Response to the Reviewer #1

We thank the Reviewer for the constructive review and address the comments below.

The authors present a new algorithm to retrieve OClO from measurements of the TROPOMI satellite instrument. Overall, the paper presents valuable information and recommended DOAS settings for OClO retrievals from space that deserves to be archived. I believe the paper should be published in AMT. The authors present convincing comparison between TROPOMI and ground-based OClO SCDs, which clearly demonstrate the value of the satellite dataset. The authors present a number of tests and new settings. Some of these are new and make a lot of sense. However, the impression is also that many complication is introduced that is not really necessary. What is missing is a summary of the retrieval settings and choices that has the largest impact on the accuracy. The English is not very good and could be improved.

We completely agree with the reviewer that some of the settings do not give large improvement as already shown in the sensitivity studies (Appendix B) and thus are optional. We will provide the requested summary in the main text of the manuscript as a separate subsection (Sect. 2.2.4):

"Sect. 2.2.4.Sensitivity to retrieval settings

To motivate the retrieval setup as introduced in Sect. 2.1, we investigated the effect of different retrieval settings on the retrieved OClO SCDs by applying modifications with respect to the standard fit scenario described in Sect. 2, Table 1. In Table 2 the specific settings for the sensitivity studies (second column) and corresponding main results (remaining columns on the right) are provided. We refer for a more detailed description of the obtained results to Appendix B. Table 2 summarizes the minimum and maximum offsets from zero (third column) for the same days introduced in Sect. 2.2.1 for which no OClO is expected (25 Aug 2018 for the NH and 25 Dec 2018 for the SH, 25 Nov 2018 (NH) and 25 Apr 2019 for SH) at SZAs of 90° and 85° for the modified settings (second column). The standard deviation of the binned mean (maximum from the investigated 4 days) is provided in the fourth column while the autocorrelation coefficient for 25 Nov 2018 (NH) at lags of 1 and 20 pixels across track, both with a lag of 0 along track, are listed in the last column. Larger absolute values than for the standard case are marked bold. Comparing these differences to the performance of the standard scenario, the case numbers of the settings (first column) are marked bold which are causing a worsening of the retrieval.

In particular, it is important to consider the OClO×λ term (compare to sensitivity case 2) and carefully select a wavelength for the calculation of the OClO SCD from the fitted OClO + OClO×λ terms (case 1) ensuring a minimization of the systematic error. The accuracy improvement here is larger than a slight increase in the random error. Also a special care should be taken when selecting the fit window (cases 3 and 4) where already small changes (case 3) can lead to a lower accuracy and increased background structures as clearly recognized by the autocorrelation analysis. For the retrieval of OClO it is important to consider the BrO absorption. Adding a BrO cross section to the retrieval as a free fit paramter however leads to large retrieval errors (case 9). Applying a BrO correction (Appendix A4) by subtracting the BrO signal retrieved in another fit window suitable for retrieval of BrO is important to account for the wavelength dependency of the BrO AMF (case 8). Interestingly, the exact BrO profile height although providing a larger offset at higher SZAs is not so important (case 7). Also the consideration of a NO_2 Ring spectrum (Appendix A3) is providing a significant improvement. It is also necessary to include in Ring spectra calculated at two temperatures (case 11) as well pseudo absorbers accounting for changes of the slit function (Beirle et al., 2017).

Not important in the context of the investigated fit settings is the use of the (theoretically more accurate) mean of the normalized earthshine spectra (Appendix A1.2) instead of the mean of the earthshine spectra (case 5). Besides that also the offset correction λ2 /I0 term can be neglected. Also the intensity weighted convolution (case 14) is considered optional leading to a correction of only about 3 times below the current accuracy level"

Table 2. Specific settings and results for the sensitivity studies, for details see Appendix B.

