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.

RC1

Review of "Global retrieval of stratospheric and tropospheric BrO columns from OMPS-NM onboard the Suomi-NPP satellite"

In this manuscript, the authors describe a new BrO data product using OMPS-NM measurements. The product includes stratospheric and tropospheric columns, uses a complex stratosphere – troposphere separation algorithm, applies pixel specific airmass factors and provides detailed uncertainty estimates. The topic of the manuscript fits well into the AMT scope, the product described is of interest to the atmospheric chemistry community and the paper is well written and includes detailed descriptions of the algorithms used. I therefore recommend it for publication in AMT.

I have however several questions, comments and suggestions to the manuscript and the algorithm, which the authors should address before the manuscript is accepted:

1) Reference sector correction

What the authors call reference sector correction in fact includes two corrections: a) the addition of the modelled BrO offset necessary when using a radiance background spectrum and b) a latitudinal correction of the slant columns based on the model. The latter correction is critical as the way I understand it, it forces the baseline of the measurements on the model values. Therefore, I think the stratospheric BrO product is lo a large extent just reproducing the model values. This is in contrast to the statements in the manuscript claiming that both stratospheric and tropospheric column are retrieved from the measurements.

Please a) discuss this point and b) include an example of the correction for one full orbit, for example one of those shown in Fig. 3.

→ As the reviewer described, the reference sector correction includes two parts. We refer to the latter part as bias correction, in which the bias correction terms (S_B) are defined as the differences between two third-degree polynomials fitted to (a) the modeled total SCDs and (b) the background-corrected SCDs ($\Delta S + S_R$) in the along-track dimension for each cross-track position. Since the third-degree polynomials are smooth enough, this approach is capable of correcting smoothly varying biases in the SCD retrievals in the along-track direction without introducing detailed spatial structures from the model into the retrievals. Therefore, this process is rather a correction of offsets than just a reproduction of the model values.

We have added the figure below to the revised manuscript to include an example of the correction for o7594, the orbit shown in Fig. 3a from the initially submitted manuscript (Fig. 4a in the revised manuscript). Panel (e) displays all four quantities associated with the reference sector correction (ΔS, S_R, S_B, and S_{total}) for the 15th cross-track position (0-based), with S_B values plotted in green. The along-track variability in the retrieved ΔSCDs (ΔS, blue) is well preserved in the total SCDs (S_{total}, red). This example demonstrates the capability of the reference sector correction.

<u>A description of the new Fig. 3 has been added to the final part of Sect. 2.2.3 (which became Sect. 2.4 in the revised manuscript).</u>

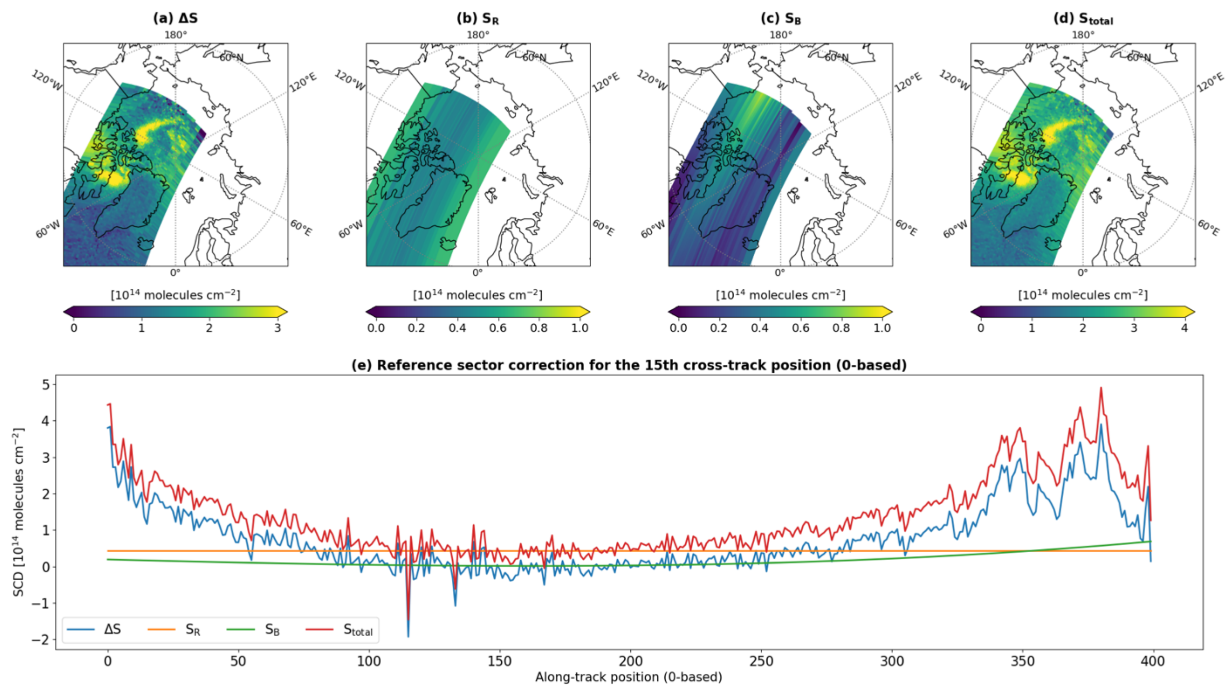


Figure 3. Description of the reference sector correction. Intermediate quantities are presented for o7594 from 15 April 2013. Panels (a–d) show the ΔSCD (ΔS), background SCD (S_R), bias correction term (S_B), and total SCD (S_{total}), respectively. Panel (e) depicts the along-track variabilities of the four quantities for the 15th cross-track position (0-based).

