Dear Editor,

With regard to the manuscript:

MS-NR: acpd-2010-0522
Title: NDACC UV-visible total ozone measurements: improved retrieval and comparison with correlative satellite and ground-based observations
Author(s): F. Hendrick, et al.

Please find below the replies to Referee #2 comments.

Sincerely yours,

F. Hendrick (**franch@oma.be**)

Anonymous Referee #2

First, we would like to thank Anonymous Referee #2 for his/her helpful comments.

In this paper, F. Hendrick and colleagues report on a new set of recommendations for the retrieval of ozone total columns from NDACC zenith-sky UV/vis observations. The recommendations are described, the error budget is discussed and the new settings are applied to a large set of SAOZ observations which is then compared in detail to satellite ozone measurements and also some Brewer and Dobson observations. The paper is well written, the analysis is thorough in many aspects and the results are interesting for people working in the stratospheric ozone field. I therefore recommend publication of this manuscript. However, I also do have some concerns about this paper as discussed below. The authors need to address these points in detail and change the manuscript accordingly before it can be accepted for publication.

1) In my opinion, this paper would probably be better suited for AMT(D) as it reports on retrieval techniques and validation but does not really contain any new information on atmospheric composition or atmospheric processes.

We agree that the first part of the paper dealing with the description of the settings for the retrieval of ozone columns from ground-based UV-visible observations is technical. However, we think that the second part about the comparisons with satellite and Dobson/Brewer, may have a significant impact on science issues since all these data sets are used in numerous studies and therefore it deserves to be published in ACP. Our main point is that accurate long-term monitoring of total ozone is one of the most important requirements for identifying possible natural or anthropogenic changes in the composition of the stratosphere.

2) In the title and in several places in the paper, reference is made to the NDACC UV-visible observations. However, the analysis and comparisons shown are limited to SAOZ instruments, which are an important part of the NDACC UV-visible network but not identical to it. In particular, the comparisons between V2 and V1 of the SAOZ analysis should not be equated with a comparison of the old and the new NDACC data analysis. I'd recommend making this difference more clear in the text and also in the title of the paper.

The DOAS and AMF settings described in the paper have been generated for application to any ground-based zenith-sky UV-visible instrument measuring ozone at twilight. The impact of these settings on the retrieval of total ozone is illustrated in the paper on a sub-set of SAOZ stations representative of the NDACC UV-vis network since a large number of the UV-vis instruments measuring routinely ozone at twilight in NDACC are SAOZ spectrometers. It is anticipated that other instruments will apply those recommendations in the future.

At the end of the Introduction, we have replaced "Here we report on these recommendations and illustrate the benefit of their use by a comparison between resulting total ozone derived from the French led NDACC/SAOZ (Système d'Analyse par Observation Zénithale) network and collocated observations performed by other instruments."

by

"Here we report on these recommendations and illustrate the benefit of their use by a comparison between total ozone measurements made by a selection of SAOZ (Système d'Analyse par Observation Zénithale, Pommereau and Goutail, 1988) spectrometers belonging to the NDACC UV-visible network and collocated observations performed by other instruments."

3) The new NDACC recommendations have two parts – one for the retrieval of the ozone slant columns, the second for the airmass factors. While the latter part is discussed in detail, the first part is only briefly mentioned and the discussion, in particular with respect to uncertainties is much less convincing. First of all, I think it is absolutely necessary to indicate what the V1 retrieval settings were, and how they relate to the settings used in previous papers applying NDACC values for satellite validation. The changes from V1 to V2 discussed in the text are interpreted as AMF changes only – does this imply that the other settings remained unchanged? And if other settings have changed as well, wouldn't it make sense to investigate what the relative importance of these changes (cross-sections, wavelength window, Ring parametrisation) are?

We have added in the beginning of the Section 4 a paragraph on the changes in the DOAS settings between V1 and V2 of the SAOZ data sets including a discussion of the impact of Ring and ozone cross-sections and ozone fitting window. Overall, the main change between V1 and V2 after applying the NDACC UV-Vis working group recommendations for DOAS settings is a decrease of ozone vertical column at twilight by 0.5% only, which is not significant.

Second, the uncertainty in the slant column is estimated by assessing the uncertainty in O3 cross-section and the variance in results of three different fitting codes on the same spectra. This is in my opinion not the full story – I would hope that least squares retrievals using the same settings on the same data should provide the same results within some limits, but this does not tell me the uncertainty in the slant column. There is uncertainty introduced from the measurements (noise, slit-function, straylight, temperature drift, etc.) and also from the analysis (choice of fitting window, cross-sections, polynomial, etc.). Together, this will be significantly more than the 1% cited in the paper, and I'm sure the authors will agree that if I put two different NDACC UV-vis instruments side by side and then compare the results, they will not agree within 1 %. I therefore think that the error discussion for the slant columns needs to be revised and extended and the estimates need to be more realistic. Also, I don't think Fig. 2 is adding any information, in particular as nothing is said on what the different scenarios were for which results are shown.