Nr.	Implemented modifications with respect to the standard fit settings	Systematic offset ($\times 10^{13}$ cm^{-2}) at SZA 90° (85°) min...max	Standard dev. of the binned mean ($\times 10^{13}$ cm^{-2}) at SZA 90° (85°, 60°) max	Autocorrel. coeff. at lags across track of 1(20) pixels for 25 Nov 2018 (NH) (%)
-	Standard scenario	-1.2...1.4 (-0.9...0.4)	1.80 (0.65, 0.29)	4.2 (0.5)
1	Calculation of the OClO SCD from fitted OClO + OClO$\times\lambda$ terms at 377 nm (instead of 379 nm)	**-1.8**...1.2 (**-1.4**...0.2)	1.63 (0.59, 0.27)	**4.5** (**0.9**)
2	Retrieval without the OClO$\times\lambda$ term	**-2.2**...1.2 (**-1.8**...0.1)	1.57 (0.57, 0.26)	**5.2** (**1.4**)
3	Slightly different fit window I (363 – 391 nm) (as e.g. by Kühl et al., 2004a)	**-1.3**...1.4 (**-1.0**...0.3)	1.78 (0.65, 0.29)	**4.7** (**0.8**)
4	Different fit window II (365 – 389 nm) (as e.g. by Oetjen et al., 2011)	-0.7...**2.5** (**-1.4**...**1.0**)	**2.09** (**0.76**, **0.34**)	**4.7** (**0.9**)
5	Fraunhofer reference as daily mean of the earthshine spectra (Appendix A1.1) instead of the mean of normalized earthshine spectra (Appendix A1.2)	-1.2...1.3 (-0.9...0.3)	1.80 (0.65, 0.29)	**4.3** (**0.6**)
6	BrO correction (Appendix A4) taking the wavelength dependency of the BrO SCD into account assuming (Fig. A1) a profile peak at 20 km (instead of 17 km)	-0.9...**2.0** (-0.8...**0.5**)	1.80 (0.65, 0.29)	4.2 (0.5)
7	BrO correction (Appendix A4) without accounting for the wavelength dependency of the BrO AMF	-0.4...**2.8** (-0.7...**0.6**)	1.80 (0.65, 0.29)	4.2 (0.5)
8	BrO correction (Appendix A4) not applied	-0.2...**4.4** (-0.4...**1.7**)	1.80 (0.65, 0.29)	**4.5** (**0.7**)
9	BrO correction not applied, but the BrO cross section included as a fit parameter in the OClO fit	**-2.4**...**2.1** (**-3.3**...-0.3)	**2.18** (**0.80**, **0.37**)	**11.2** (**3.9**)
10	NO_2 Ring spectrum (Appendix A3) is excluded	**-2.8**...0.8 (**-1.7**...-0.2)	1.73 (0.63, 0.28)	**5.0** (**0.7**)
11	Only Ring spectra for one temperature (280 K)	-0.9...**2.4** (-0.5...**1.2**)	1.80 (0.65, 0.29)	4.0 (0.5)
12	Fit without the slit function pseudo absorbers (Beirle et al., 2017)	-1.1...1.3 (**-1.3**...0.2)	1.77 (0.65, 0.30)	**9.2** (**1.9**)
13	Offset correction $\lambda^2/I0$ term excluded	**-1.3**...1.4 (-0.8...0.3)	1.76 (0.64, 0.28)	3.7 (0.5)
14	Standard convolution for the trace gas cross sections applied instead of the intensity weighted (I_0) convolution (Appendix A2)	-0.8...**1.7** (-0.7...**0.6**)	**1.82** (**0.66**, 0.29)	**4.4** (0.5)
15	Same as 14 but with the offset correction $\lambda^2/I0$ term excluded	-1.0...**1.6** (-0.6...**0.5**)	1.77 (0.65, 0.29)	3.8 (0.5)

We tried to improve the English, but also rely on the proofreading service provided by Copernicus office with respect to the English of the manuscript.

-Retrieval settings: Section 2.1

-Calibration: It is unclear whether it is performed on solar measurements or on mean radiance spectra. In the latter case, atmospheric SCDs should be fitted as part of the calibration step?

The calibration is done on the earthshine reference spectra. Absorption by ozone and NO2 are considered here. We add this information to the algorithm description.

The description in Appending A1.1-2 is unclear. I don't understand what is tau_i and I_i and what the proposed weighting (section A1.2) is supposed to solve.

tau_i is the signal of the absorption features to be fitted by the retrieval coming from a single pixel, I_i is the intensity of the signal, i.e. for pixels above the clouds there will be a stronger signal than for clear scenes.

