

Report of the Short-term scientific mission: “Analysis of Brewer Umkehr measurements” (COST-STSM-ES1207-200915-066531)

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1. Purpose of the STSM

The primary aim of the current COST Action is the creation of a homogenized dataset of ozone, AOD and UV measurements, from the European network of Brewer spectrophotometers. This dataset will provide valuable data in organizations such as the World Meteorological Organization (WMO), the World Ozone and UV Data Centre (WOUDC), the International Ozone Commission (IO3C), the Intergovernmental Panel on Climate Change (IPCC), Global Monitoring for Environment and Security (GMES) and the ozone trend assessment panels. While it has been achieved a significant progress in the characterization of the instruments and the development of the algorithms for the total ozone column retrieval, the topic of the vertical ozone distribution retrieval, through the well-known Umkehr technique, hasn't adequately addressed yet in the framework of the Action. The interest in the scientific community for the changes in the ozone profiles is very high (see for example <http://igaco-o3.fmi.fi/VDO/>) and the EUBREWNET could significantly contribute to this demand.

The Umkehr (the word Umkehr means "reversal" in German) effect, first discovered back in 1930 by Götz (Götz, 1931), is observed when measurements are made with ultraviolet spectrophotometer, of the ratio of the zenith sky light intensities of two wavelengths in the solar ultraviolet when the sun is near the horizon. The intensity (I) in the shorter of the two wavelengths is strongly absorbed by ozone, while the other (I') is weakly absorbed. When the value of $\log(I/I')$ is plotted against the sun's zenith angle, it is observed that this log-intensity ratio decreases as the zenith angle increases until a minimum is reached for a zenith angle of about 85° . As the zenith angle increases further, the log-intensity ratio increases again. Considering light which is scattered only once in the atmosphere, the light received by the instrument at the surface is contributed by light scattered downward from all the levels in the atmosphere. The amount of light contributed by scattering at any particular level depends on (a) the number of air molecules at that level and (b) the absorption by ozone and scattering by air molecules both before and after scattering. As the height increases contribution of effect (a) decreases and contribution of effect (b) increases. For a given zenith angle, the scattered light contribution comes from well-defined layer of atmosphere, which can be termed as an effective scattering height. The effective scattering height depends on the ozone absorption coefficient and on the solar zenith angle, increasing as with each of these. The effective scattering height will always be higher for shorter wavelength, which is more strongly absorbed. As the sun approaches the horizon, the two intensities decrease, but intensity I decreasing more rapidly than I' . However, when the effective scattering height for the short wavelength is above the ozone maximum, I decreases more slowly than I' , because the ozone absorption occurs mostly in the shorter vertical path after the scattering event, and the ratio I/I' increases until the effective scattering height for I' is also above the ozone maximum. This reversal (Umkehr) or inversion implies the existence of maximum of ozone concentration at some level in the

atmosphere. The Umkehr technique has been used since the 1930s [Götz et al., 1934] to estimate the vertical ozone profile from zenith sky measurements taken at a pair of wavelengths in ultraviolet (311.5 and 332.5 nm) using the Dobson spectrophotometer [Dütsch, 1959]. Since mid-80s, Brewer instruments have also taken similar measurements at eight discrete wavelengths [McElroy and Kerr, 1995]. The Brewer Umkehr algorithm utilizes the standard five "short" wavelengths used for total ozone observations as well as three additional "long" wavelengths obtained by offsetting the diffraction grating to longer wavelengths, which allows an overlap of the last two short and first two long spectral positions. Measurements are made in the strong polarization direction (the sensor's azimuth perpendicular to the sun's azimuth), but will include some multiple scattered light.

The main purpose of this STSM was the analysis of the Umkehr record of a single monochromator Brewer spectrophotometer operating in Thessaloniki since March 1982 (B005). Systematic Umkehr measurements are conducted during afternoon since 1993 and have been analyzed just partially in the past (Kosmidis et al., 2004), not with the latest available algorithm. In addition, this STSM aimed in investigating some of the parameters that affect the Umkehr retrievals, such as the out of band stray light and the temperature dependence of the ozone absorption coefficients, and thus to contribute, in the framework of the WG2, to assess quality of the Umkehr retrieval.

2. Umkehr retrievals for Thessaloniki data during the STSM

As mentioned above during an Umkehr measurement the Brewer records the zenith sky intensity at eight discrete wavelengths, the standard five "short" wavelengths used for total ozone observations as well as three additional "long" wavelengths. The full set of eight Umkehr Wavelengths includes 306.3, 310.1, 313.5, 316.8, 320.1, 323.2, 326.4, and 329.5 nm (these are the nominal wavelengths and may differ a little in particular instruments, depending on the dispersion settings of each instrument). Thus, there is an overlap of the two last "short" wavelengths (316.8 and 320.1) with the two first "long" wavelengths. These measurements are performed during sunrise and sunset in a number of different solar zenith angles (SZA), ranging from ~60 to ~92°. The measurements from two selected wavelengths centered at ~310 ("short" wavelength) and ~326 nm ("long" wavelength) form the so-called single-pair N-value (see eq. 1.1) that is used for the ozone profile retrieval.

$$N(\theta) = 100x \log \left(\frac{I'(\theta)}{I(\theta)} \right) \quad (1.1)$$

where I' is the intensity at the "long" wavelength and I the one at the short wavelength. The N-values are then interpolated at 12 nominal SZAs (60°, 65°, 70°, 74°, 77°, 80°, 83°, 85°, 86.5°, 88°, 89° and 90°) to create the characteristic Umkehr curve. The profile retrieval is based on the algorithm UMK04 and is reported in a 16 layer scheme (see table 1), with each

layer being ~5 km thick. It is recommended (Petropavlovskikh et al., 2005) the profiles to be analyzed in 8 independent layers, consisting of layers 0+1, 2+3, 4, 5, 6, 7, 8 and a broad top layer 8+, combining all layers from 8 and above. Nevertheless, in the following the results, including the comparison with the Microwave Limb Sounder (MLS) satellite data, are given for the layers 2, 3, 4, 5, 6, 7 and 8. A future step will include the analysis in the optimized 8-layer scheme, mentioned above.

