We greatly appreciate the comments and queries from the reviewer. We were able to enhance the scientific quality of our manuscript significantly by incorporating the reviewer's suggestions during the revision. We tried our best to accommodate all the comments in the revised manuscript. Our answers to the comments and questions are written below in blue. Specific revisions made in the manuscript are underlined.

RC2

In the paper titled "Global Retrieval of Stratospheric and Tropospheric BrO Columns from OMPS-NM on the Suomi-NPP Satellite," Chong and colleagues introduce a new BrO column product derived from the OMPS-NM satellite instrument. The authors focus on presenting a long-term time series of tropospheric columns while effectively distinguishing between stratospheric and tropospheric contributions. The paper's strength lies in its innovative approach, combining the strengths of two state-of-the-art methods to separate stratospheric and tropospheric columns. Furthermore, the long operational lifespan of the OMPS-NM instruments lends a distinct advantage to this product, promising continuous data acquisition over an extended period which allows long-term time series of this chemically important tracer. This BrO column product makes a valuable addition to existing satellite-derived BrO products, with potential benefits for bromine chemistry research and is therefore in good alignment with the scope of AMT. I strongly recommend considering this paper for publication after addressing the noted corrections and points.

General comments:

GC1) The manuscript appears to primarily target polar applications, with retrieval considerations and examples predominantly centered around polar regions. For instance, the effective application of the flattening technique to polar hot-spots raises questions about its performance in non-polar tropospheric enhancements, confer also GC2. Moreover, the wavelength criterion selection (Page 39, line 6-7) implies a concentration on polar applications, aiming to achieve optimal results under high latitude and SZA conditions. While this focus is justified, I suggest to explicitly mention in the abstract, introduction, and potentially the title. Consider either highlighting the emphasis on polar BrO retrieval or substantiating why polar regions pose the greatest challenge.

→ It is a valid point that a significant portion of the retrieval considerations and examples targeted polar regions. In the revised manuscript, <u>we have added sentences highlighting the emphasis on polar BrO retrieval to the abstract and introduction</u>. However, we didn't change the title as the retrievals have global coverage. <u>To incorporate the reviewer's comment, the fitting window optimization has been updated to cover non-polar regions </u>(which will be discussed in the later part of this document).

GC2) Even though it is maybe a bit overcomplicated, I generally like the authors approach for the separation of tropospheric and stratospheric columns as it combines the two pathways taken in previous studies. However, there are several steps where I can see potential issues:

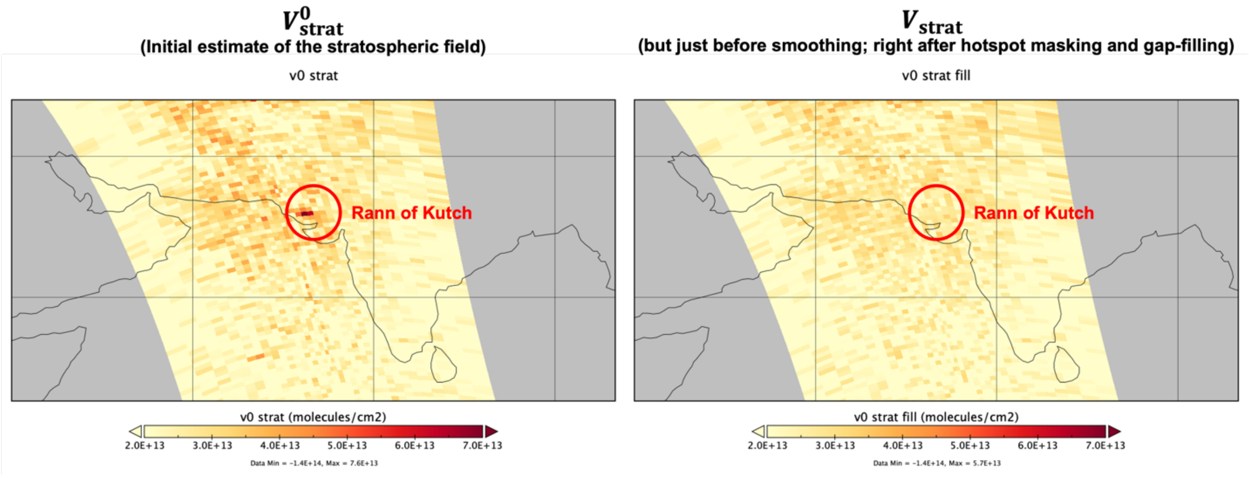
1. How does step (i), flattening the tropospheric model profile (page 17, Figure 1), perform with non-polar tropospheric enhancements? Is it specifically tailored for this scenario? For instance, if an extensive area of BrO tropospheric enhancements emerges in the equatorial Pacific due to an unusual climate change event or a significant volcanic plume, how would step (i) handle this situation? It appears that step (i) might overlook such occurrences, possibly leading to their inclusion in V_strat_0 (which may not be problematic by itself). However, a concern arises regarding the effectiveness of the O3-BrO relation-based separation (Figure 4c) under such circumstances.

→ First of all, we'd like to clarify that the flattening technique aims at non-hotspots rather than hotspots. We use flattened profiles (without tropospheric enhancement) for non-hotspots while using modeled profiles (with potential tropospheric enhancement) for hotspots.

We apologize if the description was not clear in the initially submitted manuscript. In the revised manuscript, we have added more sentences to the part where we describe the flattening technique in Sect. 2.2.4 (Sect. 2.5 in the revised manuscript). Also, we have added Appendix B to the revised manuscript to present more details about the flattening. (Appendices B and C in the initially submitted manuscript are C and D in the revised manuscript.)

Regarding the reviewer's questions, the flattening technique was designed for global applications. As described in the manuscript, the retrieval algorithm stores both pre- and post-flattening profiles for every pixel. Then, the algorithm constructs V_{strat}^{0} by subtracting the post-flattening tropospheric column ($V_{\text{trop}}^{\text{flat}}$) from the total column. If a tropospheric enhancement emerges at a certain pixel, $V_{\text{trop}}^{\text{flat}}$ is supposed to be smaller than the actual tropospheric column. Therefore, this pixel must appear in the V_{strat}^{0} field as a hotspot, just as intended. Then, this pixel is removed from the V_{strat}^{0} field, using the O_3-BrO relation. This process doesn't discriminate between polar and non-polar tropospheric enhancements. For example, the hotspots in Rann of Kutch are well detected and masked, as shown in the figure below (for March 13, 2017).

Although this figure has not been added to the revised manuscript, we have added a description to clarify that the flattening step applies globally.



V_{strat}^{0}
(Initial estimate of the stratospheric field)

V_{strat}
(but just before smoothing; right after hotspot masking and gap-filling)

2. Page 19: In step (v) the authors suggest to use the model profile as input for AMF calculation under no hotspot conditions and in difference the “flattened” model profile as input for the AMF calculation under hotspot conditions. The validity of this approach does not occur to me as I have the following concerns/points.

→ As we described above, it’s the opposite. We use the model profile under hotspot conditions and the flattened profile under no hotspot conditions.

3. How confident are you, that this model profile is representative for the tropospheric profile under “no-hotspot” conditions? Please shortly address this in the manuscript.

→ We don’t use the model profiles under “non-hotspot” condition. We use flattened profiles in that case to avoid the potential mismatch between the model and the observation. During the revision, we added Appendix B to present more details about the flattening scheme.