We agree. Section 3.1 (see page 11 of the revised manuscript) has been completely modified: the comparison of the different fitting codes has been replaced by a discussion on the different error sources in the DOAS fit and in the determination of the residual amount in the reference spectra. We have also added a new figure illustrating a typical DOAS fit result and the contributions from interfering trace gases in the 450-550 nm spectral interval (see fig. 2 on page 52 of the revised manuscript).

4) The changes in the AMFs proposed in this manuscript are relatively large and show a significant seasonality. The arguments given for the use of a seasonal and latitudinal climatology of ozone profiles are convincing and I believe the new AMFs are more realistic than the constant values used before. However, these problems have been noted and discussed before e.g. in work by Lambert et al., and I'm surprised that these previous results are not mentioned more in the current manuscript.

We agree that the problems related to the use of a constant airmass factor, the temperature and SZA dependencies of satellite retrievals have been identified for long (Van Roozendael et al., 1998; Lambert et al., 1999; Høiskar et al., 1997). Although several attempts were made to improve the retrieval of ozone at specific stations, here the objective is to provide a coordinated solution consitently applicable to all NDACC UV-vis stations.

I'm also surprised by the large change for Jungfraujoch (nearly 10% or 30 DU in winter)– is that because of the altitude of the station, and why has it not be noticed and corrected before as there is plenty of other ozone measurements available at this site?

In Figure 2 of the ACPD paper, we show the difference between the annual mean SAOZ AMFs and those extracted from the new AMF climatology. The plot for Jungfraujoch is misleading because in the data available in the NDACC database, the AMFs used for this station have been corrected a long time ago for the seasonal variation of the ozone profile and the altitude of the station. Therefore, the Jungfraujoch plot is not relevant and we have decided to remove it in the revised version of the paper (see now fig 3 of the revised manuscript).

Another surprising result are the AMFs for Bauru – I think there is no good reason for the large scatter in AMF values observed at this tropical site and would see this as indication for a problem in the LUT used.

This larger noise can be explained by the variability of the ozone profile shape above the Tropical Tropopause Layer (TTL) at altitudes from 20 to 30 km where the measurement sensitivity is largest (see Fig. 1), which therefore means a more significant impact on the AMF. As an example, the AMF in January in Bauru varies from 16.5 to 17.0, that is by 3%, when using ozone profiles extracted from the TV8 climatology for typical total column values of 244 and 278 DU. Thus, small changes in total ozone of about 50 DU on a few days time scale, as frequently observed in Bauru, result in significant changes in the AMF. For comparison, the AMF at 65°N in April corresponding to total column values of 332 and 417 DU (typical values around the mean total ozone column value at Sodankyla) is varying from 16.8 to 16.9, respectively, which corresponds to a change of 0.5% only. This explains the smaller short-term variability in the AMF at mid- and high-latitudes.

We have added the above paragraph in the revised version of the manuscript (see end of page 10 of the revised manuscript).

5) After the initial comparison of SAOZ and satellite retrieved O3 columns, the authors proceed to discuss and correct for a temperature dependence in the difference between satellite and SAOZ results. The final result shows less seasonality and better overall agreement between the two datasets. While I'm convinced that the analysis shows a valid point (the not fully corrected for temperature dependence of the UV absorption of ozone used in the satellite data), I'm a bit worried by this approach for several reasons:

a) The authors take the variations between the seasonalities in the differences to different satellite retrievals as confirmation for the absence of a seasonal bias in the SAOZ data. I don't think this is a valid conclusion – in a comparison of two (or more) data sets, one always has to accept the possibility that all of them are off.

Indeed, the temperature dependence of at least some of satellite retrievals cannot fully explain the observed seasonalities. In the revised manuscript, we have added a study of the impact of SZA at the location of the satellites (see Section 4.2.2 page 22 in the revised manuscript). In both cases, these dependences cannot be attributed to SAOZ because of the use of Chappuis bands and constant SZA (90°). We agree that there are other possible contributions to the seasonalities from SAOZ, among which are the lack of longitudinal and seasonal variations of tropospheric ozone in TV8 as well the representation of the TV8 ozone profiles at high latitude, as shown by the comparisons between AMFs calculated from ozonesondes and TV8 profiles for Ny-Alesund and Dumont d'Urville in Fig. 4 of the revised mansucript.

b) In the analysis, the difference between satellite and SAOZ is correlated with temperature, and then a correction is applied. What would have happened, if the authors had applied the same approach to SAOZ V1 data? I assume that the final results would have looked very similar, only that the correction terms would have been larger. I do believe that SAOZ V2 is better than V1 but the authors seem to take this analysis as proof that there is no seasonal bias in the SAOZ data, and I don't think this conclusion can be drawn from the data.

