nor the cloud parameters is given in Sect. 2.1.2 where it would be most prominent. The cloud parameters are included in Sect. 2.1.2. Since this question overlaps with SC6, describe the changes to the manuscript in the answer to SC6.

The detailed description of the calculation of the O_3 VCD is given in section 4.2 in lines 304-314 ("Since there are two […] molecules^{-1}"). This text is now moved to Sect. 2.1.2 after line 135 and the reference to section 4.2 in Line 135 is removed.

The paragraph starting in line 134 now reads:

"Third, for the estimation […] is derived directly from the DOAS fit. Since there are two different O3 absorption cross-sections as well as two 'Pukite'-pseudo absorbers included in the fit (see Table 1) […] molecules-1). The resulting O_3 SCD is then converted to the geometric O_3 VCD following the formalism in Sect. 2.2."

Line 304f. now reads:

„In addition, the schemes ‚ozone' and 'ozone latitude' also include the O3 VCD taken from the BrO fit (cf. Sect. 2.1.2 for a detailed description of the calculation of the O3 VCD). The O3 VCD is then used in two of the three BrO correction schemes. "

[continuing in Line 316]. Furthermore, we added a reference to Sect. 2.1.2 in line 262, which now reads:

"The high-latitude example is depicted in Fig. 5, including the FRESCO CF (Fig. 5a) as well as the O3 VCD (Fig. 5c, taken from the BrO DOAS fit, cf. Sect. 2.1.2 for a description of the calculation of the O3 VCD) […]"

GC3) The writing style could be more concise. There is room for improvement in avoiding the repetitive, and a bit confusing introduction of what is going to be presented on each section with reference to what has been presented before. Instead of saying for example "in this section we will do X, using what was done in Sect. Y", I suggest presenting directly the new step without going back to the previous steps. The introduction to the sections, as done in the manuscript, adds little to readability. Shortening this will improve the flow of the text.

We thank the reviewer for this comment and agree that the introductions can be shortened.

We therefore changed the introduction of Sect. 4 (lines 236-241) to read as follows:

> "In this section, the systematic influences of following three effects onto the BrO retrieval are investigated and discussed for the eight different BrO fit settings: Potential systematic influences of O3, influences of clouds (Sect. 4.1), and the spectral interference with HCHO (Sect. 4.3). "

At the beginning of Section 4.3 we removed in line 383: "This section is looking at the interference between HCHO and BrO for the eight BrO DOAS fits (defined in Sect. 3)." And changed the beginning of the following sentence to: "The response of the eight different BrO DOAS fits […]"

In line 437, we changed the paragraph to:

> "In order to quantify the uncertainty originating from remaining systematic uncertainties onto the retrieval, i. e. the potential 'false' BrO signal created by clouds, O3 or other systematic effects, we look at the BrO VCD of the fit SR 323 – 360 nm (after the correction with the 'ozone latitude' correction scheme)"

In Line 508, we changed it to:

> "In order to give an estimate for the total error under all the various cases and locations where volcanic plumes occur, three latitude regions are distinguished:"

Line 531 is changed to:

> "In order to accurately retrieve BrO inside volcanic plumes, potential remaining local background gradients have to be accounted for. For this information on the SO2 signal is used to mask the volcanic plume signal and derive a local background correction."

By presenting concrete suggestion on optimal retrieval parameters for volcanic BrO from TROPOMI, this article can make an important contribution to operational global retrievals on volcanoes, complementing the existing capabilities of TROPOMI to detect SO2 from even weak volcanic plumes.

We thank the reviewer again for his/her general comments, which have raised valid questions and suggestions, whose answers substantially improved the manuscript.

Specific comments (SC)

SC1) The title is a bit awkward. It indicates to present a retrieval of volcanic BrO from a satellite sensor. It should instead refer to an algorithm to retrieve volcanic BrO *column densities* from satellite sensor *data*. The authors may judge if the algorithm is only valid to this specific satellite sensor.

We thank the reviewer for this most relevant remark. The title can indeed be more precise and specific in what is measured and used. We therefore change the title to:

> "A new accurate retrieval algorithm of bromine monoxide columns inside minor volcanic plumes from Sentinel-5 Precursor/TROPOMI observations"

Furthermore, it is indeed important to indicate that the retrieval can in principle be also be applied to other satellite sensors. We therefore expand the last partial sentence of the abstract ", which allows […] bromine-rich volcanoes." with:

> ". We present a new and accurate retrieval algorithm of BrO columns from TROPOMI observations, which allows for the detection of even slightly enhanced BrO amounts inside minor eruptive plumes of bromine-rich volcanoes. While designed specifically for TROPOMI observations, the retrieval algorithm is in general also applicable to other hyperspectral satellite observations. However, some parts might require adaptation."