2) CAM-chem climatology

The algorithm described heavily relies on the CAM-chem climatology for the stratospheric columns (see above), the separation between stratospheric and tropospheric signals and for the airmass factors. However, a) it is not clear how well the climatology represents the real atmospheric BrO field and b) tropospheric BrO enhancements are very dynamic events, and their magnitude and location cannot be reflected by a static monthly climatology. This has important implications for the airmass factors which probably are often not correct as the monthly mean profiles are neither a good representation of BrO events, nor of background conditions. The algorithm foresees the use of “flattened” profiles in case the measurements with low BrO columns, but the opposite case (high BrO in a region where the climatology does not expect a BrO event) is not treated separately.

Please a) discuss the impacts of using a climatology as input for the airmass factors and b) include a figure comparing the modelled climatological tropospheric columns in comparison to the measurements, for example for the orbits shown in Fig. 3.

→ Using climatology for AMF calculations can result in both random and systematic uncertainties in the retrievals. The contribution to the random uncertainties has already been taken into account in the AMF uncertainty estimates; it is represented by the standard deviation of AMFs for each profile cluster, as described in Sect. 2.3.2 (Sect. 2.6.2 in the revised manuscript).

Regarding systematic uncertainties, there are two main quantities in the retrieval algorithm that can be impacted by using climatology: (a) the initial estimates of tropospheric VCDs and (b) the tropospheric AMFs. The initial tropospheric VCD estimates are supposed to represent the background conditions, and a flattening technique is proposed in this study to achieve it. Still, if there are biases in the flattened profiles (columns), they are propagated to the initial estimates and, ultimately, to the final tropospheric retrievals. This impact is universal. On the other hand, the impacts on the AMF calculations appear differently for non-hotspots and hotspots. The flattened profiles are employed to calculate tropospheric AMFs for non-hotspots, while climatology is applied for hotspots. This approach assumes that climatology can represent profile shapes for tropospheric enhancement events. Systematic errors can occur when this assumption fails.

We have added discussions about the systematic uncertainties due to climatology to the final part of Sect. 3.

To address the reviewer's bullet point (b), we have added Appendix B to the revised manuscript and included the figure below in it. (Appendices B and C in the initially submitted paper have become C and D in the revised manuscript.) This figure compares the modeled and measured tropospheric columns for the orbits shown in Fig. 3 (Fig. 4 in the revised manuscript). Without flattening, the modeled tropospheric columns ($V_{\mathrm{trop}}^{\mathrm{CTM}}$) show homogeneously elevated values, while the final retrievals (V_{trop}) capture dynamic tropospheric enhancement events individually.