The MLS data are reported in standard pressure levels in the range 261 – 0.02 hPa (Livesey et al., 2011), with a vertical resolution of ~3 Km and they are interpolated in the Umkehr layers so as the comparison with the Umkehr profiles to be feasible. The bad quality data have been filtered according to the directions given in the MLS data quality and description document (Livesey et al., 2011). The Umkehr algorithm provides layer-mean ozone values whereas MLS output is the ozone mixing ratio at many altitude levels not corresponding to the boundaries of the Umkehr layers. This could enlarge differences between the ozone profiles because the ozone content in certain Umkehr layer derived from MLS data is additionally influenced by the ozone mixing ratio at levels below and above this layer. This is a result of the interpolation of the MLS mixing ratio to the Umkehr layer boundaries.

Table 1: Standard Umkehr layers and their typical altitude range

Layer	Layer Boundary (km)	Pressure limits (hPa)
0	0 – 5	1013 – 506.5
1	5 – 10	506.5 – 253.25
2	10 – 15	253.25 – 126.63
3	15 – 20	126.63 – 63.31
4	20 – 25	63.31 – 31.66
5	25 – 30	31.66 – 15.83
6	30 – 35	15.83 – 7.91
7	35 – 40	7.91 – 3.96
8	40 – 45	3.96 – 1.98
9	45 – 50	1.98 – 0.99
10	50 – 55	0.99 – 0.49
11	55 – 60	0.49 – 0.25
12	60 – 65	0.25 – 0.12
13	65 – 70	0.12 – 0.06
14	75 – 80	0.06 – 0.03
15	80 – top of the atmosphere	0.03 – 0

Prior to the ozone profile retrieval, Umkehr N-values are normalized to the measurement taken at the smallest of the nominal SZAs (typically at 60° or 70°). This procedure makes the retrieval insensitive to calibration and solar flux uncertainties, and the technique can be described as “self-calibrating” (Petropavlovskikh et al., 2004). A full description of the

forward and the inverse models used in the retrievals, along with a sensitivity analysis of the parameters that have been incorporated into UMK04 can be found in Petropavlovskikh et al. (2004) and Petropavlovskikh et al. (2005).

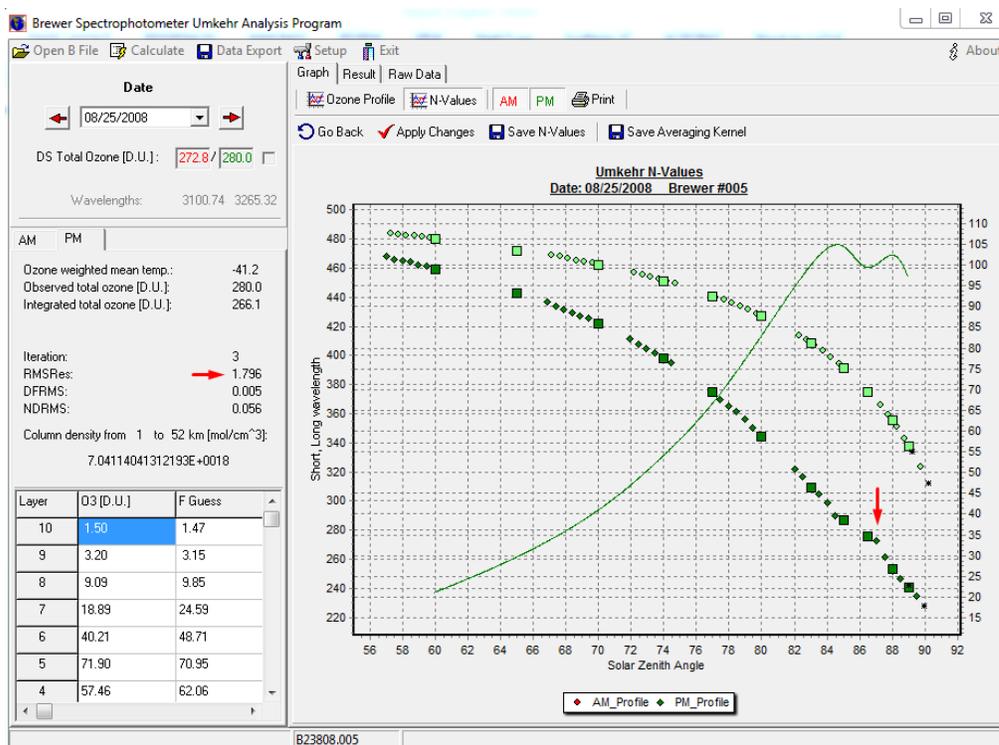
The analysis of the B005 Umkehr measurements was performed with the O3BUmkehr software, which has been developed by M. Stanek (based on the UMK04 algorithm) and is available online at <http://o3soft.eu/>. Details for the software and its set up can be found in the “readme” file that is included in the zip archive of the software. An ozone profile retrieval is successful when less than 3 iterations are required and the root mean square of the residuals is less than 1. The retrieval of the profiles can be done either in batch mode or manually; then is required the inspection of the N-values and the rejection of the bad points (e.g., those influenced by clouds) by the operator. The latter is time consuming but necessary for assuring the best quality of the retrieved profiles. In the batch mode, an essential step is the instrument’s operator to define a quality filter to remove the “bad” (cloudy) data. For the determination of the quality filter the ratio between two consecutive Umkehr measurements at 320 nm channel is used. The way to be achieved is described in the “readme” file of M. Stanek’s software, copied below:

“The way to define the best quality parameters for your own instrument is to pick a very clear day for measurements, and then open the “Raw data” tab at the top of the main window (you can see it after the set up window is complete and closed). The 320 nm ratio will be plotted as function of the SZA. You can then define the range of the natural variability in your measurement (“Range”) and the mean offset (“Offset”) parameters for you set up window. Click on the “Setup” tab at the top of the main window and change parameters, then close/save the setup. You might need to check measurements at several clear days to figure out the best adjustment for the instrument.”

Following the above instructions, an “error envelope” can be defined, so that all points that fall outside this envelope are masked as clouds, thus are not considered in the retrieval procedure. It should be noted that one should reconsider the calculation of this “envelope” when changes in the instrument characteristics are observed (e.g., after changing the dispersion settings during the calibration which subsequently affects the exact wavelength settings, after performing a high voltage adjustment and thus increase/decrease the instrument’s noise, etc.) that could affect the ratio at the 320 nm channel (possibly changes in the range and/or the offset of the parameters).

For B005 there were defined six periods with changes in the range and offset parameters (mostly after instrument calibration). However, it was decided the retrieval to be done manually, after visual inspection of the N-values and removal of the “bad” points. That was deemed necessary in order to achieve better quality of the retrievals and to further investigate the cases of some “offset” points in the “short” wavelength. These points were mostly observed in the SZA range 87 – 90°, resulting in many cases in a “noisy” (unsmooth) Umkehr curve and “bad” retrieval statistics. Since these noisy data were systematically

observed, it was attempted to be investigated whether their origin was instrumental or they may contain some information about changes in the vertical ozone distribution that may have happened during the sunset (i.e. collapse of the boundary layer or diurnal changes of the ozone in the upper stratosphere). It was examined if they are related with any wavelength shift, but nothing suspicious were found in the hg records. Moreover, the Umkehr measurements of Brewer 141, operating in Boulder, were inspected for any similar pattern in the “short” wavelength N-values, but it was not observed something similar. Figure 1 shows two characteristic cases of these “noisy” data points at SZA of $\sim 87^\circ$. The points marked with “x” are identified as affected by clouds, from the quality filter that was applied. Very strict limits for the range of the 320 nm ratio were set to allow only the instrumental noise (see Figure 2).



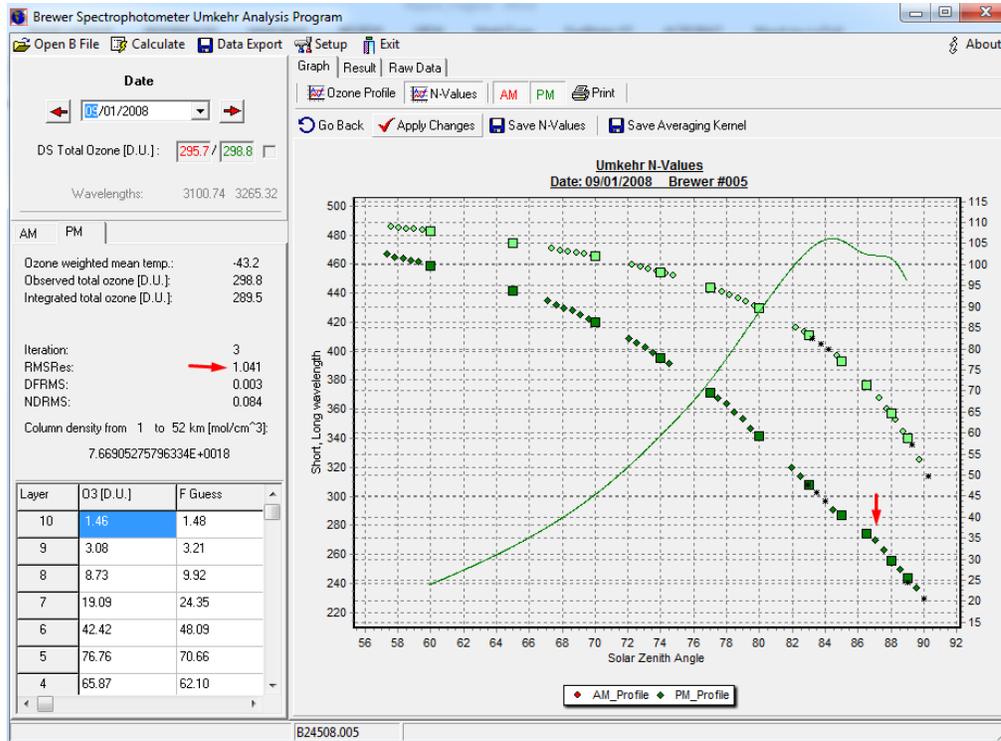
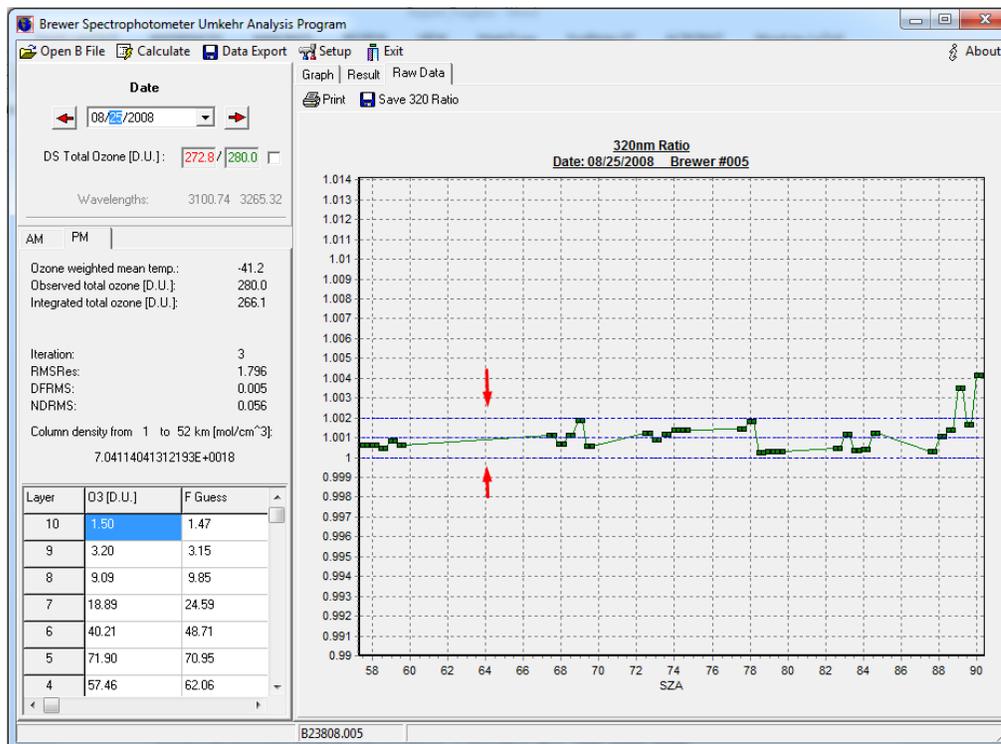