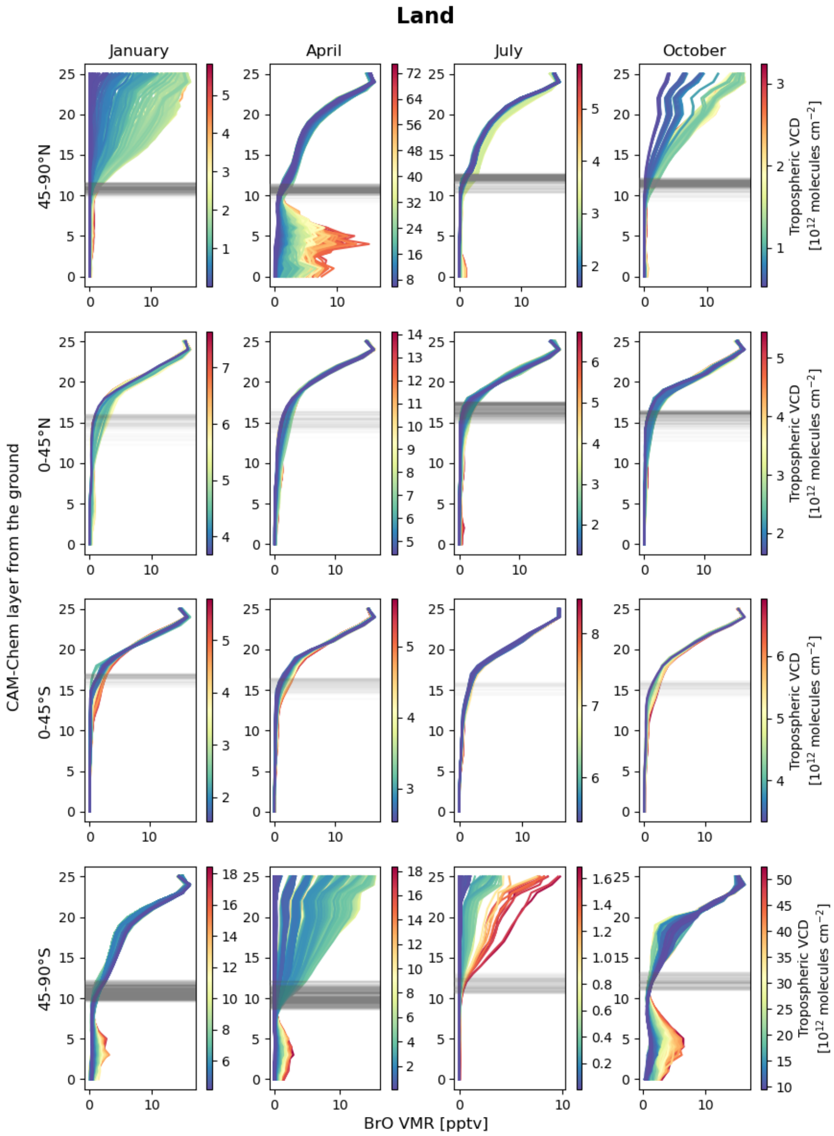
4. For hotspot cases, the authors opt for the flattened tropospheric profile. While not explicitly stated, it seems this choice stems from the model profile being decidedly unsuitable for such scenarios. However, employing a flattened profile could potentially worsen the situation. This selection could yield a lower and possibly excessively low tropospheric AMF (A_trop_select, denominator in equation 12). Considering the high S_trop (numerator in equation 12), this might lead to an excessively high V_trop. This concern is particularly relevant when contrasting with non-hotspot pixels where a non-flattened profile is used for tropospheric AMF calculation. Consequently, I have three queries/suggestions regarding the treatment of hotspot cases:

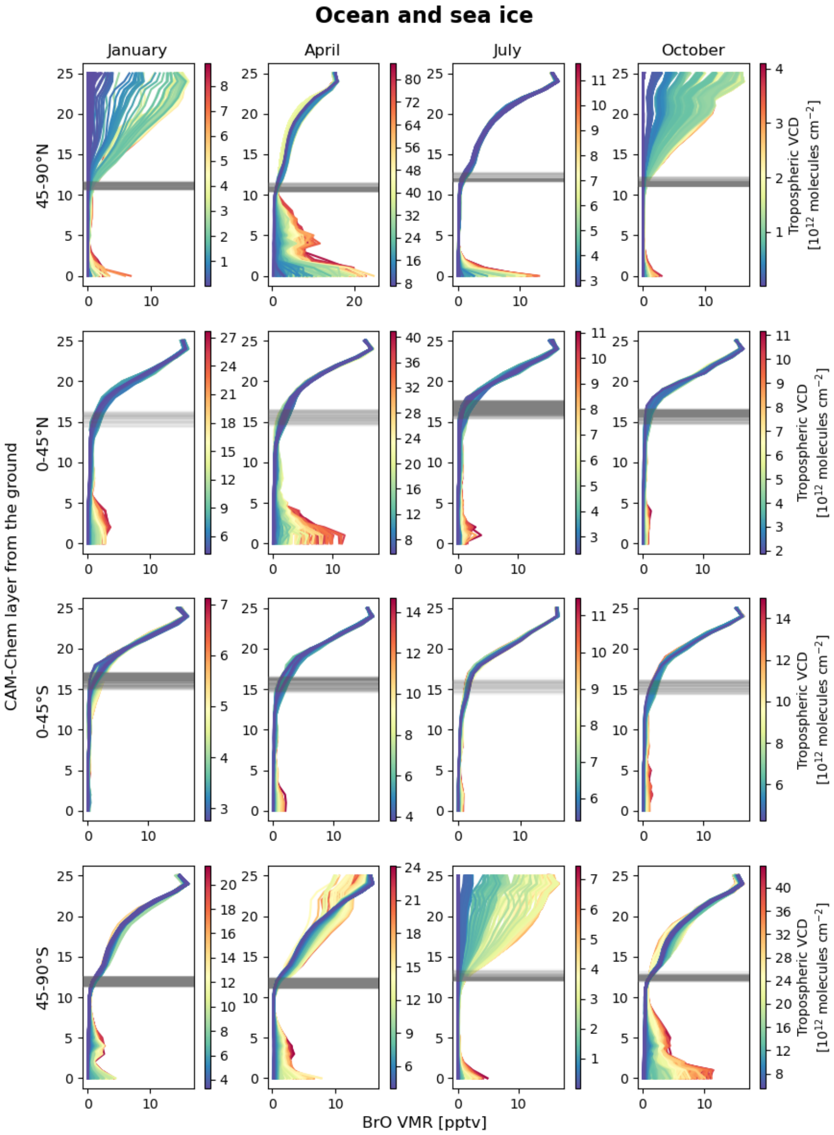
→ As discussed above, the usage of pre- and post-flattening profiles is the opposite of the description in this comment.

4.1. Is there a justification for favoring a flattened profile over the model profile as an assumption?

→ For the justification, we have added the two figures below to Appendix B of the revised manuscript (Figs. B1 and B2). The upper figure shows the variations in the tropospheric profile shapes depending on tropospheric VCD values over land. The data shown here were sampled from the CAM-Chem climatology to represent 13:30 local solar time. For January, April, July, and October, we calculated the mean tropopause pressure (P_{trop}) values for each 45° latitude band. Then, for each month and each latitude band, we sampled BrO profiles from the model grid cells having P_{trop} values within ± 20 hPa from the mean. The sampled profiles are presented in the figure below for each month/latitude band. Every profile curve is color-coded using the tropospheric VCD value. The gray horizontal lines represent the tropopause. The lower figure shows the same as the upper one but for the ocean and sea ice regions.

As stated, the purpose of the flattening technique is to simulate vertical profiles that represent the background conditions. As shown in the figures below, higher tropospheric VCDs tend to have more complex vertical structures. On the other hand, the lowest (background) VCDs typically have flat profiles with a decreasing pattern of BrO volume mixing ratios (VMRs) from the tropopause toward the ground. Based on this characteristic of the modeled tropospheric BrO VMRs, we employ the flattening approach to generate vertical profiles exhibiting gradually decreasing (or constant) BrO VMRs from the tropopause toward the ground.





4.2.Given that AMF carries a larger share of random uncertainties, could it be more accurate and practical to assume a distinct "hot-spot" profile shape (e.g., employing a ground-level profile shape as in Fig. 3e for polar hot spots, and a different assumption for tropical cases)?

→ This comment seems to describe an approach similar to what we have been employing. We use modeled profiles (with potential tropospheric enhancement) for hotspots and flattened profiles for non-hotspots. However, we don't discriminate between polar and non-polar (or tropical) pixels when applying this approach. As described above, we prepare flattened profiles all across the globe and selectively use either a modeled or flattened profile on a pixel-by-pixel basis, depending on whether a tropospheric enhancement is detected or not. As shown above, in the case of Rann of Kutch, this approach works even for non-polar regions, as well as the polar regions.

4.3. Is this factor considered in the error propagation of the AMF? Given its potential significance, it would be beneficial to explicitly acknowledge this and its impact.

→ Yes, as a way of considering the impact, we perform the AMF uncertainty estimation separately for the flattened and non-flattened profiles. As described in the manuscript, we calculate partial derivatives of AMF after binning AMFs from 2015 according to six parameters. At this point, we perform the binning separately for hotspots and non-hotspots. This way, we calculate the partial derivatives independently for hotspots (with non-flattened profiles) and non-hotspots (with flattened profiles). Therefore, ultimately, their AMF uncertainties are independent of each other.

To clarify it in the manuscript, we have added a corresponding description to the beginning part of Sect. 2.3.2 (Sect. 2.6.2 in the revised manuscript).