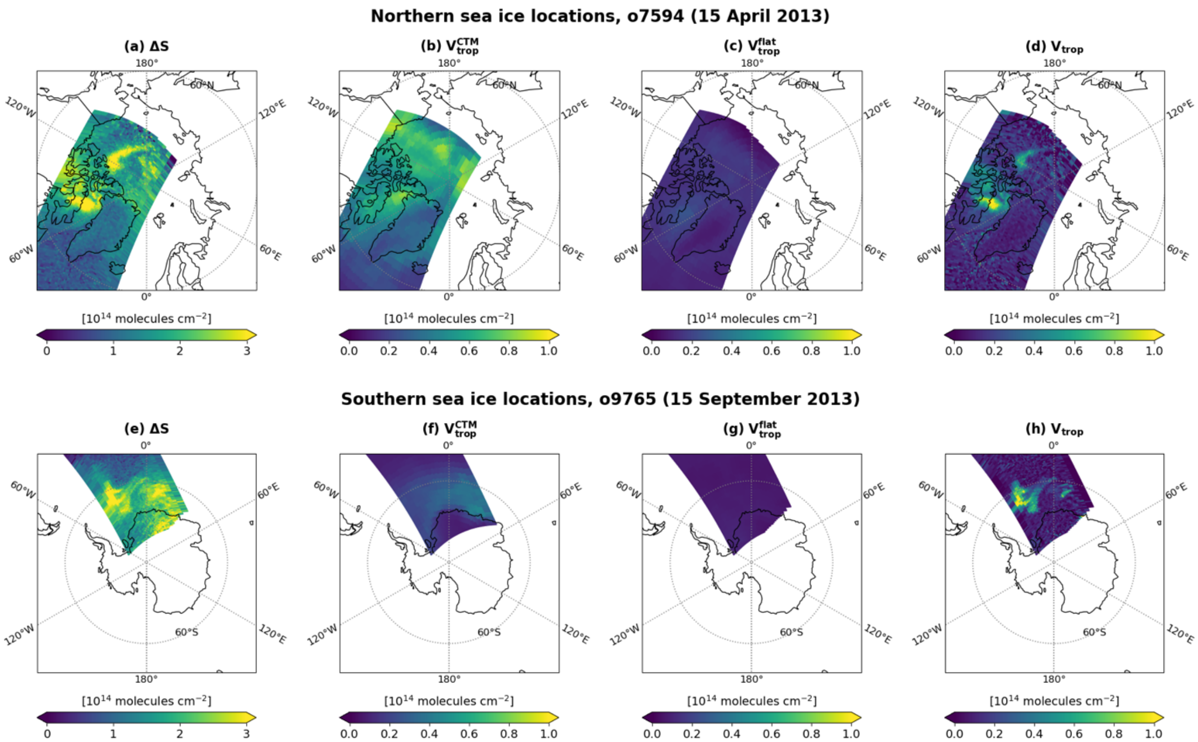


Figure B3. Comparisons between modeled and retrieved tropospheric BrO columns. Two orbits are selected to represent sea ice locations in the Northern Hemisphere (o7594, 15 April 2013) and Southern Hemisphere (o9765, 15 September 2013). Panels (a) and (e) show the retrieved ΔSCDs (ΔS). Panels (b) and (f) present the modeled tropospheric columns before flattening (V_{trop}^{CTM}), while (c) and (g) show the flattened tropospheric columns (V_{trop}^{flat}). The retrieved tropospheric columns (V_{trop}), i.e., the results of the stratosphere-troposphere separation (STS), are displayed in panels (d) and (h).

3) Polar vortex

The authors discuss the well known problem of using O3 columns as proxy for the stratospheric BrO columns and state, that their method "still preserves the overall spatial pattern of the stratospheric field". I have not understood why that should be the case. If we have ozone depletion (and possibly stratospheric BrO enhancement) within the vortex, the relationship between O3 and BrO will be different for vortex and non-vortex air masses, leading to large scatter in the O3 – BrO plot. If all the in vortex values are removed and filled with surrounding (out of vortex) values, the stratospheric BrO column will not be correct.

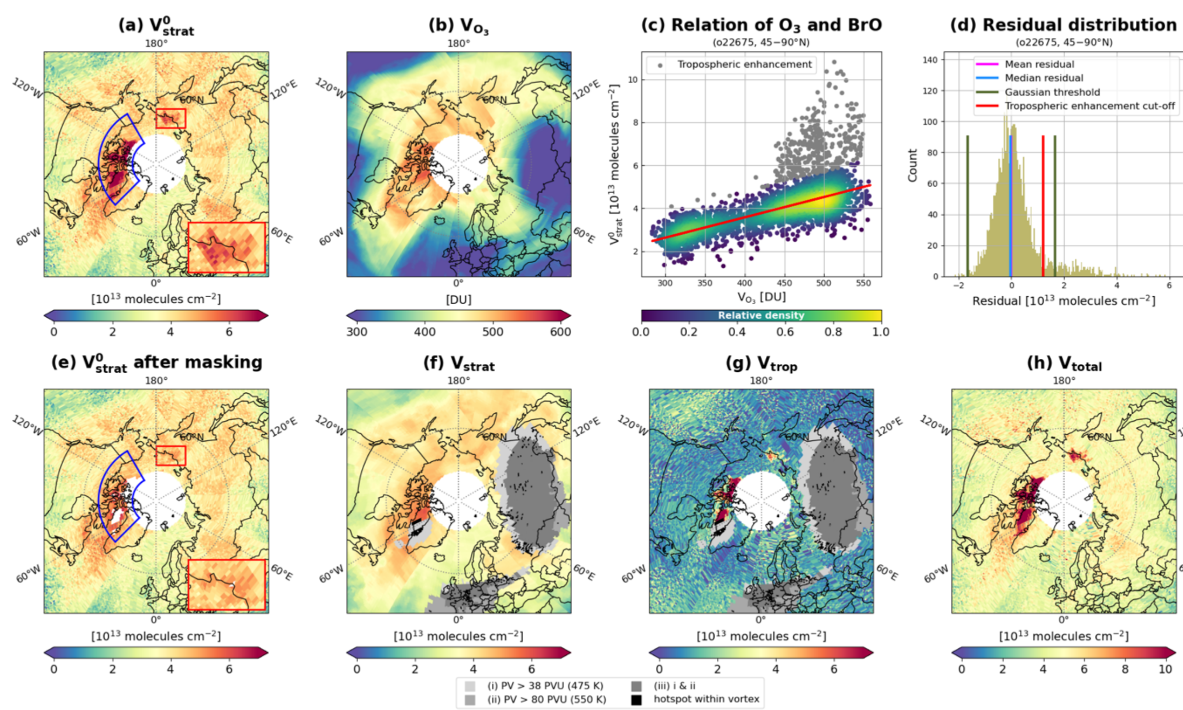
Please explain why your method is less affected by this problem than that of previous studies.

→ It is true that the stratospheric BrO columns cannot be correct in the polar vortex if all the in-vortex values are removed. However, the removed portion within the vortex is not necessarily 100% because only hotspots detected by the STS scheme are removed within the vortex.

Previous studies directly estimated the stratospheric BrO columns using O_3 columns, whereas we use O_3 columns only for detecting and reconstructing hotspots. That's why we mentioned that our method preserves the overall spatial pattern of the stratospheric field.

However, the reviewer's comment is correct. Hotspots detected within the vortex have more possibilities to be stratospheric than those detected outside of the vortex. To address this issue, we have added a new quality flag for STS, which is a three-digit binary variable. 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). For our analyses in this manuscript, we didn't use this quality flag to filter out data points to give an idea of the general retrieval performance.