Figure 1: Characteristic examples of offset points in the short wavelength, resulting in a bad shape Umkehr curve and high root mean square error of the residuals. The upper panel is for 25/08/2008 and the lower for 01/09/2008.



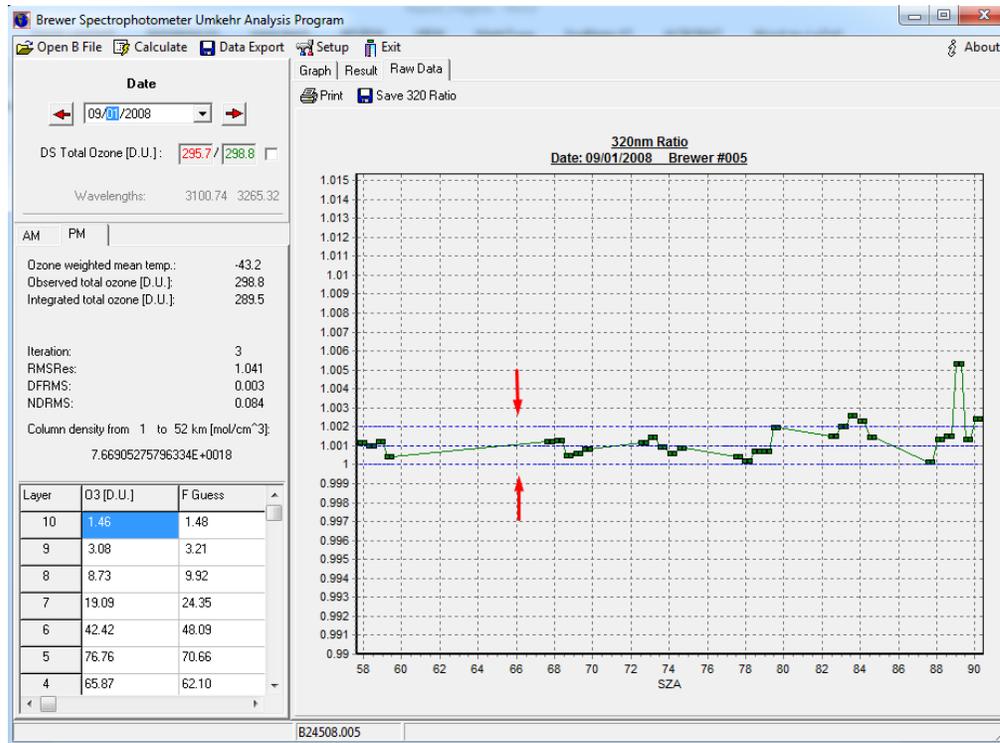


Figure 2: The ratio of the measurements at the 320 nm channel as a function of the SZA for the days presented in Figure 1. The blue lines indicate the “envelope” outside which the observations are flagged as “bad” (cloudy) points. For these examples, the limit of the range was set to 0.001 and the offset to 1.001.

Removing those points results in smoother Umkehr curve and better RMS, in the accepted limits for a successful retrieval (see Figure 3). This indicates that the retrieval algorithm is very sensitive to the N-values that are used for the interpolation at the nominal SZAs.

