Response to Referee #2 Dr. C. Sioris

We thank reviewer for his energy to review our paper. We greatly acknowledge his comments and suggestions that helped us to improve the quality of our paper. Below, we present the detailed replies to each comments. Note: original comments of the referee are given in bold.

This paper contains some interesting results such as the OClO layer in the equatorial stratosphere, but the main weakness of the paper is that the focus of this 'climatology' paper is not on the OClO number densities. In fact, it seems unlikely that the authors retrieved OClO from the annual or monthly SCD profiles in the equatorial region, since they are not discussed. I estimate that ~800 inversions would be required.
Annual: 18 latitudes * 9 years = 162 profiles
Monthly: 12 months * 6 latitudes * 9 years = 648 profiles (everywhere except Antarctic, overestimate)
+ 3 months * 1 latitude * 9 years = 27 profiles (Antarctic)
If the authors choose not to revise the paper in terms of OClO number density climatologies, they should state whether the reason(s) was due to computing time required, lack of retrieval automation, the quality of the inverted profiles, etc.
The altitude of the SCD peak will generally lie below the altitude of the number density peak. So discussion of layer altitudes should either be in terms of number density or the authors must specify in each instance that they are referring to an SCD peak height. Statements such as "the presence of an OClO layer (...) at about 35 km" in the abstract are a bit misleading.

<u>Response:</u> The question of the use of slant column densities instead of vertical profile of concentration is quite relevant. You are right to say that the use of OClO concentrations profiles would increase the number of potential users. However, we have decided to keep the SCD in the revised version of our manuscript for several reasons that we detail hereafter. First of all, even though it was originally intended to retrieve OClO using single GOMOS measurements, it turned out impossible to do it because of the low signal-to-noise ratio of a single GOMOS measurements. Thus, OClO is not retrieved with the operational processor and has never been included in the official distribution of GOMOS level 2 data. Our OClO product is <u>an off-line product</u> and, as such, to deliver them in the form of SCD with "reasonable" error bars appears to us to be the best thing to do. Secondly, the retrieval of the OClO product is based on a <u>statistical analysis</u> of several co-located GOMOS measurements. We have to keep this in mind. The retrieval is not based on a single measurement, a preliminary step is required to build a "virtual" measurement that we will use to retrieve OClO. This additional step is a first source of uncertainty. The second step is the spectral inversion (DOAS) used to retrieve the SCD of the different species involved in the attenuation of the radiations. Here again, this step implies

some uncertainties. Thereafter, it is always possible and easy in the case of GOMOS to perform a spatial inversion to retrieve vertical profile of concentration but the error bars become too large. It is difficult to be confident with a product with very big error bar. We are at the limits of the possibilities of the GOMOS instrument. Only the SCD provides error bars that make the product usable for seasonal studies, latitudinal studies,… The SCD relative errors extend from about 5% to 70% at some levels. A second panel inserted to Figure 5, shows the vertical profile of the SCD relative errors. On the other hand, the relative errors concerning the vertical profiles of OClO concentrations are generally greater than 60% and can reach about 180%. These values are too important to use scientifically the OClO concentrations. Nevertheless, we have used vertical profiles of concentration to perform the comparison with the balloon observations (Fig. 6) because we have only the concentration profiles derived from these balloon measurements. A warning has been added in the text explaining the limitations of this comparisons which is in fact a simple verification. Moreover, Figure 9 has been removed because it shows the anti-correlation between NO_2 and OClO in terms of concentration. It is a nonsense to keep this figure in the text and to affirm that OClO concentration is not a "good" product. Furthermore, we are agree with Dr C. Sioris about the altitude difference between the number density peak and the SCD peak. Thus, in the revised paper, we write 'OClO SCD peak' instead of 'OClO layer' where relevant. A small paragraph has been added at the end of section 3 to explain why we do not use the vertical profile of concentrations. We have also decided to change the title of our manuscript: "OClO slant column densities derived from GOMOS averaged transmittance measurements".