4) Uncertainties

Although a detailed discussion of uncertainties is given, I'm somewhat confused by what to expect from the data product. Are the uncertainties given for individual pixels? Has each pixel in the product an uncertainty value?

→ We apologize for the confusion. Yes, the uncertainties are given for individual pixels, and each pixel in the product has an uncertainty value. To clarify it, we have added a sentence to the beginning part of Sect. 2.3 (Sect. 2.6 in the revised manuscript).

Will uncertainties be smaller in monthly averages? If so, why are the DSCD uncertainties shown in Figs. 12 and 13 for monthly averages comparable to the median uncertainty quoted for an individual pixel?

→ It is correct that the uncertainty in a monthly average value is smaller than that for an individual pixel. However, what we presented in Figs. 12 and 13 (Figs. 13 and 14 in the revised manuscript) are "averages of individual pixel uncertainties," not "uncertainties of averages." That's why the presented values are comparable to the median of individual uncertainties. We chose to present these average uncertainties because the purpose was to investigate if individual uncertainties are low enough to ensure that individually detected ΔSCDs are above the noise level.

To prevent readers' confusion, we have added a sentence to the caption that these are "averages of individual pixel uncertainties."

How is the uncertainty of having a high surface BrO event in the data at a location where the CAM-chem climatology has background conditions taken into account?

→ This situation can occur; however, we consider these systematic errors in retrievals. Since we estimate only random uncertainties in this study, these mismatches are not taken into account.

This comment aligns with the reviewer's comment 2 (CAM-chem climatology). As described in our response to that comment, we have added discussions about the systematic uncertainties due to climatology to the final part of Sect. 3.

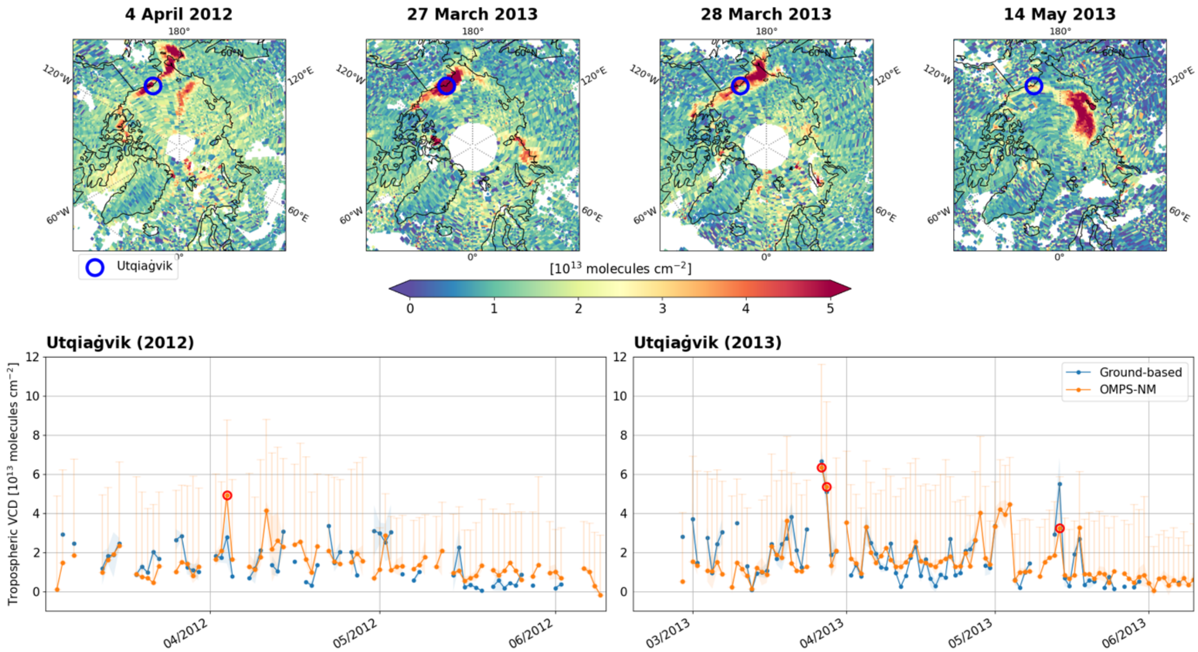
Can the product be used on a daily basis (Figs. 3 and 4 suggest that this is the case) or should it better be used on a monthly basis?

→ Daily retrievals are capable of capturing tropospheric enhancements, as presented in the figures. However, as shown in Fig. 8 (Fig. 9 in the revised manuscript), daily retrievals have larger errors than monthly averages. The decision to utilize either daily or monthly data hinges on the specific analyses or requirements of the users. The random uncertainty estimates provided for individual pixels can assist the decision.

Please add error bars to the satellite data in Fig. 7 and a paragraph on data usage.

→ Error bars have been added to the figure, as shown below. However, the lower error bars, which are supposed to have the same lengths as the upper ones, are not plotted for display purposes. This description of the lower error bars has been added to the caption.

Also, we have added a paragraph on data usage to Sect. 5 (Discussion and conclusions).