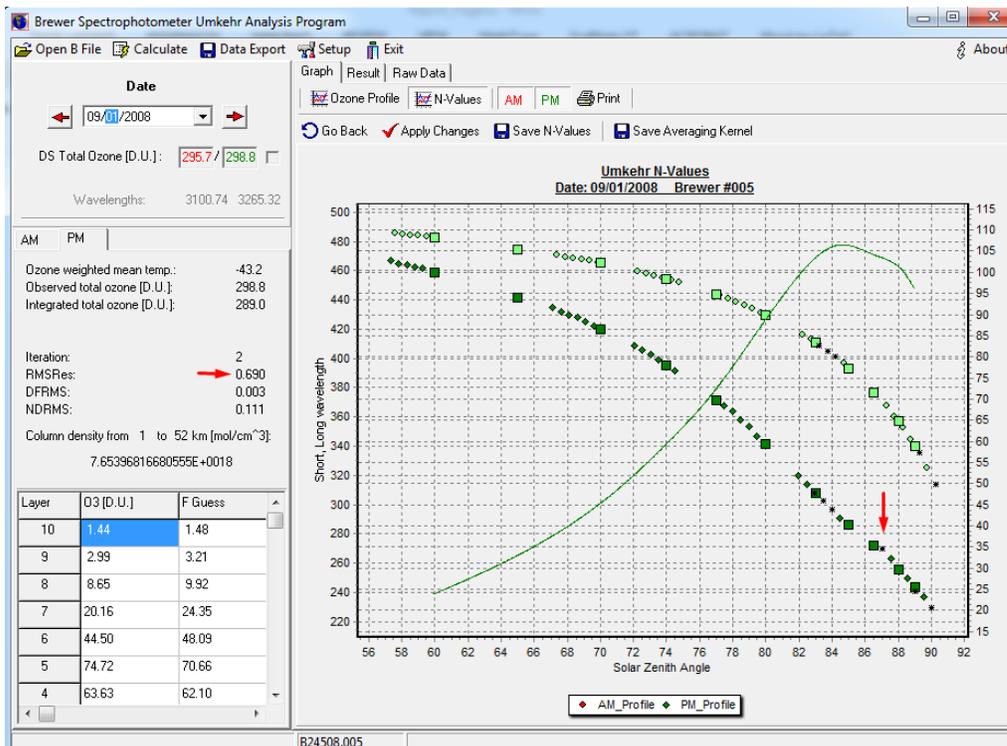
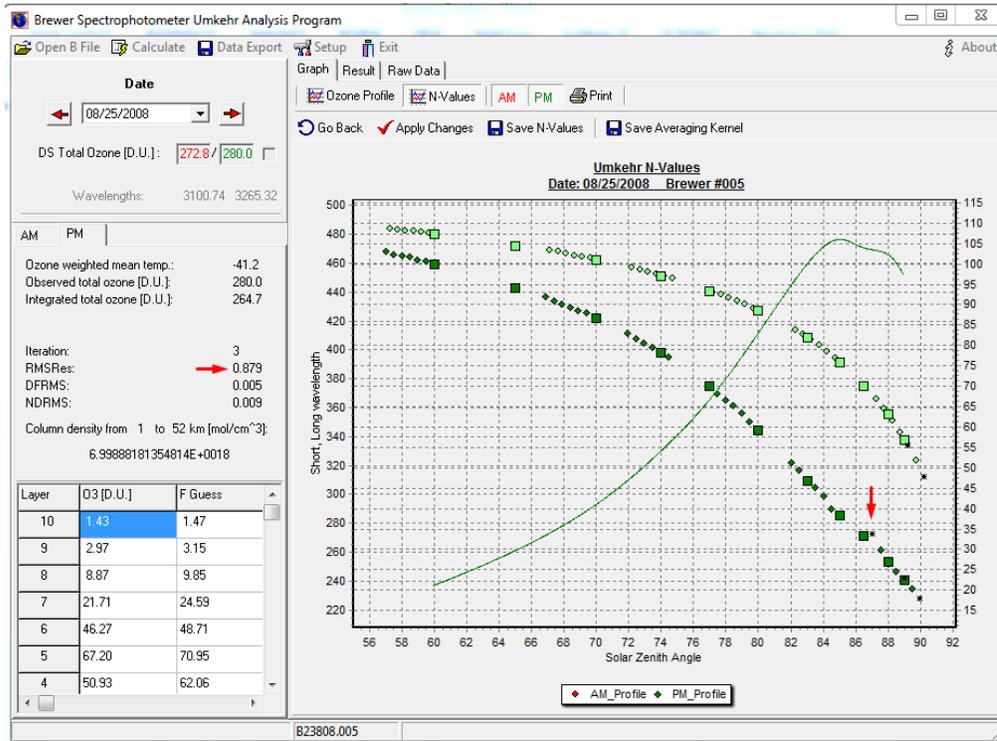


Figure 3: Same as Figure 1 after removing the offset points at SZA of ~87°. Note the improvement in the shape of the Umkehr curve and in the value of the RMSRes.

Figure 4 shows the retrieved ozone profile for 01/09/2008 when the offset point at ~87° SZA is included and excluded from the analysis. Moreover is plotted the interpolated profile from the MLS satellite overpass for the same day, which is assumed to be the true profile. It

is obvious that the removal of the point results in better agreement with the MLS profile and thus all these “offset” points have been removed prior to the retrieval.