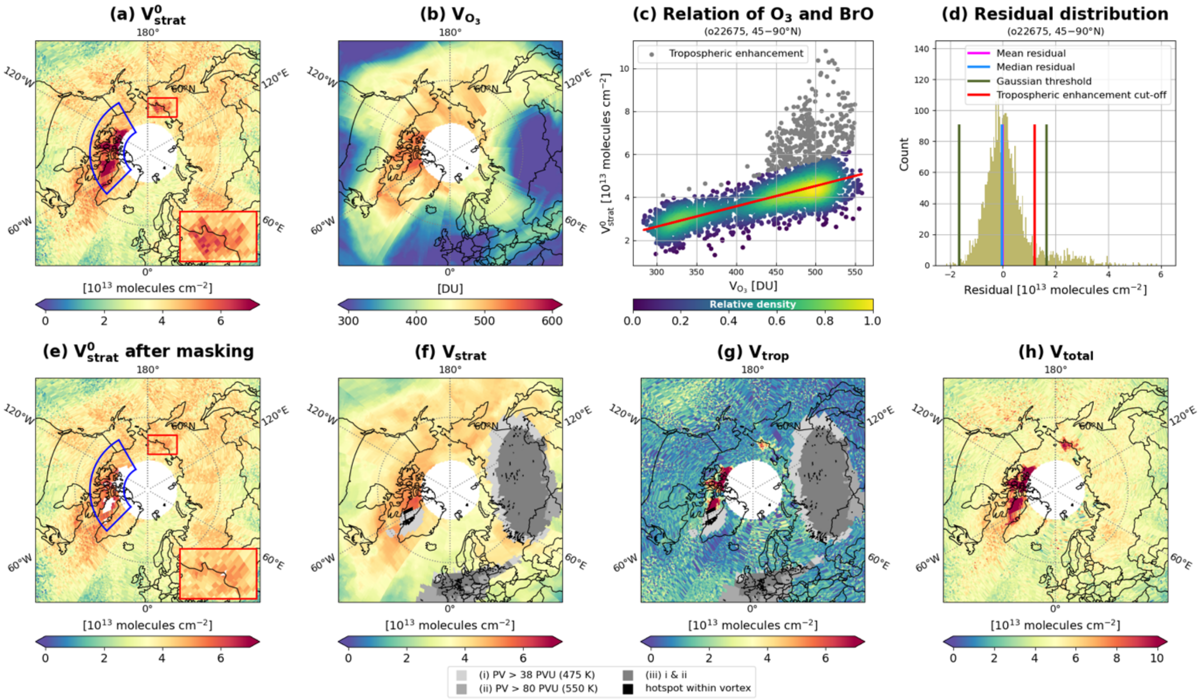
5. Page 19, line 13: Like Sihler et al. (2012), step (iii) employs the assumption of a constant O3/BrO relation to quantify tropospheric enhancements. However, the authors note that this assumption becomes problematic in polar vortex scenarios, potentially introducing bias to your data. Is this data utilized in the final product? If yes, provide rationale for its inclusion and discuss implications for the data quality flag.

→ To be precise, the issues are with hotspots detected in the polar vortex rather than all pixels within the polar vortex. That's because we perform only hotspot detection/reconstruction using the O_3 fields rather than constructing the entire stratospheric BrO field.

Nonetheless, we still use all data within the polar vortex in the analyses shown in the manuscript, including the hotspots, to give an idea of the general retrieval performance.

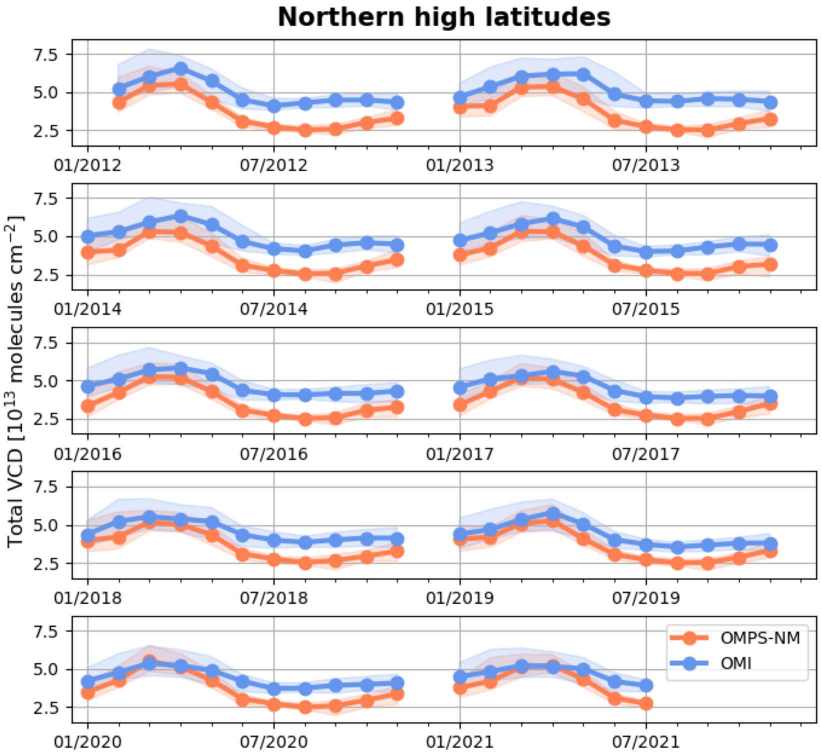
During the revision, however, we added a quality flag variable to the product for users who would like to filter out hotspots within the polar vortex. This variable is a three-digit binary. The first digit represents whether a hotspot is detected, and the second and third represent whether the potential vorticity is larger than a threshold at 475 K and 550 K potential temperature, respectively. The thresholds are 38 PVU (475 K) and 80 PVU (550 K) in the Northern Hemisphere, while those are –55 PVU (475 K) and –90 PVU (550 K) in the Southern Hemisphere. For this purpose, we used potential vorticity data from MERRA-2.

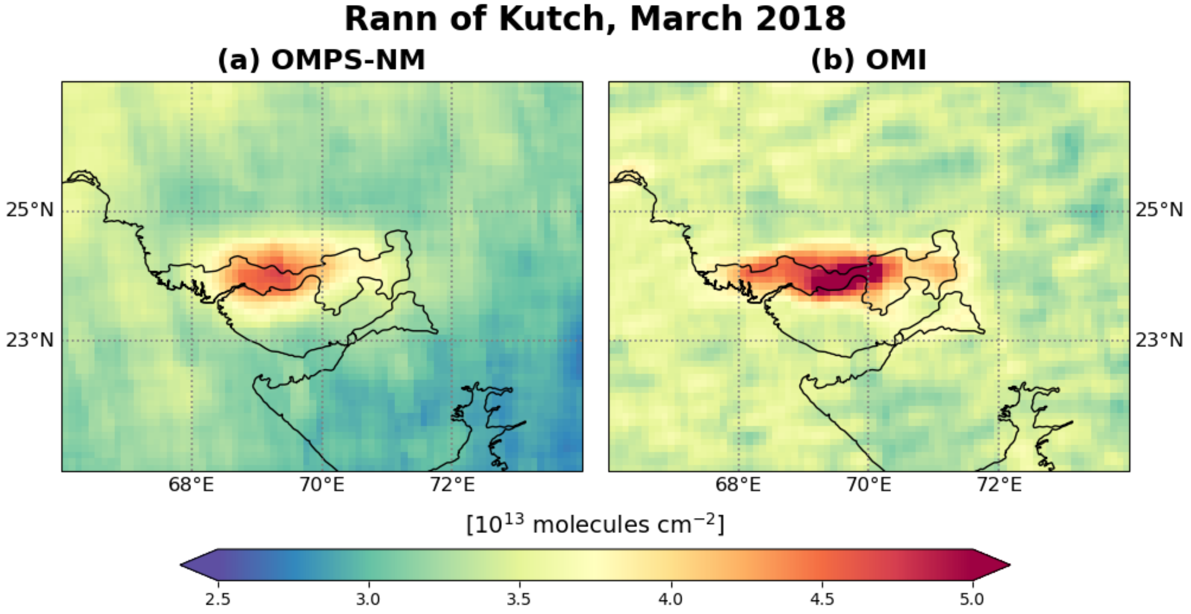
Figure 4 (Figure 5 in the revised manuscript) has been revised as below. Polar-vortex pixels are marked with gray colors in panels (f) and (g). Hotspots within the polar vortex are marked with black (see legend for details). In the revised text, we now recommend that users decide whether to utilize the hotspot data within the polar vortex based on their specific analyses and requirements. If they choose not to use them, it is recommended to filter out only black pixels in panels (f) and (g).



GC3) The paper would benefit from a comparison with other satellite studies. This does not need to be thorough, but differences and agreements should be adressed both with regard to the polar observations as well as the BrO from Rann of Kutch.