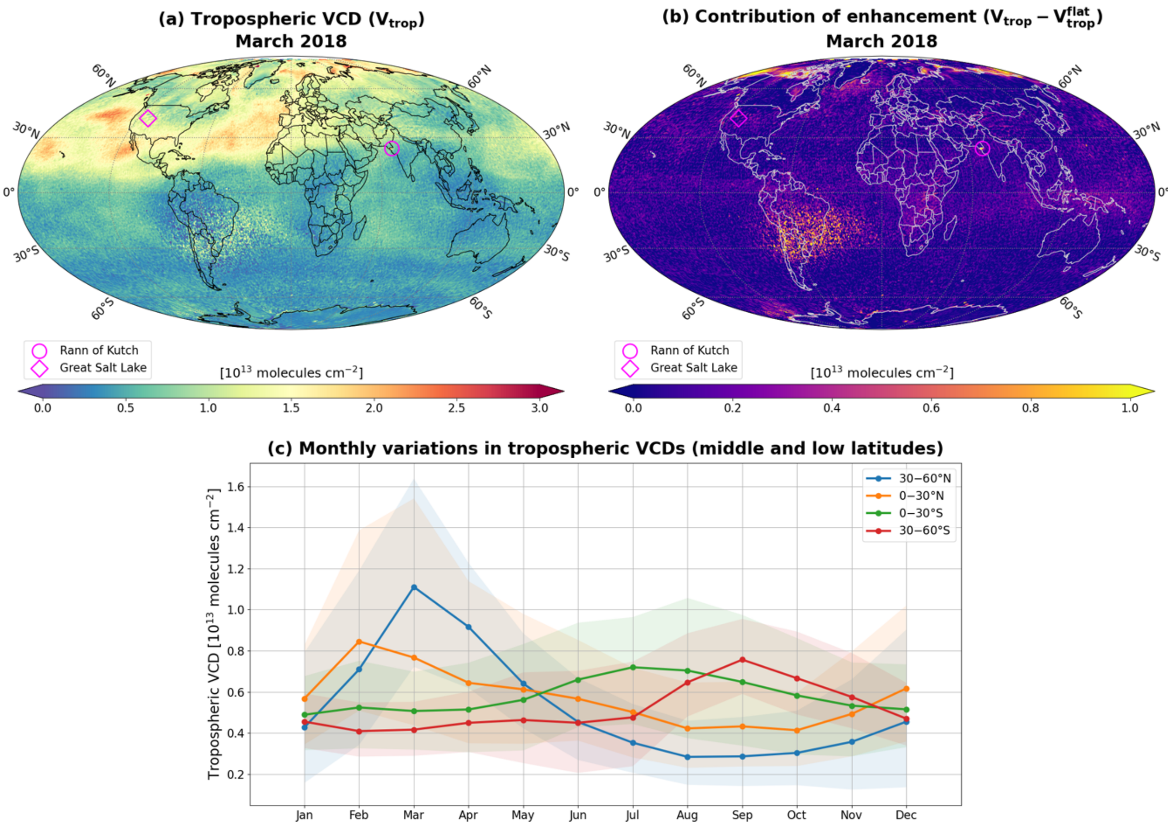
5) High values over the ocean

In Fig. 11, large BrO columns are shown over much of the NH oceans, and as far as I can see, the largest BrO columns in that month are not found in the Arctic but somewhere over the Pacific. Do you think this is realistic? How do these findings compare to other satellite products and independent measurements? Are these high columns already visible in the slant columns or are the introduced by the CAMS-chem based airmass factors?

→ Relatively high values in the final retrievals (V_{trop}) can be either from the flattened columns ($V_{\mathrm{trop}}^{\mathrm{flat}}$), predominantly contributed by the free troposphere, or the detected tropospheric enhancements. Separating those two impacts can be achieved by calculating the difference between V_{trop} and $V_{\mathrm{trop}}^{\mathrm{flat}}$, i.e., the contribution of enhancement $V_{\mathrm{trop}}^{\mathrm{enh}} = V_{\mathrm{trop}} - V_{\mathrm{trop}}^{\mathrm{flat}}$. If the elevated values in the V_{trop} field are due to tropospheric enhancements, those pixels should appear in the $V_{\mathrm{trop}}^{\mathrm{enh}}$ field.

The spatial distribution of $V_{\mathrm{trop}}^{\mathrm{enh}}$ for March 2018 has been added to Fig. 11 (Fig. 12 in the revised manuscript), as presented below. The new panel (b) shows the $V_{\mathrm{trop}}^{\mathrm{enh}}$ values, demonstrating where the enhancements were detected from the OMPS-NM instrument.