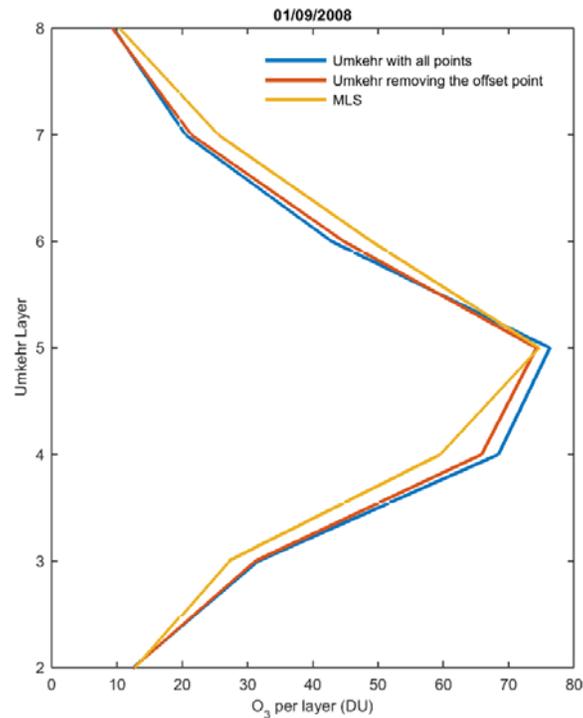


Figure 4: Retrieved ozone profile for 01/09/2008 from the Umkehr measurements when the offset point at $\sim 87^\circ$ SZA (see Figure 1, lower panel) is included in the analysis (blue line) and when it is removed (red line). The orange line shows the MLS profile interpolated at the Umkehr layers for the specific date.

Nevertheless, the issue of the offset points has not been addressed adequately and for this reason we initiated regular Umkehr measurements, since November 2015, with the collocated double monochromator Brewer with serial number 086 (B086). Surprisingly, these kind of “offset” points have been observed in both instruments, an indication that supports the assumption that are not of instrumental origin. Such an example is given below, in Figure 5, where is evident that the point at SZA $\sim 89.2^\circ$ deviates significantly from the other points in both Brewers (B005 and B086).

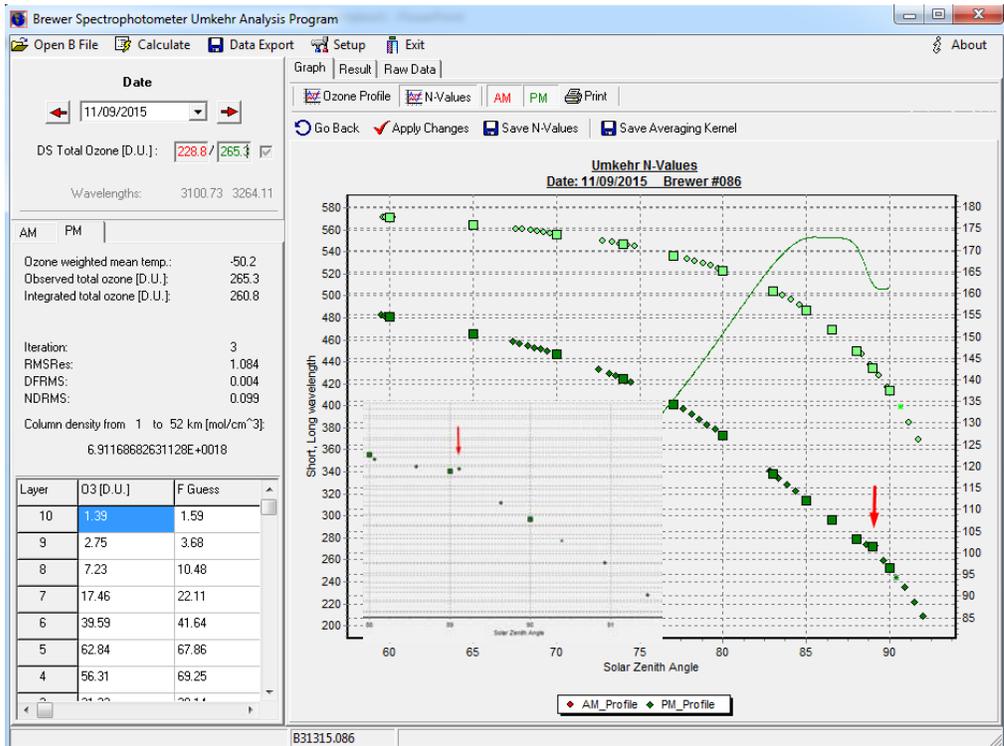
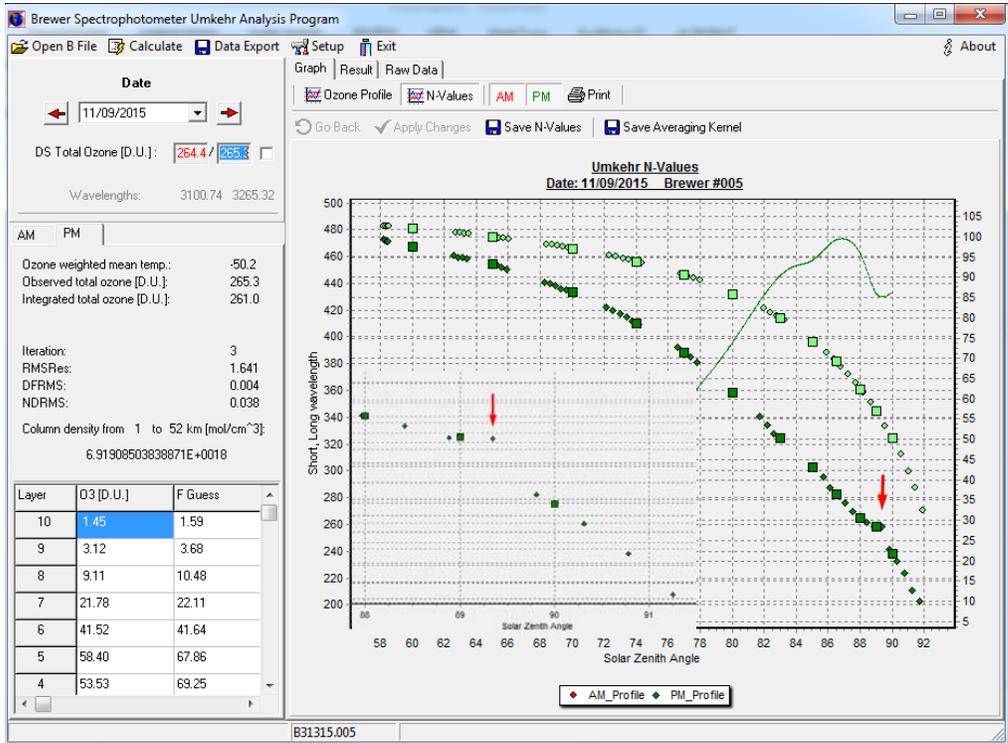