We add in the introduction after line 94:

> "Even though our BrO retrieval algorithm is designed for TROPOMI observations, the improvements tested are – aside from satellite specific adaptations – in principle applicable to all hyperspectral satellite observations."

At the end of the conclusion (line 650), we add the following:

> "Even though our BrO retrieval algorithm is designed for TROPOMI observations, the improvements proposed here are in principle applicable to the BrO observations inside volcanic plumes from any hyperspectral satellite observations. However, satellite specific adjustments might be required."

SC2) The abstract could be shortened by reducing the information on the first two paragraphs to concentrate on the contribution of the study.

We changed the two paragraphs to read as follows:

> "Bromine monoxide (BrO) is a key radical in the atmosphere, influencing the chemical state of the atmosphere, most notably the abundance of ozone (O3). Ozone depletion caused by the release of bromine has been observed and modeled in polar regions, salt pans, and in particular inside volcanic plumes. Furthermore, the molar ratio of BrO and SO2 – which can

be detected simultaneously via spectroscopic measurements using the Differential Optical Absorption Spectroscopy (DOAS) method - is a proxy for the magmatic composition of a volcano and potentially an eruption forecast parameter.
The detection of BrO in volcanic plumes from satellite spectroscopic observations is limited by the precision and sensitivity of the retrieval, which so far only allowed for the detection of BrO during major eruptions. The unprecedented spatial resolution of up to 3.5x5.5km2 and high signal-to-noise ratio of the TROPospheric Monitoring Instrument (TROPOMI) onboard Sentinel-5 Precursor (S-5P) enables to observe and monitor volcanic bromine release globally even for minor eruptions or even quiescent degassing."

SC3) Expand on importance of measuring BrO. How much can O3 be destroyed and by which mechanisms? How the columns of BrO or column ratios of BrO respect to SO2 can help to understand volcanic processes?

We expand the second sentence of the introduction "Volcanic bromine […] ten-year period (Brenna et al., 2019).". It now reads as follows:

"Also Volcanic bromine release is thought to affect atmospheric O3 chemistry (Bobrowski et al., 2003; von Glasow, 2010; Surl et al., 2021). However, O3 measurements inside volcanic plumes are limited and the strength of the O3 destruction of volcanic bromine is still not fully clear (an estimation can be found in the review by Surl et al., 2015). Recently, the development of chemiluminescence instruments for the detection of O3 inside volcanic plumes show promising results (Rüth, 2021; Bräutigam, 2022) and could help to better constrain the volcanic O3 destruction, as they allow interference-free measurements of O3 in volcanic plumes, in contrast to the standard UV-O3 monitors often used today. Another important aspect is the influence of bromine on the stratospheric O3 chemistry. Strong eruptions ejecting bromine into the tropical stratosphere could potentially impact global stratospheric O3 abundance over a ten-year period (Brenna et al., 2019)."

In addition, we change lines 35-38 "and the BrO/SO2 ratio […] Warnach et al., 2019)." to now read the following:

". Ground-based (Bobrowski and Platt, 2007; Gutmann et al., 2018), as well as satellite observations (Hörmann et al., 2013) report inter-volcanic variations of the BrO/SO2 ratio of three orders of magnitude, strongly suggesting a link to differences in the geological settings of the individual volcanoes (Platt et al., 2015). Additionally, variations of the BrO/SO2 over time have been attributed as a proxy for changes in the volcanic systems, for instance at Mt. Etna, Italy (Bobrowski and Giuffrida, 2012), Nevado del Ruiz, Colombia (Lübcke et al., 2014), Cotopaxi, Ecuador (Dinger et al., 2018) and Tungurahua, Ecuador (Warnach et al., 2019). These variations are suggested to be linked to differences in the partitioning of bromine and sulphur from the magmatic melt, i. e. that bromine and sulphur are released at different depths below the surface. However, the interpretation remains difficult, as the partitioning of bromine from the melt is not yet well-constrained."

We added the following references:

Bräutigam, E.: Construction of an airborne chemiluminescence ozone monitor for volcanic plumes, Bachelor thesis, Heidelberg University, https://doi.org/10.11588/heidok.00032085, 2022.

Platt, U. and Bobrowski, N.: Quantification of volcanic reactive halogen emissions, p. 115–132, Cambridge University Press, https://doi.org/10.1017/CBO9781107415683.011, 2015.

