

Response to the Reviewer #1

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

General Comments:

In this work, Differential optical absorption spectroscopy (DOAS) technique is applied to TROPOMI data to obtain OCIO Slant column densities (SCDs), for Arctic and Antarctic latitudes, from November 2017 until October 2020. These SCDs have been also compared with meteorological data from the ECMWF model (temperature and potential vorticity) and CALIOP PSCs observations. Through this study, the temporal and spatial evolution of the OCIO SCDs can be examined, as well as the correlation with the studied parameters, allowing also identifying possible causes of chlorine activation. A comparison between both hemispheres has also been presented, and some interesting unusual episodes concerning formation, development or deactivation of polar vortex have been studied.

The research performed in this work has been clearly presented and explained and represents useful information for the Atmospheric science community. Thus, I think that this paper should be published in ACP. However, I think that some questions should be clarified.

Specific Comments

- Has some cloud-screening been applied to the DOAS data? Could tropospheric clouds have a significant impact in the presented DOAS measurements?

No cloud screening has been applied. Since OCIO as a stratospheric trace gas is above the tropospheric clouds, no cloud shielding occurs. There can still be a small effect on the air mass factor due to the dependency of multiple scattering effects on the backscatter albedo (up to 5-10%) which, however, certainly would not justify a cloud filtering.

We add to the manuscript at the end of the paragraph about L95: “Furthermore, the occurrence of OCIO in the stratosphere ensures that no cloud filtering needs to be applied because no shielding by tropospheric clouds is expected.”

- Page 5, lines 133-135: Most of the information provided by the DOAS measurements come from air masses located at certain altitude and distance from the observation point, depending on the geometry of observation, Solar zenith angle, etc.. Has been this taken into account in the comparison between the TROPOMI and the ECMWF or CALIPSO data? Is this what you mean when talking about the multilinear interpolation? Do you use a spherical radiative transfer model to do so?

The described collocation procedure considers the instrument viewing geometry by interpolating the meteorological data to the geographic coordinate along the instrument's line of sight at 19.5 km (as already stated in the paper). The multilinear interpolation means a trilinear interpolation of the meteorological parameters to this coordinate (latitude, longitude) as well as the time of the measurement. To make it more clear, we replace “multilinear” by “trilinear” in the manuscript. The consideration of radiative transfer would necessarily require

a-priori constraints about the concentration variability along the light path which, given the high spatial variability of the OCIO number density, would mean a dependence on additional constraints on the atmospheric state like chemical composition and PSC distribution which would introduce additional uncertainties. Thus no radiative transfer modelling is applied in these calculations. In response to the comment of the reviewer and also given that such an investigation up to our knowledge has not been done so far, we performed a sensitivity study by means of a 3D radiative transfer model to estimate the range of the possible sensitivity area of the OCIO SCDs measurements. Also the possible effect of a horizontal shift of the comparison location towards the Sun is investigated. We found that the effect on the comparison is rather limited thus not affecting the findings of the manuscript.

We added the following statement about these findings to the paper:

“No radiative transfer modelling is applied during the assignment. Radiative transfer effects indicate that the mass centre of the sensitivity area of the measured OCIO SCDs is expected to be located towards the direction of the Sun from the line of sight coordinate. The consideration of the radiative transfer would require a-priori constraints about the spatial variability of the OCIO number density. Given its high variability and also the dependence of RTM on additional constraints on the atmospheric state, especially also the highly variable PSC distribution, it would introduce additional uncertainties. We have found in sensitivity studies (see Appendix A) that this displacement is expected to be less than 100 km and typical PSC concentrations do not largely affect it. It is thus below the resolution of the applied meteorological data set and the systematic effect on the performed comparison is estimated as rather limited (variation in temperature of 1K and below and in potential vorticity of 5PVU or below), therefore not affecting the findings of the study.”

We also provided the details of the investigation in the Appendix A

- Second panel from top of figure 2 and similar figures: Just as suggestion, the colour scale of these colour maps are contrary to the rest of the panels of these figures (red means low values of PV and blue means high values). Perhaps, using similar colours scale for all the panels would be more visually intuitive.

We selected a contrasting colour scale for this panel because it shows a different quantity in contrast to the other panels. But we can follow the suggestion and use the same colour scale if this seems more intuitive.

- Figures using “Longitude” as Y axis: even if positive and negative values of longitude are usually assigned to East and West longitudes, respectively, this should be clarified somewhere in the figure captions or in the text.

We added this clarification in the figure captions.

- Page 12, line 211 and page 13, line 212: The provided longitude values correspond to East longitudes instead West longitudes, Is it right?

Yes, indeed. We corrected this typo.

- Page 16, line 242: The provided OCIO SDCs values include also those below the detection limit?

We do not filter the OCIO data set in the figures just to show SCDs above the detection limit. Instead we have discussed and provided the detection limit in Sect. 2. We just pay attention here that the observed enhancements during the last days of November are very small (technically below the detection limit) but discuss them since they are persisting for several days (hence they seem statistically significant)

- Page 28, lines 407-409: The commented exceptional OCIO increase could be related to aerosols, as commented previously by the authors (page 3, line 59)?

In principle we agree with the reviewer that there could be a relation. Indeed we see increased backscatter ratios in May 2020 comparing to those in previous years. However we do not see a clear local correlation between the backscatter ratios and OCIO SCDs when they are at low levels. We added this information to the text by changing the description for the SH winter 2020:

So far we do not have a clear explanation for this finding except of increased backscatter ratios in CALIOP data in May 2020 compared to those in previous years. For the polar mean PSC evidence (..) values distinguishable from zero can be observed already at the beginning of May which was not the case for the previous SH winters. The local PSC evidences (..) have sporadic values slightly above zero which however seem not to be correlated with the collocated SCDs (top panel). Also we do not see a clear local correlation between the backscatter ratios and OCIO SCDs when they are at low levels (see Appendix B).

We modified also the last paragraph of the conclusions:

Further investigation are still needed with respect to the exceptional OCIO increase which goes along with increased backscatter ratios compared to previous winters but is not correlating with the stratospheric meteorology in late March and April in 2020 in the SH where a larger OCIO SCD signal above the typical uncertainty range was observed ($5E13 \text{ cm}^{-2}$) which is also observed in the S5P+I data.

Technical Corrections:

- Some sentences are too long. I think some “,” should be introduced. As example: Page 2, lines 29-31; Page 6, line 166: “For the comparison, ..”; Page 6, line 169: “In addition, ..”; Page 6, line 166: “For this winter, ..”; etc.

We proceeded as suggested. We also rely on the English proofreading service offered by the Copernicus office.

- Page 4, line 113: Introduce the meaning of the ECMWF acronym

We introduced now the meaning at the first occurrence (same page, line 99)

- Page 5, line 135: “..19.5 km of altitude”.
- Page 5, line 137: “..The obtained correlative dataset..”.

All corrected as suggested