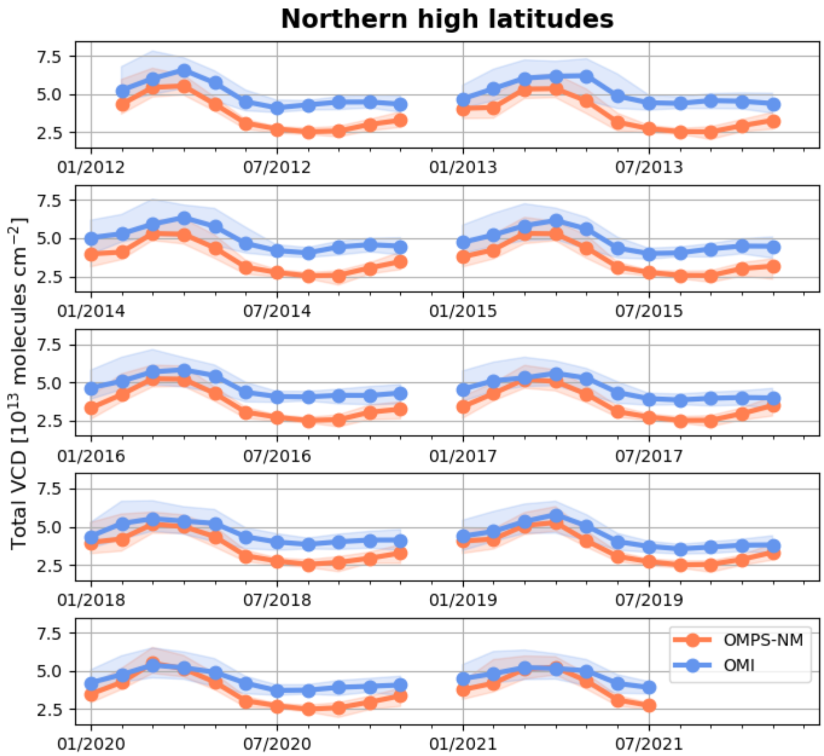
Regarding the comparison with other data, unfortunately, we were not able to find independent measurements that can be used to validate the tropospheric columns over the Pacific.

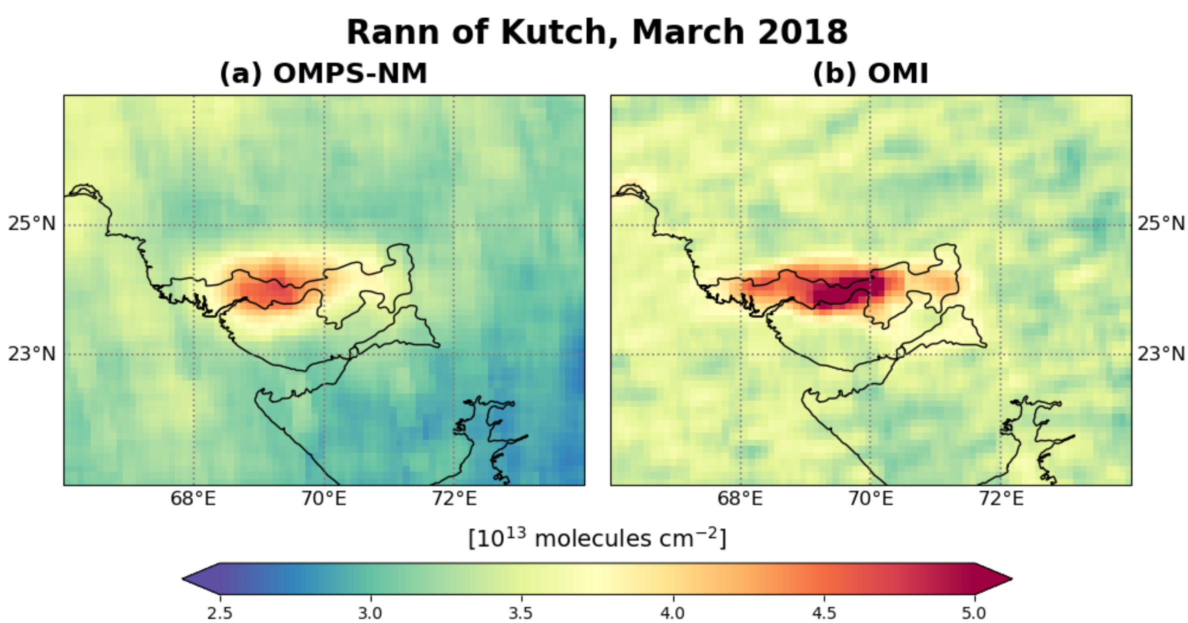


6) Comparison to other satellite data

Why is there no comparison to other satellite data? The data set is advertised as extension of the OMI afternoon BrO time series, and I think it would be good to include some kind of comparison between BrO observations from the two platforms, even if it is just a visual side-by-side comparison of a monthly average.

→ This is a valid point. 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 the OMPS-NM BrO retrievals are capable of extending the afternoon BrO time series. The 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.