Figure 5: The N-values as function of the SZA for 09/11/2015, for Brewers B005 (upper panel) and B086 (lower panel). The red arrow indicates the points that deviate from the normal pattern. It is annotated inside the figures a zoom of the area where the offset points have been observed.

Furthermore, it was discovered that when the normalization of the N-values was done in the 60° the difference between the integrated ozone from the Umkehr retrieval and the observed total ozone column (from the DS measurements) was quite big, in most cases. When the normalization was done in 70° or 74° the agreement was much better, thus it was decided that the smaller SZA (where the normalization of the N-values is performed) will be 70° or 74°. Such an example is given in the Figure 6, below.

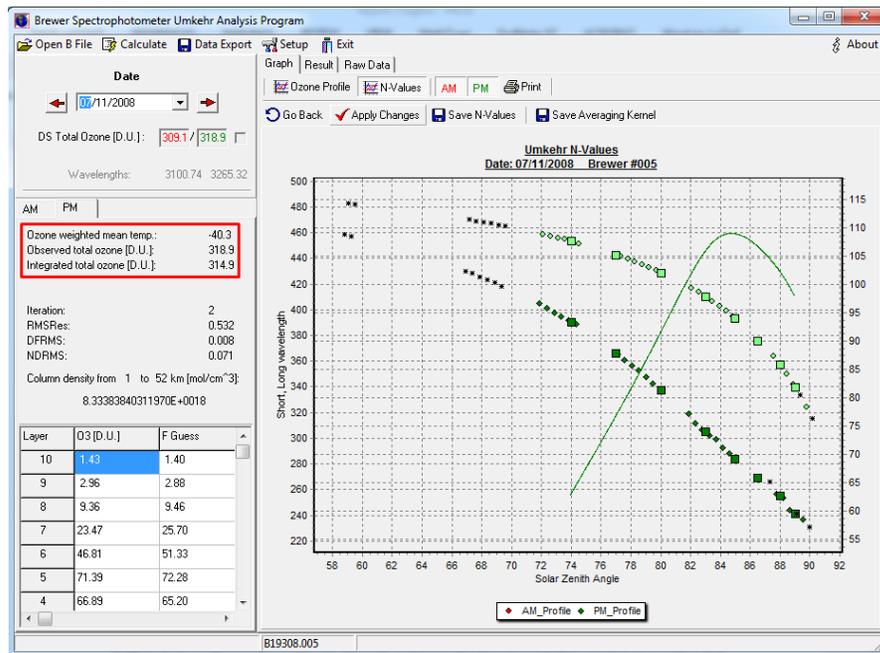
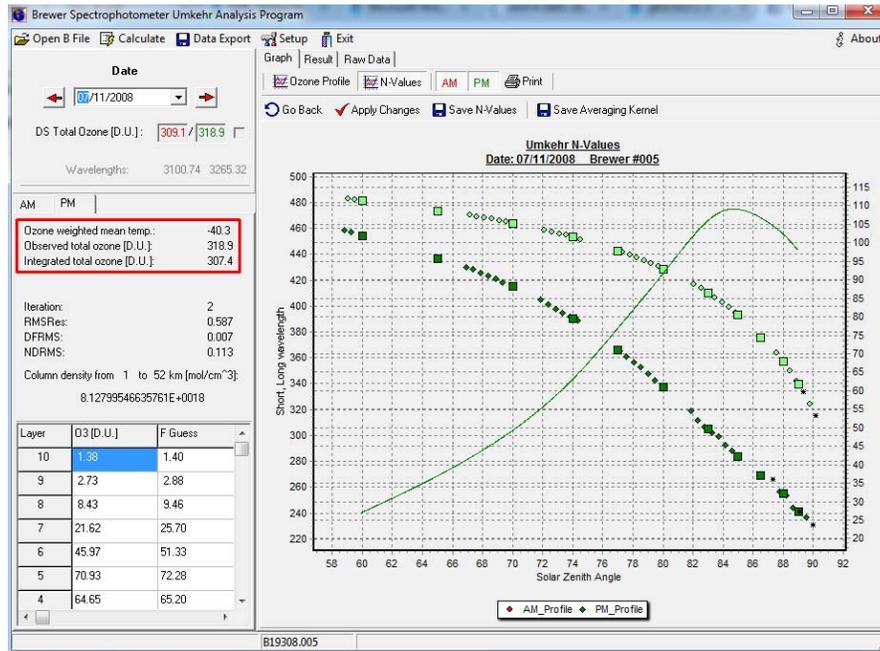


Figure 6: Example of a “good” Umkehr retrieval. Note the big difference between the observed total ozone column and the integrated one when the normalization of the N-values is done at 60° (upper panel) and the better agreement of the two quantities when the normalization is done at 74° (lower panel).