The weighting approach ensures that the mean offset of the fit result in the logarithmic domain (optical depth space) in the offset region is zero because the absorption signals are not weighted anymore by the intensities of different pixels. We show in sensitivity studies that this (theoretically better) setting in the OClO retrieval does not play a role which is a good sign: it means that there are no (or at least no spatially variable) OClO signal in the reference region which could provide an offset in case of applying the mean of the spectra in the reference region. Also it indicates that there are no other (no spatially variable) unexplained spectral structures highly correlating with the absorption structures of OClO that also would provide an offset in the retrieved OClO SCDs.

We added the required explanation "…tau_i being the signal of absorption features from a single pixel i to be contributing to absorption parameters fitted by the retrieval and the intensity I_i being the intensity of the signal, e.g. for pixels above the clouds there will be a stronger signal than for clear scenes."

Also the motivation in the last paragraph of Sect A.1.1. was improved (as a consequence of Eq. A2) "Thus, such a reference spectrum in the DOAS analysis generally would lead to an offset for the fitted parameters even in the reference region if their SCDs are not homogeneous in this region. Also if there is no expected absorption of a particular absorber in the reference region (like it is the case for OClO), the potential errors and the incompleteness of the representation of the atmospheric state by the DOAS model can in theory induce an offset because part of the signal could interfere with the absorption cross sections of the considered absorbers. Fortunatelly the performed sensitivity studies (Appendix B5) show no additional effect from these considerations to the retrieved OClO SCDs. Nevertheless, we eliminate even theoretically such an offset by considerations in the next subsection."

In Sect A.1.2. we added a note that the calculation of the weighted mean reference requires practically no additional calculation effort.

-The sentence on l106 "The effect for this application is however negligible" is strange. Why introducing something in the text which has no effect?

It is introduced because from the theoretical point of view it is a better setting avoiding an extra offset (see also previous point) and it is always better to use theoretically better approaches if they do not require any additional effort.

We reformulated the two sentences referred here to better motivate the use: "The use of the normalized spectra (Appendix A1.1.2) for the calculation of the daily mean (at practically no additional calculation effort) ensures that also spectral features that are not related to OClO but correlate with its cross-section are not producing an artificial offset. The effect of this theoretically better approach for this application is however negligible."

-the description of the Ring effect is unclear. It is explained that Ring spectra are calculated at 2 Temperatures from the reference spectrum. Do you mean the reference for SZA 60-65°? If yes, I don't understand how it is calculated. Are the 4 Ring cross-sections fitted?

The Ring spectra are calculated from the mean earthshine reference, as it would be calculated from a Sun spectrum. The Ring spectra calculation from the earthshine reference corresponds to the earlier research referenced in the study (e.g. Kühl et al., 2004b, 2006, 2008) Yes, all 4 Ring cross-sections are included in the fit.

We have also performed a test using measured Sun irradiance spectra for the calculation of the Ring spectra but did not find an improvement. Also the use of the Ring spectrum as defined in the S5P+I product does not provide an improvement. For illustration see Fig. R1.

We also made this more clear in the text: "The Ring effect is accounted for by Ring spectra calculated at two temperatures (280K and 210K) in order to account for dependency of Ring structures on temperature, which we found is important (see Appendix B9). The two Ring spectra are calculated from the Earth-shine reference spectrum and included in the fit. Each also is scaled with λ^4 according to Wagner et al. 2009 (additional two spectra). The use of an Earth-shine reference spectrum for the calculation of the Ring spectrum is in accordance with previous studies (e.g. Kühl et al., 2004b, 2006, 2008) and is found to give a slight improvement with respect to the calculation of the Ring spectra from measured Sun irradiance spectra."

A statement with respect to Ring spectrum as for S5P+I is also added at the end of Sect. 4. See the response to the Reviewer 2.