→ We have added the two figures below to the revised manuscript (Figs. 15 and 16). The first figure shows the comparison between OMI and OMPS-NM BrO total columns over the Northern high latitudes (60–90°N). The monthly variations agree well, demonstrating that OMPS-NM BrO retrievals are capable of extending the afternoon BrO time series. OMPS-NM BrO retrievals are typically lower than OMI, and that's likely due to differences in the retrieval algorithms. For example, the OMI retrieval algorithm uses different settings for the source spectrum (solar irradiance) and the fitting window (319.0–347.5 nm). The second figure presents the comparison of total BrO columns from OMPS-NM and OMI over Rann of Kutch for March 2018, demonstrating the consistency between the two data sets.





Specific Comments

Abstract:

The abstract could benefit from a clearer articulation of the paper's primary goal and focus. Consider adding a succinct sentence at the beginning of line 4 to outline the central theme of the study. This could lead into the subsequent statement, "To address this concern and improve upon the current methods, our study introduces..." This adjustment would help provide a smoother transition into the specific achievements and advancements discussed.

→ We have revised the abstract following the reviewer's comment.

Introduction:

Page 3, lines 10-29: The provided overview of the broad variety of separation schemes used in the literature is commendably thorough and informative. Nonetheless, its level of detail seems too thorough for an introduction. In the introduction, the focus should be on the paper's new method, and a concise acknowledgment of various approaches would suffice to put the paper's method into perspective

Given the absence of a designated "methods" section to accommodate such content as a subsection, I understand the authors' predicament in determining its placement. To address this, a practical solution could be integrating it as a subsubsubsection within Page 14, subsubsection 2.2.4.

→ Following the reviewer's comment, we have shortened the part where we presented the overview of various separation schemes in the introduction. Then, we added Sects. 2.2.4.1 and 2.2.4.2 (Sects. 2.5.2 and 2.5.3 in the revised manuscript). The former provides the overview, moved from the introduction, and the latter delivers the texts originally written in Sect. 2.2.4.

Page 3 line 35-page 4 line 5: The paragraph's primary emphasis, as underscored by the authors first lines, lies in the substantial potential of OMPS to provide an extensive and enduring time-series well into the 2030s. This aspect should take precedence, and should be highlighted in the paragraphs first sentence, shifting the focus imediately to this critical attribute. Accordingly, I suggest starting with the assertion about OMPS's long time-series capability, and subsequently incorporating the initial sentence, "OMPS-NM instruments ... decommissioning of TROPOMI (Nowlan et al., 2023)," at the paragraph's conclusion.

→ We have reorganized the paragraph as suggested by the reviewer.

Page 4, line 16-18 and fig. 1: There are two confusing aspects:

1. The use of the numeration (1) – (4) in reference to fig. 1, lets the reader look for the numbers 1-4 in fig. 1. However, the corresponding fields are noted with (A)-(D). Please use the same symbols in text and figure

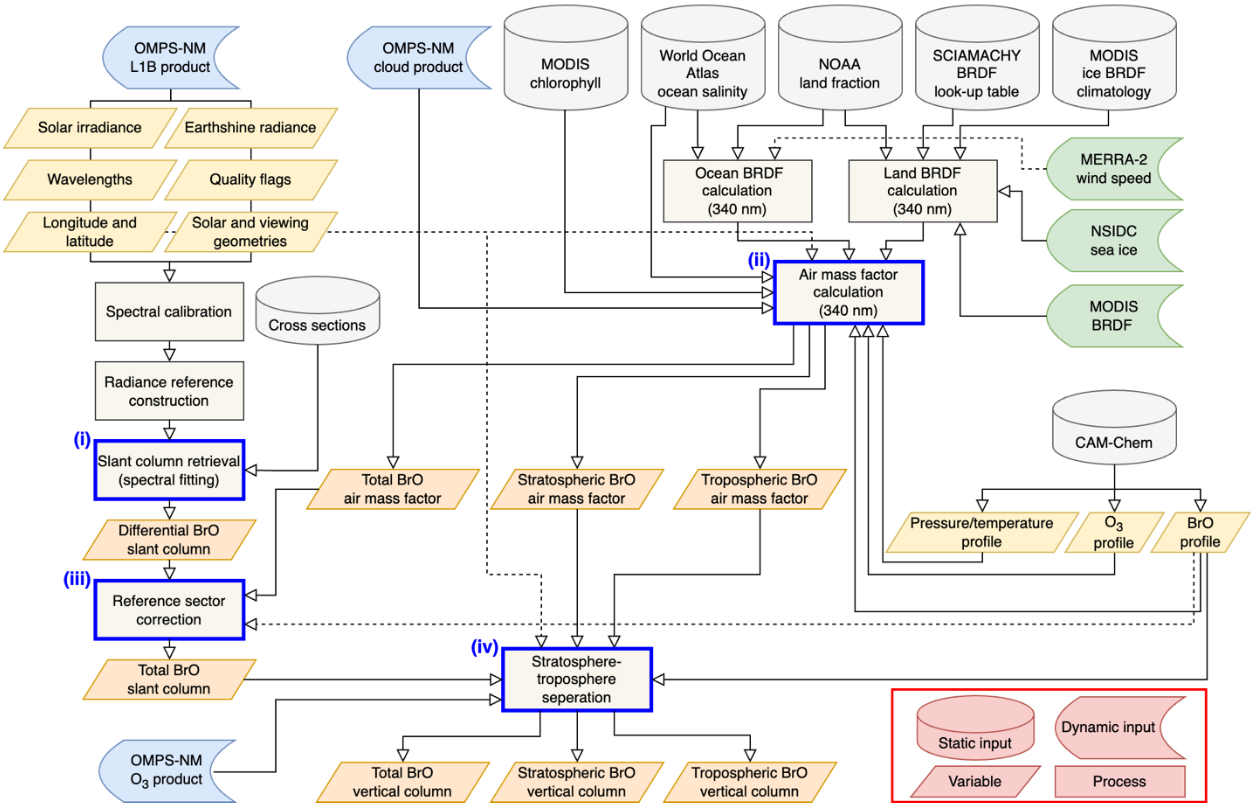
   → We have changed the symbols to (i)–(iv) both in Fig. 1 and the text.

2. When reading "highlighted in blue in fig. 1", the first look in figure 1 will be to the fields which have a blue background "OMPS-NM L1B product" etc. As Figure 1 contains a lot of information it is difficult to find the highlighted fields. Either specify in the text that they are "encircled/framed in blue" or reconsider the coloring within fig. 1 to avoid this

confusion. Also consider to increase the size of the border line to highlight the 4 fields. Furthermore, consider to place the A-D always at the same location w.r.t. the fields they refer to (either all on the top left or top right).

→ We have changed the wording to “framed in blue” and increased the size of the borderline. Also, we have placed the bullets (i)–(iv) at the same position (at the upper left corners).

The resultant figure is shown below.



Page 4, lines 16-18 and Fig. 1: This section presents two points of confusion:

1. The utilization of the numerals (1) – (4) in reference to Fig. 1 prompts readers to search for numbers 1-4 within the figure. However, the corresponding elements are actually labeled as (A)-(D). To enhance clarity, it's advisable to employ consistent symbols both in the text and the figure.

   → This appears to be a duplicate comment mirroring the one above.

2. When the text mentions "highlighted in blue in Fig. 1," readers instinctively turn their attention to fields with a blue background, such as "OMPS-NM L1B product," within Figure 1. Since the figure contains extensive information, locating the highlighted elements becomes challenging. To address this, you could specify in the text that the relevant fields are "encircled/framed in blue." Alternatively, reconsider the color scheme

> within Fig. 1 to alleviate this confusion. Additionally, consider enhancing the border line's size to accentuate the four designated fields. Furthermore, for consistency, contemplate consistently placing the labels (A)-(D) at the same relative position with respect to the corresponding fields (either all at the top left or top right).
>
> → This appears to be a duplicate comment mirroring the one above.

By harmonizing symbols and refining visual cues, these adjustments can substantially improve the reader's comprehension.

→ We agree. Thank you for the comment.

Page 4 and 6: The mention of 2 times "retrieval" in headline 2 and 2.2 is redundant, I suggest to move all the subsubsections in 2.2 up by one rank in hierarchy (e.g. 2.2.1-> 2.1, etc.).

→ We have revised the hierarchy following the reviewer's comment.

Page 6, line 6: The phrasing currently implies that Beirle et al., 2017; Nowlan et al., 2023 originated the super-gauss concept. I recommend revising it to "super Gaussian and adopt the approach outlined in Beirle et al., 2017; Nowlan et al., 2023."

→ We have revised the sentence as recommended.

Page 6, line 23: The authors specify their utilization of a 20° latitude portion within a single orbit for the computation of the earthshine reference spectrum. With an assumed along-track pixel footprint of 50 km, this approach implies that each across-track reference spectrum would be derived from 40-50 individual spectra. Notably, other investigations involving 2D CCD satellites like OMI and TROPOMI adopt larger sectors (e.g., Seo et al., 2018: 150°E – 240°W, 30°S-30°N for BrO; Theys et al., 2017: 120-160°W, 10°N-10°S for SO2). Please Justify the rationale behind employing a relatively compact reference sector and argue why such a low statistic is deemed satisfactory for your study.