Scientific comments:

P3513 - The authors should also discuss denitrification which involves hydrolysis of N2O5 to form nitric acid

Response: you're right. Since HNO_3 is a reservoir species for NO_2, this denitrification leads to low NO_2 concentrations in the stratosphere and therefore the reaction between ClO and NO_2 is limited. This has been added in the text.

P3513L7- credit to the authors for making this point: done.

P3516 - The mission baseline scenario is not too relevant. From Fig. 1, it appears that "coverage" is near-global (50°N-80°S) and takes several months to achieve. This statement is misleading.

Response: Figure 2 are not discussed in the section 2 (general description of the GOMOS instrument). Figure 2 corresponds only to the GOMOS measurements used to retrieve OClO in our studies. For clarity, this has been added in the caption of Figure 2. In the section 2 (The

GOMOS instrument), we consider all the GOMOS measurements (without restriction), and a global coverage takes effectively about 3 days to achieve. In addition, Figures and captions 1 to 3 are confusing , we have addressed this issue in the revised paper (now, the caption corresponds to the right figure, we are sorry about that).

P3519L17 – This is not the cause. The difference in transmittances is more likely related to older air (smaller air densities) inside the vortex than outside of it. The transmittance difference between the two modes is 0.06. This implies that 23% (= 1-0.2/0.26) of the 404 nm light at a tangent height of 20 km is absorbed by OClO. This is easily 2 orders of magnitude more than OClO absorbed could explain without even considering transmittance changes due to NO2.

Response: You're right. We have added in the text that the difference in transmittances is related to older air inside the polar vortex. We still kept in the text that a part of the difference observed can be attributes to the low concentration of NO_2 inside the polar vortex.

P3519L23 – What is used to weight the median transmittance? Provide a reference for the jackknife method.

Response: the transmittances are weighted with respect to their estimated measurements error (added in the text). We have also provided a reference for the jackknife method (Quenouille, M.H.: Note on bias in estimation, Biometrika, 43, 353-360, 1956)

P3520 – I appreciated having Eq. 3. Using the reduced chi-square statistic, the authors should consider whether a 2nd order polynomial is sufficient to account for the slowly-varying component, particularly for the 355-425 nm window. It may not be a coincidence that the window with the smallest residuals is the one with the narrowest fitting window. Furthermore, the authors should test the impact of not detrending the absorber cross-sections of OClO, NO2, and O3. They may be throwing away valuable information particularly for the 355-425 nm window in doing so. As well as comparing residuals, it makes sense for the authors to check whether the OClO SCDs from the 355-381 nm window are biased relative to the other windows.

Response: You're right. The choice of a second order polynomial to fit the slow-varying component has been carefully considered. Indeed, various functions were tested for all ranges of wavelengths. It turned out that a second order polynomial was very suitable. Therefore, we found it unnecessary to use higher order polynomials, it would only increase the computation time without add any values. We have added a sentence explaining this in the text: 'Several other functions have been tested and it appears clearly that it is sufficient to use a second order polynomial to fit the slowly-varying component.'

We have considered that the absorption cross sections of OClO, NO_2 and O_3 contained also two components. The first slowly varying is fitted using a second order polynomial (it represents the global trend of the cross sections) and the second is the rapidly varying component which is used to express the modeled differential transmittance.
Finally, we have compared the OClO SCDs obtained from the 4 spectral windows selected. Almost no difference is observed. Therefore, the choice of the wavelength range can not be based on this.

P3521 "extracted from the jacobian matrix". Please elaborate.
Response: . We have defined more precisely the way we extract the retrieval error. Once we have found the slant column densities of each species by using a minimization of the chi-square function defined in the text, we derived the random retrieval error using the hessian matrix of the chi-square function as explain in Press et al. (2007). We have added a sentence in the section 3 explaining this calculation: "Once the SCDs retrieved, the errors made ΔN_{gas} are given by the root square of the diagonal elements of the error matrix (the inverse of the curvature matrix which is equal to one-half times the Hessian matrix of the chi-square function)". We thank the referees because thanks to their comments about uncertainties we have found a small error in our computation of the error. It has been corrected and the new error bars appear in the revised version of our manuscript.