Rüth, M.: Characterisation of a chemiluminescence ozone monitor for volcanic applications, Bachelor thesis, University Heidelberg, https://doi.org/10.11588/heidok.00029947, 2021.

Surl, L., Donohoue, D., Aiuppa, A., Bobrowski, N., and von Glasow, R.: Quantification of the depletion of ozone in the plume of Mount Etna, Atmospheric Chemistry and Physics, 15, 2613–2628, https://doi.org/10.5194/acp-15-2613-2015, 2015.

And removed the following references:

Bobrowski, N., von Glasow, R., Giuffrida, G. B., Tedesco, D., Aiuppa, A., Yalire, M., Arellano, S., Johansson, M., and Galle, B.: Gas emission strength and evolution of the molar ratio of BrO/SO2 in the plume of Nyiragongo in comparison to Etna, Journal of Geophysical Research: Atmospheres, 120, 277–291, https://doi.org/10.1002/2013jd021069, 2015.

Kern, C., Sihler, H., Vogel, L., Rivera, C., Herrera, M., and Platt, U.: Halogen oxide measurements at Masaya Volcano, Nicaragua using active long path differential optical absorption spectroscopy, Bulletin of Volcanology, 71, 659–670, https://doi.org/10.1007/s00445-008-0252-8, 2009.

SC4) The description of the DOAS method supported by Eq. 1 misses the essential feature of separation between high- and low-frequency components of the optical depth, that characterizes this method.

We thank the reviewer for noticing this. 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, e. g. Rayleigh and Mie scattering.”

SC5) Explain carefully the motivation to use the entire equatorial Earth-shine band (at all longitudes) for the background “reference” correction. This region includes quite different albedo regions (land, ocean), and many sources of BrO. The motivation of this choice is far from obvious.

This question is included in GC1) and addressed/answered in detail there.

SC6) Provide essentials of the FRESCO algorithm to obtain cloud height and fraction products used for the sensitivity study.

We changed the sentence “Both data-sets are calculated […]” in line 121 to:

> “Both data-sets are calculated using the Fast Retrieval Scheme for Clouds from the O_2 A-Band (FRESCO, Compernolle et al., 2021) algorithm, which derives a radiometric cloud fraction and cloud pressure using the reflectance spectrum at 760nm (in the O_2 A-band) assuming a Lambertian cloud model. The FRESCO data products are provided within the NO2 operational product (van Geffen et al., 2021, NO2, 2021).”

SC7) Provide essentials of the algorithm used to retrieve the HCHO product used for the sensitivity study. In particular, refer to how BrO interference was treated in such retrieval or if it was neglected?

We expanded the paragraph starting in line 132 to:

> “Second, for the study of the influence of HCHO onto the BrO retrieval, the HCHO slant column densities (SCDs) -- provided within the operational TROPOMI HCHO L2 product -- are used (De Smedt et al., 2018). The HCHO SCDs are derived from a DOAS fit in a similar fit wavelength range as BrO (328.5-346nm). In order to minimize interference with BrO, the BrO SCD is fixed using a SCD derived in an independent pre-fit in a larger fit wavelength range (328.5-359nm) (De Smedt et al., 2018).”

SC8) Section 6.1 introduces SO2 measurements, here some of the acronyms are spelled out for the first time. The first two paragraphs of this section could be moved to the Introduction.

We moved the first two paragraphs of section 6.1 (lines 537-545) in the introduction after line 56. We additionally checked that all acronyms are spelled out at the first mention.

SC9) The spectral evaluation setup for SO2 could be included in Table 1.

We like to thank the reviewer for this suggestion and included the spectral evaluation setup for SO2 in table 1 by adding a column “incl. in SO2 fit”. In addition, we included the phrase “All the fit parameter included are noted in Table 1 in the column ‘incl. in SO2 fit’.” in line 557.

Technical comments (TC, followed by line number)

TC1) L33- Spell out “sulphur dioxide” before chemical formula on first mention.

We changed the text as suggested.

TC2) L35- The payload is not designed to “determine” the composition, i.e., the instrument cannot determine the composition (this is determined by natural processes), but to measure certain properties of the atmosphere.

We replaced “determine” with “gain information on”. It now reads “[…] tool both to gain information on the composition of a volcanic plume […]”

TC3) L38- Define acronyms on first mention (GOME, SCIAMACHY).

We added the full designations for GOME & SCIAMACHY in line 38 and OMI in line 46.

TC4) L49- Correct spelling of “measurments”.