→ The reason why we use a narrower latitude band is that we found spatial and temporal variabilities of BrO concentrations in the modeled data (CAM-Chem) over the Pacific. The wider the latitude band, the less representative the background SCDs are. Our objective here was to avoid having large spatial and temporal variabilities of BrO within the reference sector.

As mentioned by the reviewer, there are previous examples of using wider latitude bands (e.g., Gonzalez Abad et al., 2015, 2016; Nowlan et al., 2023; Seo et al., 2019; Theys et al., 2017). Except for Seo et al. (2019), who assumed a constant BrO VCD within the reference sector, those retrieval examples mostly targeted a species primarily residing in the troposphere (e.g., HCHO and SO_2) with minimized variations in total columns over the Pacific. For BrO, which resides in the stratosphere with significant and varying amounts, a narrow reference sector can be safe as we derive only a single background SCD to represent the entire sector.

After deciding to use a narrower reference sector, we considered the resulting SNRs of the averaged radiance reference spectra to determine the exact width of the sector. Given that there are retrievals employing solar irradiance data, it would be enough if we could generate a radiance reference spectrum with a comparable SNR. The SNR tends to be proportional to the square root of the input signal. Given that the intensity ratios between radiance and irradiance values are typically ~0.05 in the BrO fitting window, the SNR of an individual radiance spectrum would be smaller than that of irradiance by a factor of $\sim\sqrt{20}$. Considering that the noise (error) of an average value is inversely proportional to the square root of the number of samples, we need ~20 radiance spectra for averaging to achieve our goal. In the case of the OMPS-NM instrument, which has 50 km footprints, it corresponds to a ~10°-wide latitude band.

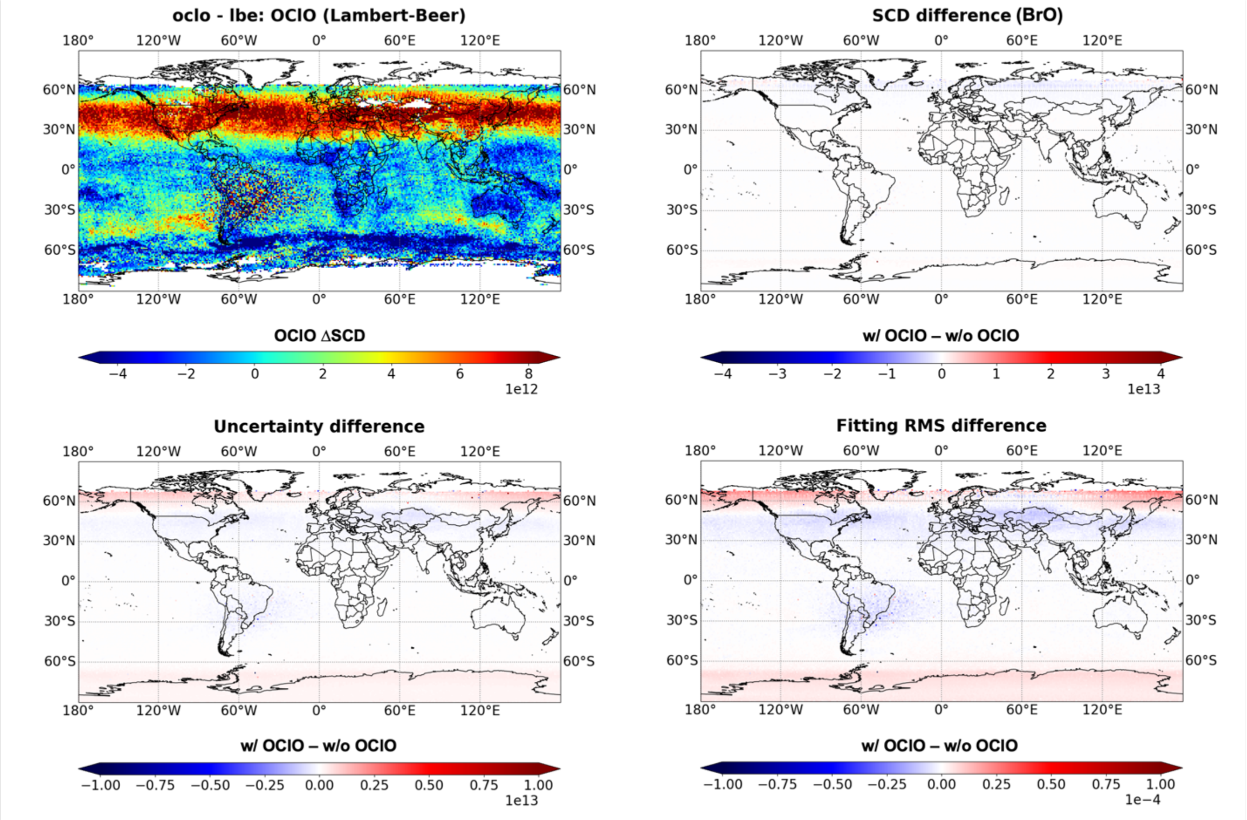
We have added a brief description of why we chose this reference sector to Sect. 2.2.1 (Sect. 2.2 in the revised manuscript).

Page 8, Table 1: Regarding the SCD retrieval: All (to the reviewers knowledge) recent other BrO DOAS and "DOAS-like" spectral retrievals include OClO in their spectral retrieval (e.g. Suleiman et al., 2019; Herrmanns et al., 2022; They et al., 2011; Sihler et al., 2012; Seo et al., 2019). Please justify your choice not to include it especially with respect to the potential spectral interferences (see overlapping absorption peak at 344nm).

A similar argument can be made for SO2 although it was only implemented in the most recent publications (Suleiman et al., 2019 (Proposed to be implemented); Herrmanns et al., 2022; Sihler et al., 2012; Seo et al., 2019). Please explain why you have not chosed to include it and how strong you estimate for spectra affected by SO2 (e.g. strong pollution emitter or volcanoes) as well as how substantial this impact is on the global data-set.

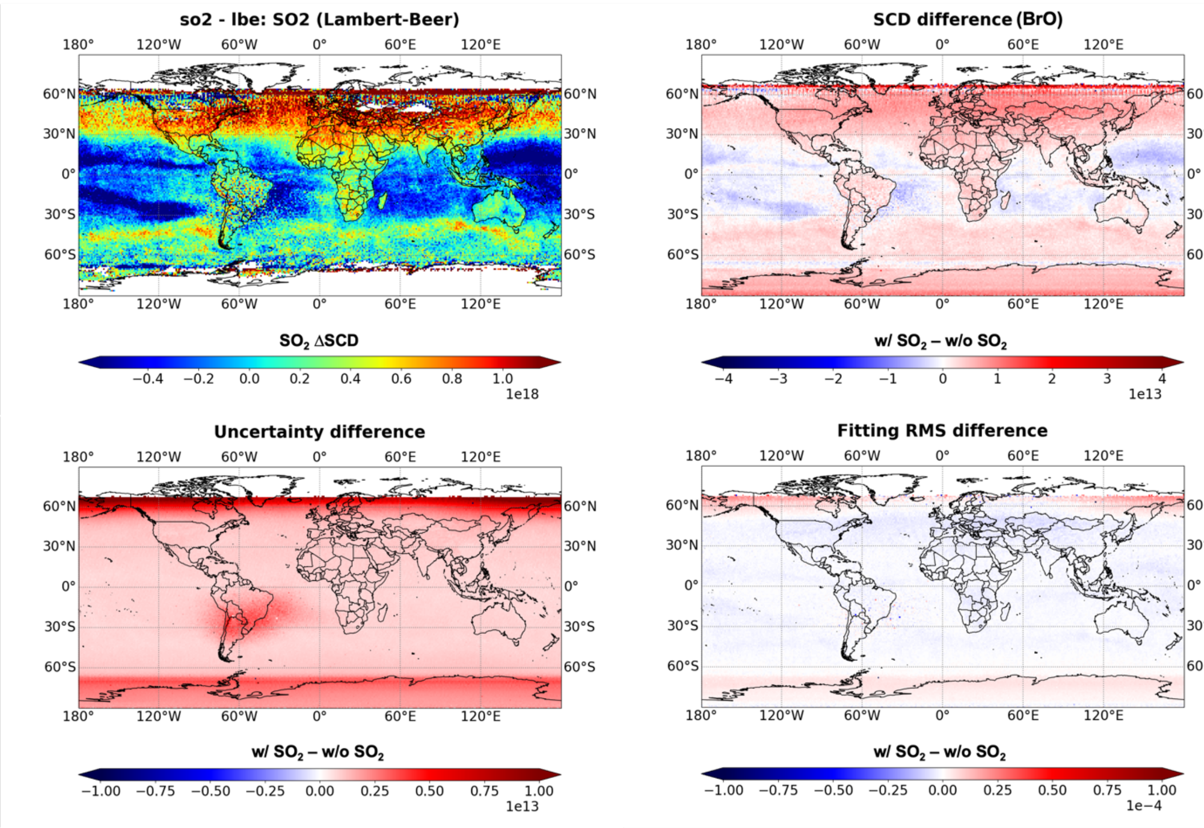
→ We have excluded OClO and SO_2 from the spectral fitting for the following same reasons: (a) the spatial distribution of the ΔSCDs of the species from the fitting didn't look reasonable (or physical), and (b) the inclusion of the species led to increases in fitting RMS and uncertainty in polar regions, where the BrO chemistry is of particular significance.