The same calculations with V1 instead of V2 provide the same temperature dependence, although more noisy. But most unexpected is the large amplitude of the temperature dependence of TOMS and OMI-TOMS although their respective retrieval algorithms include a temperature correction. The fact that this dependence is smaller or event absent (OMI-DOAS) in other satellites makes difficult to attribute it to SAOZ.
There may be indeed an influence of the seasonality of the TV8 profiles at high latitude, but then why on TOMS and OMI-TOMS only. For OHP where the AMF LUTs from TV8 are consistent with those derived from the sondes, we have no other satisfactory explanation to suggest.

c) I'm concerned by the overall approach to see good consistency between SAOZ and satellite data after T-correction as validation of the new retrieval settings. While this is certainly a nice result, the SAOZ data are often used as validation data set for the satellite retrievals, and therefore should not themselves be "validated" by comparison to satellite data. The comparison to Dobson and Brewer is much more relevant in this context, as would have been comparison to sonde data. I recommend that this part of the

paper is formulated a bit more cautious making clear which data set is validating which and which statements are firm conclusions and which are just plausible.

We agree on the fact that only the comparison to Dobson and Brewer can be considered as a validation of the new SAOZ data sets. That's why this section now appears before the section on the comparisons with satellite data. The comparisons with satellites must be seen as a study of the consistency between all data sets, based on what we believe is a consolidated SAOZ data set generated by application of the newly established NDACC WG recommendations. It is certainly not our purpose to use satellites to validate the new SAOZ data.

We also modified the title of the paper accordingly: "…comparison with correlative satellite and ground-based observations" replaced by "…comparison with correlative ground-based and satellite observations"

6) The impact of tropospheric ozone needs more attention. Tropospheric ozone has several possible effects – it can enhance the observed signal, in particular in the presence of clouds, fog or snow; it can affect the comparison of satellite and ground-based observations as they have different sensitivities to the troposphere and it can change the real AMF if the true tropospheric column is different from the climatological one. In fact, the authors mention the ghost column added to the satellite observations in the presence of clouds, but at twilight, the climatological tropospheric ozone used in the AMF calculations has a quite similar role in the ground-based observations.

The discussion on the influence of tropospheric ozone is improved in several paragraphs:

- The addition of Section 2.2 (page 6 of the revised manuscript) showing the averaging kernel of the zenith sky measurements at 90° SZA. As can be seen, the sensitivity to tropospheric ozone is low, with averaging kernel value smaller than 0.5 below 10 km while the sensitivity to the stratosphere is larger (averaging kernel values close or larger than 1 in the 18-30 km altitude range). So it is clear that our zenith-sky total column measurements are strongly weighted by the contribution of the stratosphere.
- The inclusion of a discussion on the possible impact of tropospheric ozone on the residual seasonality of satellite-SAOZ and Dobson-SAOZ in OHP(see Sections 4.1 and 4.2)
- The impact of tropospheric ozone on the systematic bias between Bauru and Reunion Island (see page 26 of the revised manuscript).

Moreover, more details are now given on the impact of clouds in section 3.2 (page 14 of the revised manuscript):

“The small impact of clouds on zenith-sky ozone UV-vis measurements at twilight is due to the fact that the mean scattering layer is generally located at higher altitude than that of the clouds. However, there are two exceptions: in the tropics where thunderstorms accompanied by heavy rainfall can reach 15-16 km, and at high latitude in the winter where Polar Stratospheric Clouds (PSC) are sometimes present, disturbing the ozone measurements. These episodes are easily removed from the ground-based data series by detecting the large enhancements of 70% or more of the absorption by O_4 and H_2O in the tropics in the presence of thick clouds and rainfall, and by the use of a color index (ratio between irradiances at 550 and 350 nm) in case of PSC (Sarkissian et al., 1991).”

As a side note, it is also worthwhile to consider the risk of a circular argument when the same ozone climatology is used in the ground-based observations and in the OMI observations which are then used to derive the tropospheric column by subtracting the MLS columns. Consistency between measurements using the same assumptions does not necessarily imply that they are correct. In the case shown in the paper, the excellent agreement with ozone sondes at OHP is of course independent confirmation for the tropospheric ozone columns derived.

To our opinion, the argument of circularity is not valid here because the sensitivity of the ground-based UV-visible and satellite O3 measurements to the a priori ozone profiles (in both cases from the TV8 climatology) is different. Thus the uncertainty of the TV8 climatology will impact the ground-based and satellite retrievals in a different way.

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