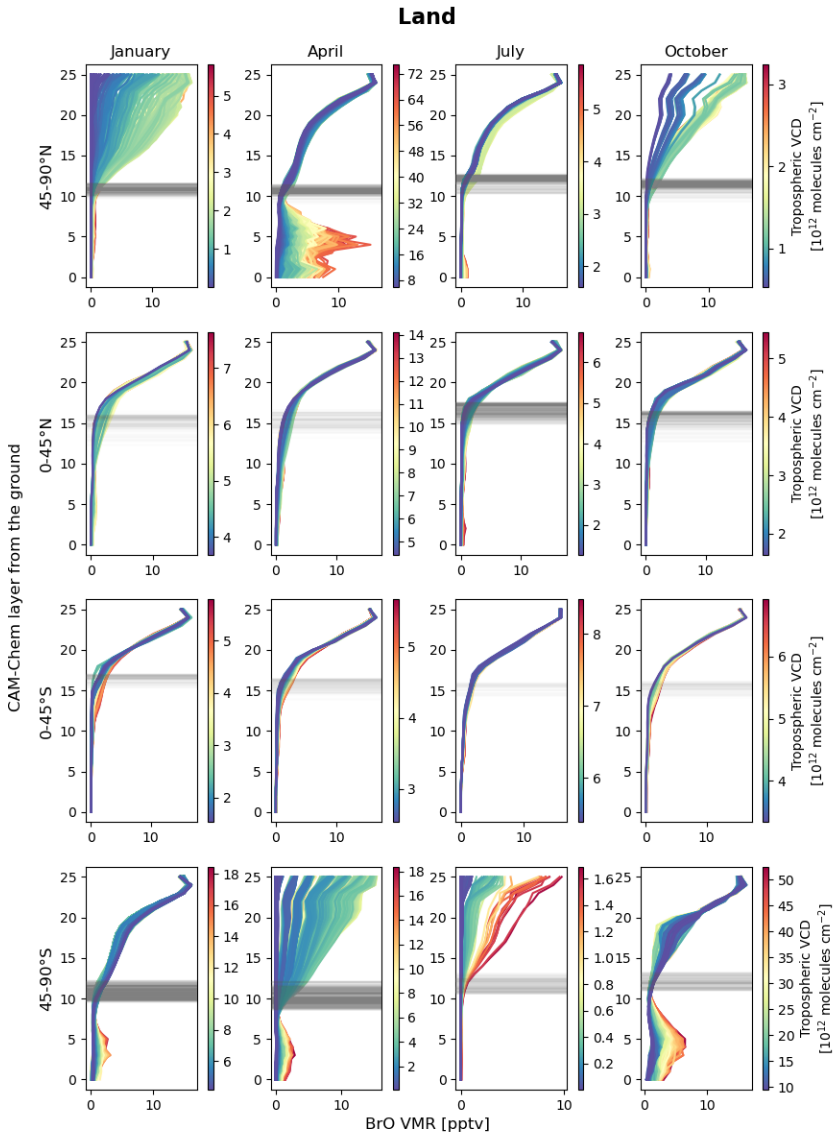


7) Profile flattening

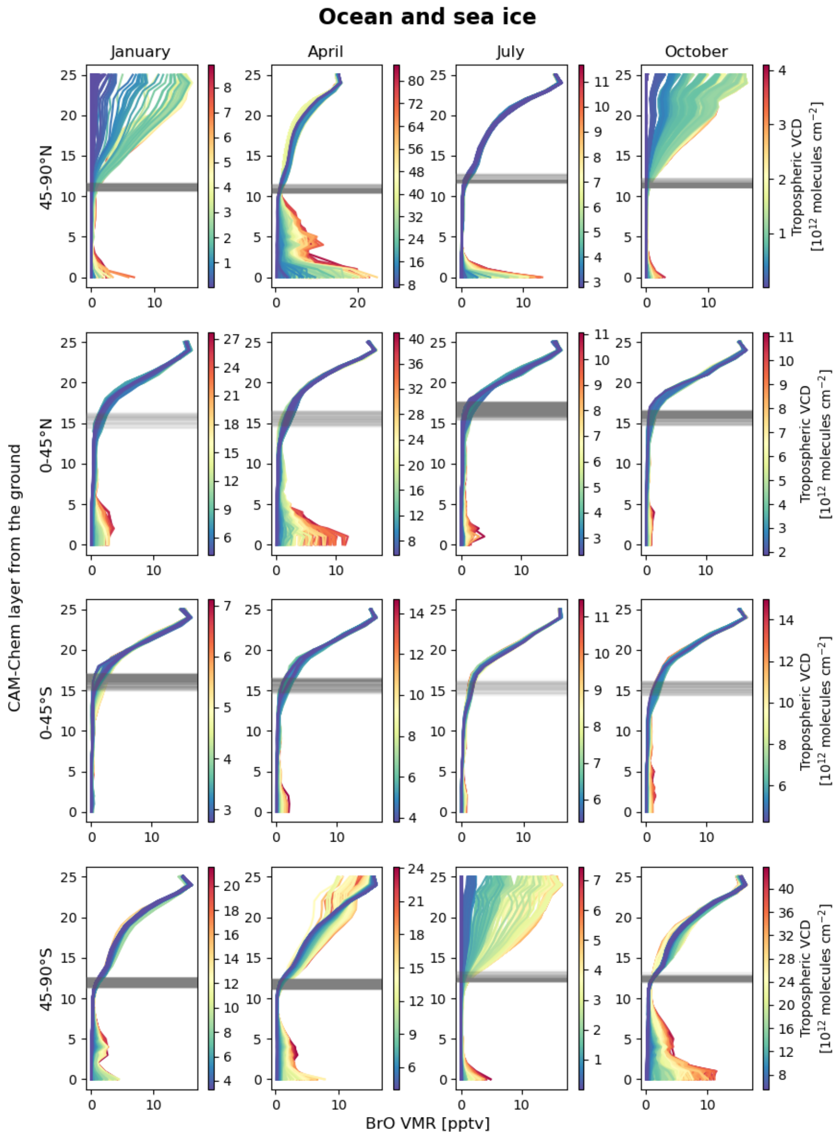
This procedure seems arbitrary to me. I do not see why such a profile should give reasonable tropospheric BrO columns or realistic airmass factors. Please justify your approach.

→ 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.