We changed it to “measurements”

TC5) L60- Use consistent notation for all molecular species, i.e. “O3” instead of “ozone”.

We changed this throughout the manuscript, except when talking about the “cloud ozone correction scheme”

TC6) L63- What is lower altitudes and higher latitudes? Better to indicate “tropical”, “mid-latitude” etc., or even better to tell percentage of active volcanoes within +-30 deg, to back up this assertion.

We changed the latitude designation to “tropical, mid, and high latitudes” to now read the following:

> “Second, volcanic plumes mainly occur at tropical latitudes, where O3 columns and the SZAs are considerably lower compared to mid- and high latitudes. Third, the use of an earthshine reference spectrum recorded around the equator reduces the optical density of O3 to zero at the equator, further reducing a potential O3 interference at tropical latitudes.”

TC7) L81- Add “typical” before “volcanic BrO columns of small eruptions”.

We added it accordingly.

TC8) L83- Define “VCD” on first mention.

We thank the reviewer for noticing this. As it is not needed to talk about VCDs in the introduction, and on each other instance in the introduction we talk about “columns” we changed it to “column” in this line as well in order to increase readability.

TC9) L103- Use upper case for the name “Precursor”.

We changed it accordingly.

TC10) L112- Correct spelling of “characteristica”.

We changed it to “characteristics”.

TC11) L113- Better to write “Selection of spectra” instead of “spectra… used” as sub-section title.

We thank the reviewer for this comment. We changed the wording to “Selection of spectra: TROPOMI UVIS”

TC12) L125- Use upper case and complete name for “Sentinel 5-Precursor Expert Users Data Hub”.

We changed it accordingly.

TC13) L174- Correct “far off”.

We changed it to “far-off”

TC14) L229- Define “dSCD”.

We added the abbreviation “(dSCD)” in line 180, it now reads “differential SCDs (dSCDs)”, where it is first introduced.

TC15) L243- Use upper case for “Pacific”.

We changed it accordingly

TC16) L435- Better to use the noun-phrase “Investigation of systematic…” instead of the continuous verb form “Quantifying the systematic…” in the title of a sub-section.

Many thanks for this suggestion! We prefer to use the noun-phrase “Quantification of the systematic effects”, as the title “investigation of systematic effects” is already the title of the section (cf. line 234).

TC17) L472. Similar than previous comment.

Also here we prefer to use the noun-phrase “Quantification of the statistical uncertainties”. The reasoning is the same as in TC16.

TC18) L577- Does the value indicated as typical for TROPOMI correspond to one or four standard deviations of SO2? And is this the standard deviation of the residual?

The value 2×10^{16} molecules cm^{-2} corresponds to four times the std. dev. The std. deviation refers to the std. dev. of the SO_2 background distribution. We clarified this in the text by changing the sentence to:

> “This is chosen in this study as four times the standard deviation of the SO_2 background distribution (for TROPOMI the standard deviation is typically in the range of 5×10^{15} molecules cm^{-2}, i.e. the threshold is in the range of 2×10^{16} molecules cm^{-2} […]”

Figures

Fig. 1) Indicate which references cross sections were used before convolution (reference to authors), preferably as a legend or caption to the figure.

We added them in the caption, which now read as follows:

> "Absorption cross-section of BrO (blue, based on Fleischmann et al., 2004) and HCHO (red, based on Meller and Moortgat, 2000) convolved with a typical TROPOMI instrument spectral response function."

Fig. 2) Add labels to y-axes.

We included them.

Fig. 3) Reference to Fig. 4 and Fig.5 in the figure description is not appropriate because one needs the three figures to understand the meaning. Better to explain briefly the reason of the two regions, e.g., with relation to the study of the cloud and stratospheric O3 interferences.

We changed the second sentence in the caption to:

> "The areas in the equator and mid-latitude region (marked by red rectangles) are used as case studies to investigate and quantify the effect of clouds and O3 respectively (see section 4.1)."

Fig. 4 (and all following maps) Add units of "deg" to all axes showing lat and lon.

We changed it for all figures to "latitude [°N]" and "longitude [°E]" respectively.

Fig. 5) The O3 background varies very drastically with latitude. It would be good to discuss if the reason for this steep gradient can be found in terms of the general features of O3 dynamics (e.g., presenting a plot of typical O3 latitudinal gradients for comparison).

In the case presented here, the gradient in O3 originates from a change occurring on the boundary of the polar cell and the Ferrel cell, e.g. polar air masses north of 50°N and mid-latitude air masses south. Firstly, the polar jet stream defines this boundary. Secondly the tropopause height decreases in the polar cell.