P3521 – How small do the SCD errors get (best-case altitude and latitude)?
Response: We have added a panel in Figure 5 showing the vertical profile of relative errors for the 3 latitude bands. The range of errors extends from 5% to about 70%. The best case (5%) is for the Antarctic latitude band at 20 km.

P3522 – The reason it is not easy to validate is that measurements by balloons are geographically sparse. It does not have to do with the fact that this product is "new".
Response: agree. We have removed the first sentence of the paragraph.

P3523 – For validation, it is not clear whether the mission-average or the year of the correlative balloon flight is used when using the 20-day window. The authors should try both and go for the one that provides better agreement and explicitly state whether the 20 day window is for one year or all years. The authors should also look at whether the transmittances have a bimodal nature during this 20-day window (if they have not done so) and state this explicitly. If the authors separate the modes, the authors should state that they are picking the "in the vortex" mode. The authors should also try increasing the latitude range to 60-90°N (entire polar region) and then could try reducing (to <20) the number of days in the window. The trend over 20 days, particularly in early March is not linear. Between January and February, relative decreases may be more minor, whereas between February and March, the decrease can be 1 order of magnitude in some years. The authors could use their monthly time series to find the mission-averaged monthly variation and use it to weight the date of the window center. I suggest this because the low bias for the Sirius occultation could be a result of the window center. The lack of bias for the Alnilam case may be a fluke.

Response: Thanks to the comments of all the reviewers about this point, we can not talk about validation using this simple comparison. It is just a simple verification and it has to be considered as such. We have added a sentence at the beginning of this section explaining this: "… as a result, the following discussion should be considered as a simple verification and not as a validation of the GOMOS OClO product.".

We are agree with all the comments of the reviewers but his comments will be taken into account in the future when a complete exercise of validation will be done (including satellite instruments and photo-chemical models).

We are using a 20-day window corresponding to the year of the balloon flight. This has been clarified in the text. Following the advice of the reviewer, we have done the comparison using an average over all the years and the best agreement is achieved using one year (those corresponding to the balloon flight).

All data sets used to compute averaged transmittances have been statistically analyzed, including those used for comparisons with the correlative balloon flights. For the two cases studied here, no bimodal distribution have been detected. Now, this is stated explicitly in the text.

There is no need to increase the size of the bin for March 2003 (AMON comparison) because there are no useful measurements available above 75° (see Fig. 2). For the other balloon measurement, indeed, if we increase the latitude range to 60-90°N, we have some measurements above 75°N but they occurred about 6 days before the date of the balloon flight. Thus, if we reduce the number of days in the window, there are no measurements above 75°N. Therefore, we have kept our 20 days window and the 60-75°N latitude range.

We are agree about the problem of the trend of OClO over 20 days, particularly between February in March. The proposal to weight the date of the window center using our monthly time series is really interesting and will be implemented in the frame of the future complete exercise of validation.

P3524 – include error bars for all number densities on this page.

Done

P3524 – Mention why the vertical sampling of the two AMON profiles is different.

Response: They differ because the two flights does not occurred at the same date (1 and 3 March 2003) and the meteorological conditions were not the same. Thus, the AMON balloon has ascended faster for the occultation of Alnilam. This has been added in the text.

P3524L10 – "for the entire altitude range" -> "for most of the altitude range" (see e.g. 27.5 km).

done

P3524L14 – The 2 km vertical resolution of SALOMON is not the likely cause of difference in OClO number density peak height. The vertical resolution for GOMOS is 1.7 km (for vertical occultations), so there is consistency between the vertical resolutions of the instruments. Furthermore, the SALOMON measurement at 20 km that has a 2 km vertical resolution, depending on the shape of the averaging kernel, will likely be sensitive to the true number

density in the 19-21 km range and the GOMOS one at 18 km will be sensitive to the true profile between ~18 km and ~19 km. A more likely explanation is that the peak height "can vary according to the" specific "area of the vortex".
Response: We are agree. In the text, we have written that the vertical resolution of the two instruments are close and we have added that the explanation of the different peak heights can be attributed to the location of the measurement in the polar vortex.