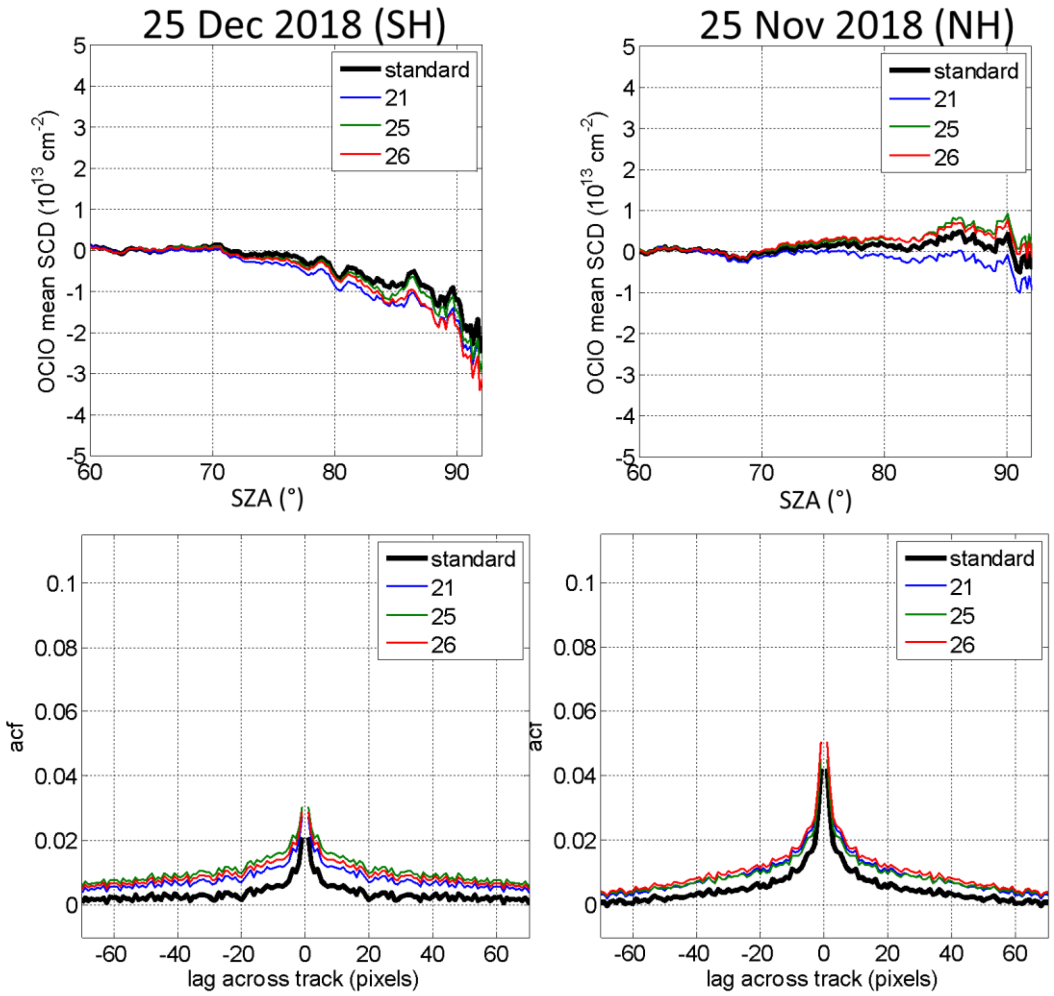

Fig. R1. Top: Retrieved daily mean OClO SCD as function of SZA (resolution 0.2°) (similar as Fig. B1 in the manuscript) for days in two different seasons for the retrieval using a Sun reference spectrum for the calculation of the Ring spectra (21), using the same Ring spectrum as for the S5P+I retrieval (25), using the Ring spectrum as for the S5P+I retrieval and also scaling it by λ^4 (26) in comparison to the standard settings (standard). Bottom: calculated autocorrelation coefficients for the mentioned cases (similarly as in Fig. B3 in the manuscript).

-Section 2.2

To help the reader, I suggest to add directly in Fig1 the indication of which days are expected to have enhanced OClO or not. In Fig1 left, the marker "x10" should be "divided by 10"?

Many thanks for these suggestions, which we implement as suggested.

-Figure 3: it would make sense to show the standard error also or instead of the std (which is already shown in Fig1).

We do not understand this suggestion. Fig. 3 already shows standard deviation of the binned mean (which assuming independent random variables is standard error of the mean). Fig. 1, however, shows mean standard deviations of single pixels. In other words, Fig. 3 does not show the same quantity as Fig. 1 and already illustrates the standard error of the binned data.

-Section 3:

-the ground-based data are not analyzed using the same settings as used for TROPOMI. It is not fully clear to what extend and how this can explain the observed differences. E.g. in Fig6 left, there is a clear offset of ~1e13 cm-2 between TROPOMI and ground-based data for low SZA. Is this related to different DOAS settings, sampling bias, other? Please discuss this in the text.