The jet stream can be identified by a strong wind pattern in roughly 9km altitude. As can be seen in the wind speed at 9km (cf. fig. A1) such a strong wind pattern occurs directly on the edge of the O3 gradient, indicating that the gradient occurs at the location of the jet stream and hence at the boundary of polar and mid-latitudinal air-masses. This is strengthened when looking at the height where the potential vorticity is 2 potential vorticity units (10-6 m^2Ks^{-1} kg^{-1}) – which is one

definition of the tropopause height. This height and hence the tropopause height is decreasing from 12 km south of 50°N to 8km north of 50°N in congruence with the O3 gradient (cf. Fig. A2).

We made the following changes within the manuscript:

In line 264 we write instead of "This follows the gradient line of O3 along the same latitude, indicating that this increase is most probably due to a higher stratospheric column." The following:

> "This follows the gradient line of O3 along the same latitude (cf. Fig. 5c). Both BrO and O3 gradients coincide with the transition from mid-latitude to polar air masses indicated by the presence of the jet stream and a change in the tropopause height (cf. Fig B1), where O3 is higher and the stratospheric column is increasing."

We added the section 'High-latitude air-masses influence on BrO and O3' in the appendix including the following text and figure of the tropopause height and wind speed at 9km:

> "In order to interpret the coinciding gradients of O3 and BrO in Fig. 5 with respect to meteorology, we look at the location of the jet stream estimated as band of strong wind-speed at 9 km altitude and the tropopause height estimated as the height where the potential vorticity is 2 potential vorticity units (PVU). The corresponding data are taken from ECMWF ERA-5 model data at 12 am UTC for the respective region and is depicted in Fig. B1."

We added the following figure in this section of the appendix:

(a) (b)

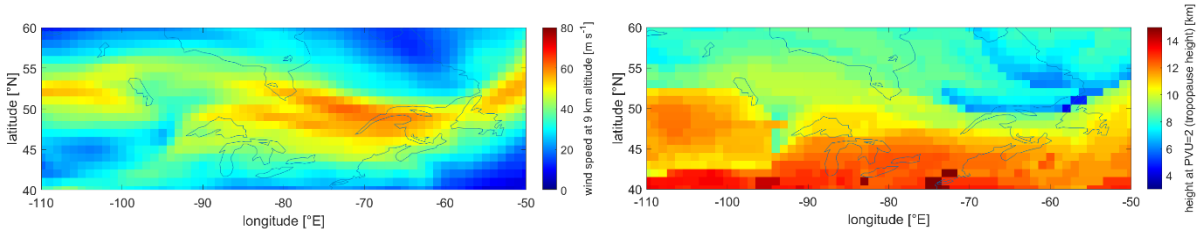


Figure B1: Meteorological conditions for the high-latitude case (cf. fig. 5) on 1 October 2018, taken from ECMWF ERA-5 data at 18:00h UTC: (a) the wind speed at 9km indicating the presence of the jet stream and (b) the tropopause height indicated by the height where the potential vorticity is equal to 2 potential vorticity units.

Fig. 6) This sequence of figures, presented in the sensitivity studies, is difficult to understand. It would help to add text in the caption to guide the reader towards a conclusion. The pattern cannot be understood at first glance. What complicates matters is that the scales for VCD are different.

We added the following explanation to the caption:

> "The almost linear influence of both cloud height and cloud fraction is clearly visible for tropical latitudes (middle row). This changes for mid- to high latitudes (upper and lower row), where other influences related to the stratospheric column independent of the cloud parameters can also be seen."

We realize that the different VCD scales complicate the matter and we thank the reviewer for addressing this point. We see the need that this should be addressed somehow. However, as the centers of the different colormaps deviate by 3.5×10^{13} which is almost twice the range of the current color scale (2×10^{13}), using a single color scale for all subplots would drastically reduce the visibility of the effects in question. We therefore decided against a universal color scale and added the further line to the caption:

> "Note the different color scales for each subplot. They always encompass the same range of 2×10^{13}, but start at different BrO VCDs."

Tables

Table 1) "Shift and squeeze" and "ISFR" should be classified as instrumental corrections and not "pseudo absorbers". Define "ISFR" and its parameters in the table's description or as a footnote.

We moved the ISRF and shift & stretch to a new subsection named "Pseudo absorbers for instrumental effects" and defined ISRF and its parameters in two footnotes.

Table 3) It could be limited to include only new information not presented already in Table 1.

We removed this table, 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).

Lastly, 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."