The figure just below shows the monthly averages of (a) OClO ΔSCDs, (b) changes in BrO SCDs, (c) changes in fitting uncertainties, and (d) changes in fitting RMS values when we included OClO for December 2018. For the ΔSCD panel, only pixels with cloud fractions < 0.3 were used. Apparently, the spatial distribution of OClO ΔSCDs doesn't reflect well the physical distribution of OClO in the atmosphere. This finding aligns with Pukite et al. (2021), who found that the inclusion of BrO led to significant biases in OClO retrievals from TROPOMI. Pukite et al. (2021) chose to exclude BrO from the fitting and, as an alternative, employ BrO columns determined from an independent fitting for a correction. However, this type of correction was not necessary for the OMPS-NM BrO retrieval because the difference in BrO ΔSCD between with and without OClO was very small, as shown in the upper right panel. Still, since the inclusion of OClO increased the fitting uncertainty and RMS values over polar regions (the lower panels), we simply excluded OClO from the fitting.



The same type of figure for SO_2 is presented below (again for December 2018). The spatial distribution of SO_2 ΔSCDs doesn’t reflect well the physical distribution of SO_2 in the atmosphere. Notably, strong negative values are found over the ocean. It’s worth mentioning that the fitting window we employ (331.5–358 nm) doesn’t cover the strongest SO_2 absorption features. The inclusion of SO_2 in the fitting even leads to non-negligible changes in BrO ΔSCDs as well as increases in fitting uncertainties all across the globe (the upper right and lower left panels). Therefore, we decided to exclude SO_2 from the fitting.

However, it is a valid point that excluding SO_2 might lead to biases in BrO retrievals over volcanoes. Although we haven’t added these OClO and SO_2 figures to the manuscript, we have added a brief description to Sect. 2.2.1 (Sect. 2.2 in the revised manuscript).



Page 8, Table 1: I suggest to highlight the trace gas absorption spectra in the list. For example by horizontal lines. Also add "the parameter are listed in their order of appearance in eq. 2".

→ We have added two horizontal lines and also added the phrase suggested by the reviewer.

Page 9, Figure 2: It would be beneficial to also include the residual spectrum in this plot as it gives information on potential residual structures originating from absorbers which are not accounted for.

→ We apologize for the confusion. The residual structures are already presented, as the blue curves in Fig. 2, referred to as "measured optical depths" in the caption, represent the sum of the modeled optical depths and the residuals.

We have added the following sentence to the caption to clarify it in the manuscript: "The measured optical depths are defined as the sum of modeled optical depths and residuals."

Page 13: In other studies (such as Seo et al. 2019) a uniform background of 3.5x1013 moleculesc cm-1 is used (based on Richter et al., 2002). Include how your background correction S_R typically is with respect to this value.

Richter et al., 2002: Richter, A., Wittrock, F., Ladstatter-Weissenmayer, A., and Burrows, J. P.: GOME measurements of stratospheric and tropospheric BrO, in: Remote Sensing of Trace Constituents in the Lower Stratosphere, Troposphere and the Earth's Surface: Global

Observations, Air Pollution and the Atmospheric Correction, edited by: Burrows, J. P. and Takeucki, N., Adv. Space Res., 11, 1667–1672, 2002.

→ The total VCD values from CAM-Chem for the reference sector are typically ~2.0–2.2×10^{13} molecules cm^{-2}, which is smaller than the value from Seo et al. (2019) and Richter et al. (2002) (3.5×10^{13} molecules cm^{-2}). However, there's a difference in the reference-sector latitudes between Seo et al. (2019) and this study. The CAM-Chem total columns vary spatially and temporally. For example, the VCD range within 30°S–30°N is 2.0–5.9×10^{13} molecules cm^{-2} for February, with the mean of 2.9×10^{13} molecules cm^{-2}, which is closer to that from Seo et al. (2019).

We have added a brief description to Sect. 2.2.3 (Sect. 2.4 in the revised manuscript).

Page 13 line 17-31: It would improve the readability to include the names "S_R" and "S_B", when talking about these quantities in the text.

→ We have added S_R and S_B in the text.

Page 37 line 20: Please remove "including volcanic plumes". Volcanic application is not mentioned at all in the result section and as the major volcanic constituent "SO2" is not accounted for in the spectral fitting, this statement is questionable.

→ This is a valid point. We have removed that phrase.

Page 38, line 13-18, concerning the "modeled stratospheric BrO DeltaSCD": I assume the "modeled Delta SCDs" in line 17 is the same as the "modeled stratospheric BrO Delta SCD". Name both the same, and consider to mention this in the first sentence of its explanation (line 13).

→ The reviewer's assumption is correct. We have changed "modeled ΔSCDs" in line 17 to "modeled stratospheric BrO ΔSCDs." Also, in Line 13, we have changed the wording "stratospheric BrO columns from CAM-Chem" to "stratospheric BrO ΔSCDs from CAM-Chem." Then, we have added "stratospheric" throughout the paragraph to prevent readers' confusion.

Page 38, Line 15-17: From your explanation, it looks like the "modeled stratospheric BrO Delta SCD", which is subtracted by is defined in a way that its mean is zero at 0-10°N and non-zero elsewhere. Thus the "Delta SCD bias" will then be the complete retrieved SCD subtracted by zero at the equator and non-zero

→ The reviewer's comment is accurate, although it appears that a part of it might be missing.

Page 38-39 and figure A1, regarding the correlation with O3:

1. How have you combined the different O3 SCDs to one O3 SCD? Did you follow the formula proposed by Pukite and Wagner (2016) eq. 16? If so, please add a reference.

→ In this appendix, we didn't combine the two O_3 SCDs. That's because we didn't use the SCD values directly when calculating the correlation coefficients (R). As stated in the manuscript, the R values presented in this appendix are between the Jacobians derived within the retrieval algorithm, not between the SCDs.

However, we did combine the two O_3 SCDs for the calculation of the O_3 optical depth in Fig. 2. For that calculation, we used the formula proposed by Pukite et al. (2010) rather than Pukite and Wagner (2016). During the revision, we added the 2010 reference to the part where we describe Fig. 2 instead. (This reference was already cited several times elsewhere in the manuscript because we use the Taylor-series parameters for the retrieval.)

2. I do not see the benefit of looking at two O3 absorptions at 243 and 273K, as there is only O3 absorption. If you do not gain any benefit from using the two, then I suggest to skip this.

   → The reason why we don't combine the two O_3 temperatures here is that, during the retrieval process, the BrO cross section technically interferes with each of the two O_3 cross sections individually rather than interfering with a single combined O_3 absorption spectrum. Figure A1 demonstrates that there are fitting windows where we can avoid interference with one of the O_3 temperatures but not with the other. Our objective was to find a fitting window where we could avoid strong interference from both O_3 temperatures.

3. Should you chose to keep the distinction between 243 and 273K, how did you avoid a cross correlation between the two spectra (which are very similar)? Have you orthogonalized the O3 absorption spectrum at 273K w.r.t the one at 243K? Please state this in the text.