The comparison was performed with datasets of ground base datasets with settings obtained by independent studies. We already demonstrate the effect of a different setting for Kiruna (Appendix C) showing a worse result, nevertheless also there the offset is still below 2e13 cm-2. For Kiruna we have found that the usage of a reference spectrum from a different day can slightly modify the offset. Nevertheless the offset is below the accuracy of the retrieval and thus can be neglected which we will add as a statement to the manuscript. A more detailed investigation, in particular a study towards unifying OClO retrieval settings for different instruments, would be an important investigation on itself and as such is more as just a validation exercise and would reach far outside of the scope of this paper.

-Interestingly at Neumayer, the scatter of the SCD differences is much higher than for Kiruna. Is it because the SCDs range is larger? Or is there an instrumental related difference? Or something else? Please elaborate.

We can only speculate for the reason of larger scatter of the differences at Neumayer. Surely the SCD range and diurnal variation of the SZA is larger there because of the different latitudes of both sides and the specific TROPOMI orbital properties. There are also systematic differences in the difference plots from year to year (Fig. 8) which results in larger scatter as can be seen in Fig. 9, left. Also different retrieval settings compared with the Kiruna analysis could play a role but this cannot be confirmed without additional investigations.

We add: "We can speculate that the scatter for Neumayer in comparison with Kiruna is larger because of the different latitudes of both sites and the specific TROPOMI orbital properties along with the different retrieval settings."

-Section 4: *it would be good to understand what is the dominating factor explaining the offset between the 2 OClO data sets. I imagine it is probably related to the use of irradiance as reference spectrum and it is likely the largest source of error of the retrievals.*

We have found that the use of an irradiance spectrum as reference spectrum does likely not explain the differences as it would lead to a rather constant offset along the whole orbits. The comparison of both datasets to the ground based data revealed that the difference is limited to high SZAs for cases with low OClO. We added the information to the paper that the reference spectrum can likely not explain the offset. See also the response to the Reviewer 2 for more details.

-The Appendix B is hard to digest. I suggest to add a summary table (extending Table B1) in the main text with typical errors on the SCDs coming from the main sensitivity tests so that the reader can have a rapid idea of what matters and what not.

This comment comes back to the general comment at the beginning of the review. We will follow the suggestion and add the suggested table and to the main text (see the answer to the comment about the summary of the sensitivity studies above).

How the errors from the sensitivity studies are relevant compared to the typical OClO values and the differences from the validation exercise?

Given that the differences in the validation exercise can include additional errors (e.g. different radiative transfer, collocation, instrumental and retrieval settings), it is probable that some of the retrieval settings for TROPOMI could be still relevant even if they cause smaller differences than found in the validation exercise. Thus we think that the errors from the sensitivity study should be put in a relation to the retrieval errors of the standard setting as already estimated in Sect.2.2.1. Thus we list in the summary table of the sensitivity study the errors of the standard scenario as well and compare the performance of the sensitivity cases with the performance of the standard scenario.

-Sensitivity studies 5, 6, 13,14,15 have very little impact on the results. Consequently, one could argue that the related settings introduced are not really necessary. E.g. the mean of normalized earthshine spectra, the offset correction quadratic term could be optional.

Yes, of course. We will add a discussion for this along with the information in the new table as suggested above.

Minor comments

-Abstract: the first 10 lines are too generic for an abstract and should belong to the introduction section. "OClO" is defined twice in the abstract.

We remove the sentences from the abstract. The information is already provided in the introduction.

- *"so called" -> "so-called"*

corrected

-wording such as "Last but not least" should be avoided.

We reformulate affected sentences

-lines 315-316: a reference to a next section (Sect. ??) is erroneously made. Please remove.

Here should be a reference to the manuscript https://acp.copernicus.org/preprints/acp-2021-600/ which in an earlier stage was part of the manuscript presented here. In that earlier version it was presented as a separate section to which a reference was made. After suggestion by the former editor, we split the original paper into a technical part (AMT) and a part with TROPOMI results and their meteorological interpretation (ACP). We oversaw the old formulation while splitting. We add now the correct reference to the second manuscript.