Land
January
April
July
October
45-90°N
0-45°N
0-45°S
45-90°S
CAM-Chem layer from the ground
BrO VMR [pptv]
Tropospheric VCD
[10^{12} molecules cm^{-2}]



8) Coastal artefacts

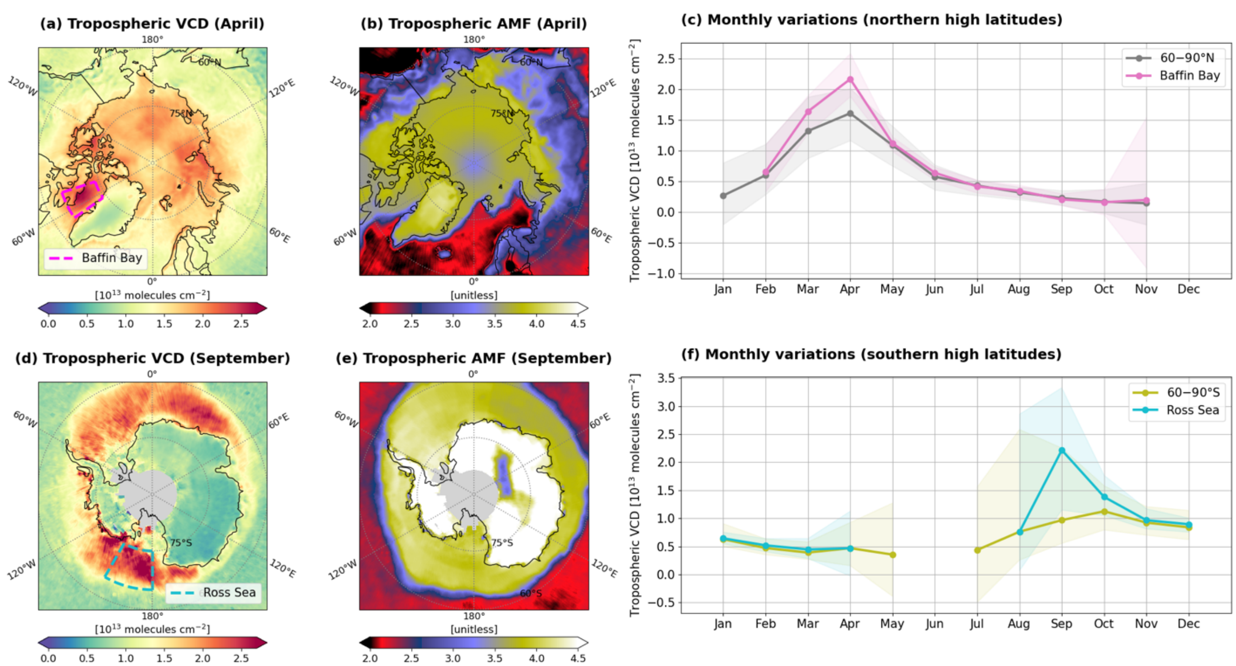
In Fig. 10, there are many localised spots of suspiciously high tropospheric BrO along the Antarctic coast but also at the sea ice edge. As this is a longterm average, it is clear that the high values are from too small airmass factors, and this is confirmed by figure (e). In my opinion, this should not be part of the product as it clearly is an artefact. I expect similar artefacts in the daily images also in the NH, for example close to Spitsbergen. My suggestion is to at least include a flag for those retrievals having very small airmass factors, and to increase the uncertainty estimates for such pixels.

→ Thank you for your comment. While addressing this issue, we were able to find the root cause and fix the problem. The cause was the improper treatment of 'ice shelves' around Antarctica in the retrieval algorithm. Specifically, the land mask data employed in this study didn't treat the ice shelves as land. Since they were not sea ice either, our retrieval algorithm treated them as if they were water bodies. In reality, however, the surface of the ice shelves can be very bright, primarily associated with snow. Therefore, applying water-body surface albedo leads to significant underestimations of tropospheric AMFs.

During the revision, we found out that the ice shelves are treated as land in the NSIDC sea-ice data, and MODIS BRDF values are also given there. Therefore, we applied NSIDC's definition of land when estimating surface reflectivity for latitudes < –39°, which led to the use of MODIS BRDF values for the ice shelves. As a result, the localized spots of suspiciously high tropospheric BrO disappeared. The updated figure is presented below.

We haven't found such an issue for the Northern Hemisphere, but just in case, we changed the algorithm configuration to use MODIS BRDF if the IMS data reports the presence of snow over water pixels.

Descriptions have been added briefly to Sect. 2.2.2 (Sect. 2.3 in the revised manuscript).



9) Large AMF above ocean

I'm surprised by the large values of the airmass factor over much of the oceans. Values above two indicate the presence of a significant fraction of the BrO in the free troposphere. Do you think this is realistic?

→ Unfortunately, we were not able to find independent measurement data sets that could be used for verifying BrO columns or profiles in the free troposphere.

This comment aligns with the reviewer's comment 5 (High values over the ocean). As described in our response to that comment, we have revised Fig. 11 (Fig. 12 in the revised manuscript) to address this issue.

10) Data availability

I understand that the product is not yet released, but to my knowledge, the data has to be available in some form for the manuscript to be published in AMT. Maybe just add that it is available on request?

→ The product has been accepted for online data distribution through GES DISC (NASA). We have recently received a DOI (10.5067/PSPSYHVDNSJE), but uploading the files has yet to be done. After the product is uploaded to the GES DISC server, it will be publicly available.

We will revise the 'Data availability' paragraph in accordance with the status of the data upload. If the uploading process is not finished by the final stage of the entire revision process, we will add that the product is available upon request before the official release.