P3524L21 – "very well ... slightly less well" -> "well ... less well". This statement is repeated in p3528L8. Also the conclusion of sufficient quality for scientific use depends on the application. I would be willing to use GOMOS OClO number density data to study seasonal evolution of peak height, but I do not feel the community knows enough from comparing with these three correlative profiles whether we can do quantitative comparisons with model to identify problems with these models.
Response: Agree. We have tempered our conclusions about the scientific use of the GOMOS OClO product, both in section 4 and in the conclusion by explaining that, at this point, the quality of this product is sufficient for seasonal or latitudinal studies but that a more thorough validation is required to use these data whatever the scientific application.

P3525 – As with the abstract, analysis of the layer height should use inverted data.
Response: as specified previously and expected by Dr. C. Sioris, in the revised version we talk about the altitude of the OClO SCD peak (instead of the altitude of the layer).

P3526L14 – Is this the maximum at any tangent altitude (15-45 km)?
Response: These values (5e15 to 3e16 cm-2) correspond to the maximum OClO SCD reached each year in the lower stratosphere in the Arctic region. 5e15 cm-2 is the maximum reached during the winter 2003 /2004 and 3e16 cm-2 is reached during the winter 2010/2011.

P3526L16 – The number density range is much narrower than the SCD range. I would expect the opposite. As in previous comment, what do 5 and 9 e-7 cm-3 represent? Are they the maximum monthly-mean number density for each year at any altitude?
Response: Yes, you're right. We would also expect the opposite so we have checked our calculations. They are correct. These values represent the maximum number density reached during each winter in the lower stratosphere. These values are in the range of the maximum observed by the balloon-borne measurements (see Tab. 1).

P3527- see 1st scientific comment (p3513): N2O5 + H2O -> 2 HNO3
Response: Yes, you're right but Figure 9 has been removed and the discussion about it also.

P3527L6 – One or two months in austral autumn appear in some years.
Response: We have added this point in the text.

P3527L11 – The range of number densities is more reasonable given the range of SCDs, but still oddly small. I wonder if there is a retrieval issue that also led to the decision to not retrieve in equatorial regions.

Response: We are agree, this range is more reasonable. The reasons why we have not performed the concentration retrieval have been already discussed above. There is no retrieval issue. Furthermore, we have retrieved OClO SCD in the equatorial region (middle panel of the last Figure).

P3527L22 – If you retrieved OClO number density profiles and uncertainties for each month, you could see if it is significantly higher than the 'background' level in those time periods. If you don't bother with inversions, you could still more compare the SCDs to see if the enhancements are statistically significant considering respective uncertainties.

Response: We are agree, thank you for this helpful comment. In a first step, we have computed the average of the maximum OClO SCD for each months (excluding the 2 periods mentioned in the text). It corresponds to the maximum of the background level of OClO (it is equal to 3.6e14 cm^{-2}). The associated uncertainty is equal to ~9%. Then we have calculated the average of the maximum OClO SCD for the two periods mentioned in the text. We have found respectively 5.03e14 cm^{-2} and 5.54e14cm^{-2} and the associated uncertainties are 21% and 25%.

Taking into account the uncertainties, the background level of OClO SCD is between 3.3 and 4e14 cm^{-2} whereas the maximum for the first period is between 4 and 6e14 cm^{-2} and for the second period, between 4.15 and 6.9e14 cm^{-2}. Since the former interval is below the two others, we can conclude that these enhancements are statistically significant and that it is justified to keep them in the text. This has been added in the text.

Fig. 6 – The GOMOS error bars seem driven by natural (OClO) variability. This is a good sign.

Agree

Editing:

All suggested corrections have been taken into account.