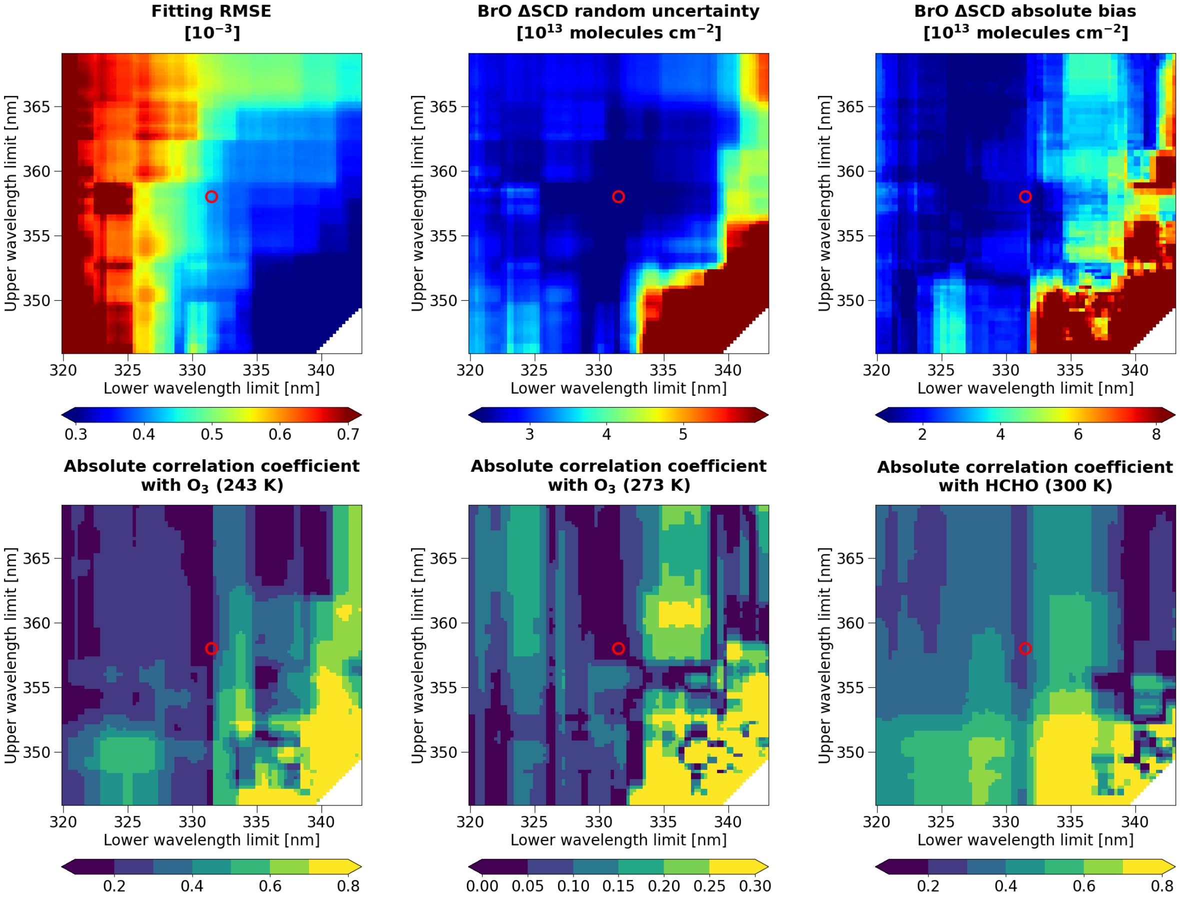
   → We haven't orthogonalized the O_3 cross section. An additional step is not required because what we aim to assess here is the correlation between the Jacobians of BrO and the others, which are all equally treated in the retrieval algorithm (or program) regardless of the species. The algorithm provides a covariance matrix as one of the outputs, and we simply chose the proper elements in this matrix to assess the correlation between the BrO Jacobian and another. We have added a brief description to this appendix.

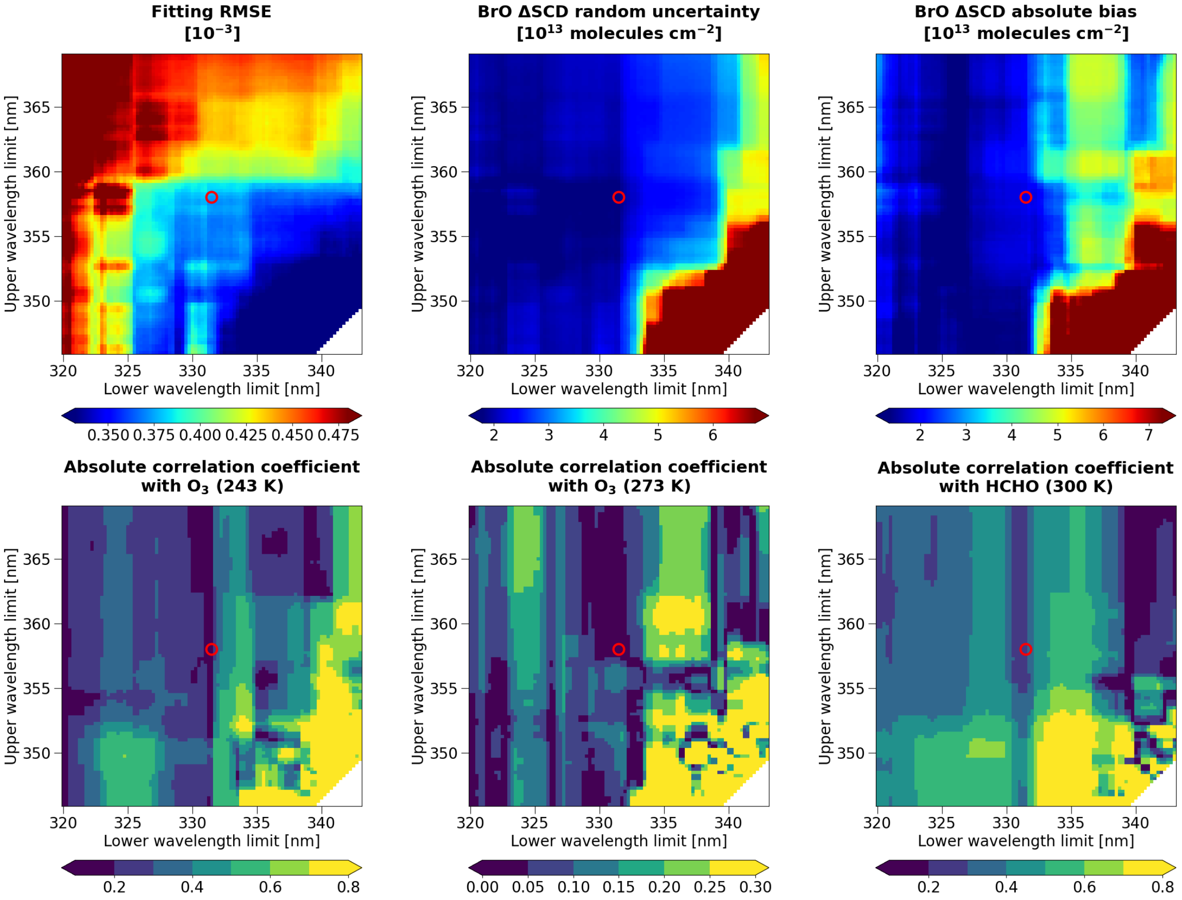
Page 39 line 8: The choice of a percentile seems suboptimal to me and introduced strong data-selection biases and I would urge to change this. For instance, the pixel at high VZA will have a higher SCD compared to the nadir looking pixel and they will be selected thus more frequently. Additionally, tropospheric enhancements will be more dominant. If you want to select for high latitude and high SZA, then why not use latitude and SZA as a selection criterium?

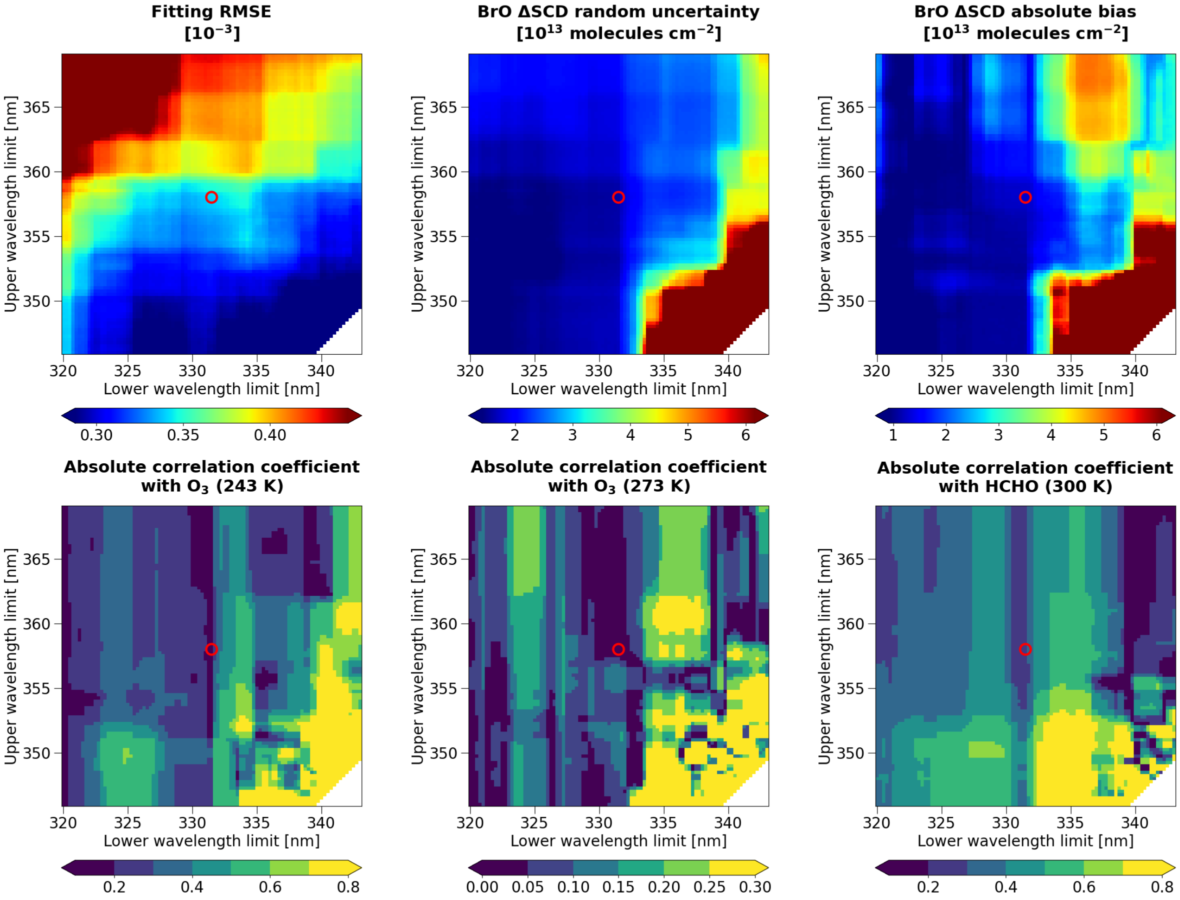
Additionally, the paper is about a global product of BrO. Please justify why you have not also looked if the fit is also performing well at other regions (cf. Seo et al., 2019, who performed a retrieval interval mapping for several cases and also for an equatorial region).

→ We have made several changes in this analysis following the reviewer's comment. First, we have changed the selection criterion. Considering correlations between latitudes and SZAs, we used only latitudes. Second, we have included all latitude ranges. We have done the same analysis for (a) 90°S–60°S and 60°N–90°N combined (high latitudes), (b) 60°S–30°S and 30°N–60°N combined (middle latitudes), and (c) 30°S–30°N (low latitudes). In addition, to constrain the conditions for stratospheric bias analysis, we have excluded pixels with (a) cloud fractions > 0.2 and (b) snow or ice. Also, we have excluded pixels with SZAs > 80°.

The three figures just below show the results for high latitudes, middle latitudes, and low latitudes, respectively, in the order of presence. The results for the same month (April 2018) is presented as in the previous analysis.

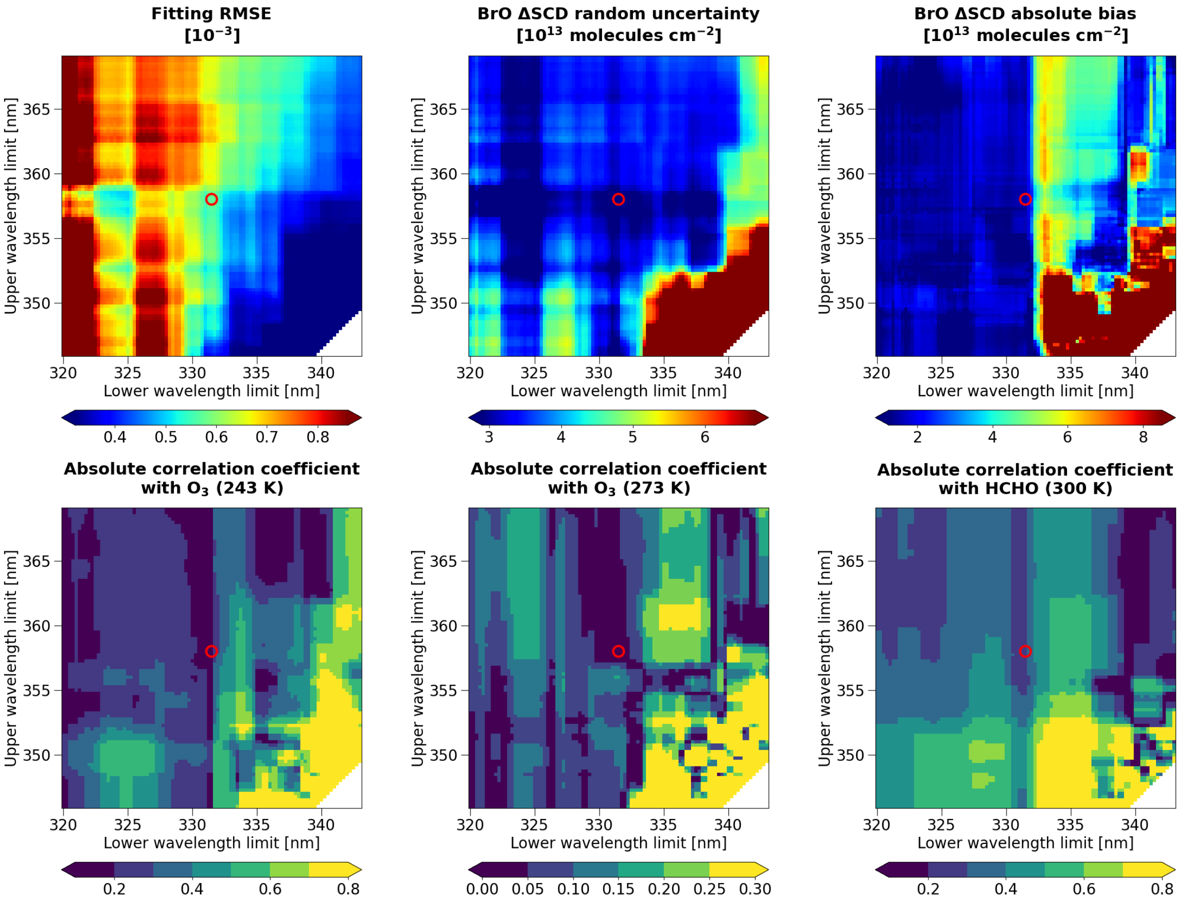



Fitting RMSE
[10^{-3}]
BrO ΔSCD random uncertainty
[10^{13} molecules cm^{-2}]
BrO ΔSCD absolute bias
[10^{13} molecules cm^{-2}]
Upper wavelength limit [nm]
365
360
355
350
320
325
330
335
340
Lower wavelength limit [nm]
0.350 0.375 0.400 0.425 0.450 0.475
2 3 4 5 6
2 3 4 5 6 7
Absolute correlation coefficient
with O_3 (243 K)
Absolute correlation coefficient
with O_3 (273 K)
Absolute correlation coefficient
with HCHO (300 K)
0.2 0.4 0.6 0.8
0.00 0.05 0.10 0.15 0.20 0.25 0.30
0.2 0.4 0.6 0.8



Since the fitting window originally selected shows an excellent performance here, we didn't change the window. Furthermore, the example below for January 2018 for high latitudes demonstrates that biases could steeply increase if we choose a longer wavelength for the lower limit of the fitting window.

We have updated Appendix A based on these new results.



There is one paper about long-term BrO time-series:

Bougoudis, I., Blechschmidt, A.-M., Richter, A., Seo, S., Burrows, J. P., Theys, N., and Rinke, A.: Long-term time series of Arctic tropospheric BrO derived from UV–VIS satellite remote sensing and its relation to first-year sea ice, Atmos. Chem. Phys., 20, 11869–11892, https://doi.org/10.5194/acp-20-11869-2020, 2020.

I suggest to include this paper in your introduction. You can frame this as an advantage of the OMPS data-set who in difference to Bougoudis et al., 2020 does not require a complicated inter-calibration of the time-series of the different sensors.

→ The reference has been added following the reviewer's comment.

Technical comments:

Page 2, Line 30: As the authors are very thorough in giving a complete list of relevant references in the introduction, I would here also strive for completeness and include the other two studies of BrO from GOME-2: Hörmann et al., 2013 and Sihler et al., 2012 (already included in the references)

→ The references have been added.

Page 3, Line 14: also here I would complete the list of references who used an area as an estimate for the stratospheric correction and include Hörmann et al., 2013 to the list of Wagner et al., 2001; Hörmann et al., 2016).

→ The reference has been added.

Page 4 line 22: I believe you mean “local solar time”?

→ It has been revised.

Page 13 line 17: The comm in “(i.e., […])” is not needed.

→ The comma has been removed.

Page 13 line 20: add “separately” after “across-track position”

→ It has been added.

Page 38 footnote: somewhere there is a bracket missing or one too many.

→ The bracket has been removed.