3. Effect of stray light and temperature on Umkehr retrievals

In addition to the thorough investigation of the retrieval procedure, during the STSM it was examined the effects of the stray light and the temperature dependence of the ozone absorption coefficients in the retrieved profiles. The results, which are presented below are in agreement with those of an earlier study of Petropavloskikh et al. (2011), confirming that the out-of-band stray light has a major impact on the retrievals, while the effect of the temperature dependence of the cross-sections is minor. In order to test the Umkehr retrievals for the effects of stray light and changes in the true ozone effective temperature, during the STSM the O3BUmkehr software of M. Stanek has been updated several times and the latest available version is now 3.2. At the beginning of the STSM, the version number was 2.7 and did not include a stray light correction or the option of reading the ozone effective temperature from an external file. For this purpose, the measured slit function (Figure 7) of B005 was modelled to include the effect of the stray light (wings) and was incorporated in the latest version of the O3BUmkehr software.

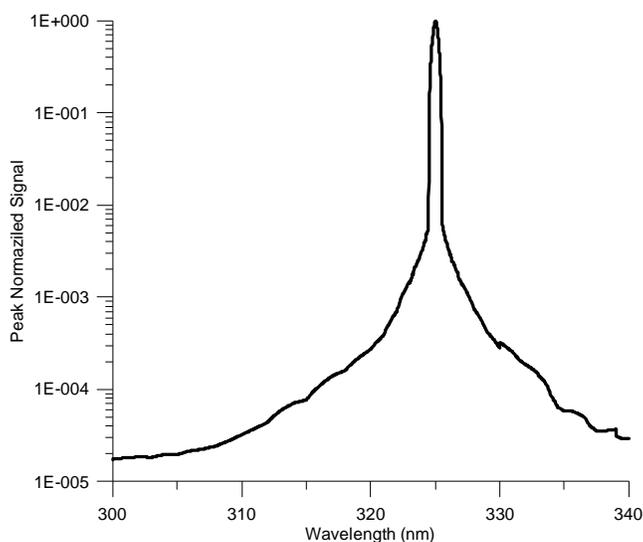


Figure 7: Slit scattering function of Brewer 005, measured through slit #1 during the X Intercomparison RBCCE Campaign with a HeCd laser.

A drawback of the O3BUmkehr software when used for testing the Umkehr algorithm is that it does not keep a record of the “bad” points that are removed by the user. For this reason, analysis that is done manually is more time consuming, since the user has to remove the bad points each time the software is run (e.g., for testing different versions of the software). This could be possibly solved in a future release, by giving the user the option to choose whether the software reads the Umkehr measurements from the B-files (like it is now) or to read the N-values from a file where they have been saved by the user previously. For this reason and because the analysis was performed in a manual way and many different

versions of the software were tested, the Umkehr record of Thessaloniki was only partially analyzed during the STSM. Nevertheless, the analysis is going on after the return in the home institution.

In order to investigate the effect of the stray light in the ozone profiles, the Umkehr retrievals were calculated for a period of one year (2008), first without any stray light correction (based on O3BUmkehr v.2.7) and then again by applying a stray light correction (with the O3BUmkehr v.3.2). These results were subsequently compared with the overpass MLS profiles. Moreover, an extra step of analysis included the retrieval of the Umkehr profiles using the ozone effective temperature as calculated from the overpass MLS ozone and temperature profiles instead of the climatological ones used in the retrieval algorithm. The effective ozone temperature is defined as the integral over altitude of the ozone profile-weighted temperature, normalized with the ozone profile integral. As can be seen in Figure 8, the “actual” ozone weighted temperature (MLS) for Thessaloniki is on average 5 - 10 °C lower than the one calculated from the climatological temperature profiles.

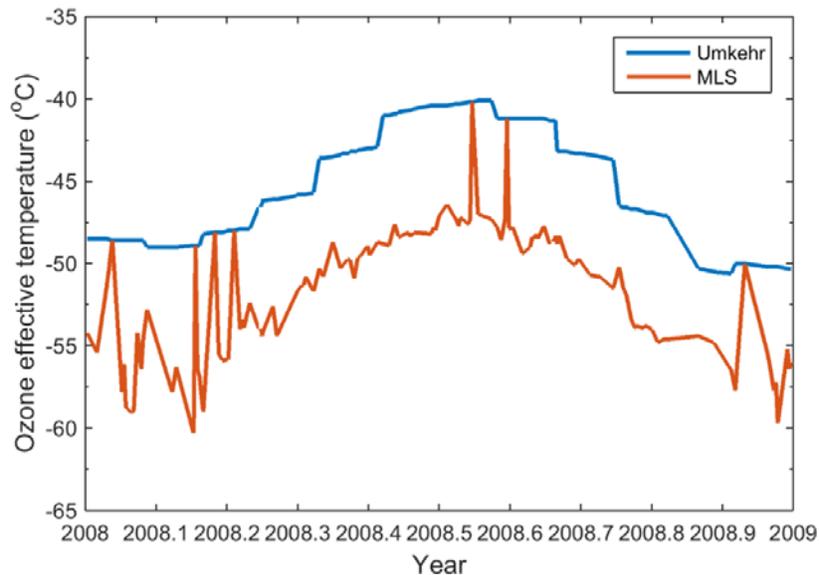
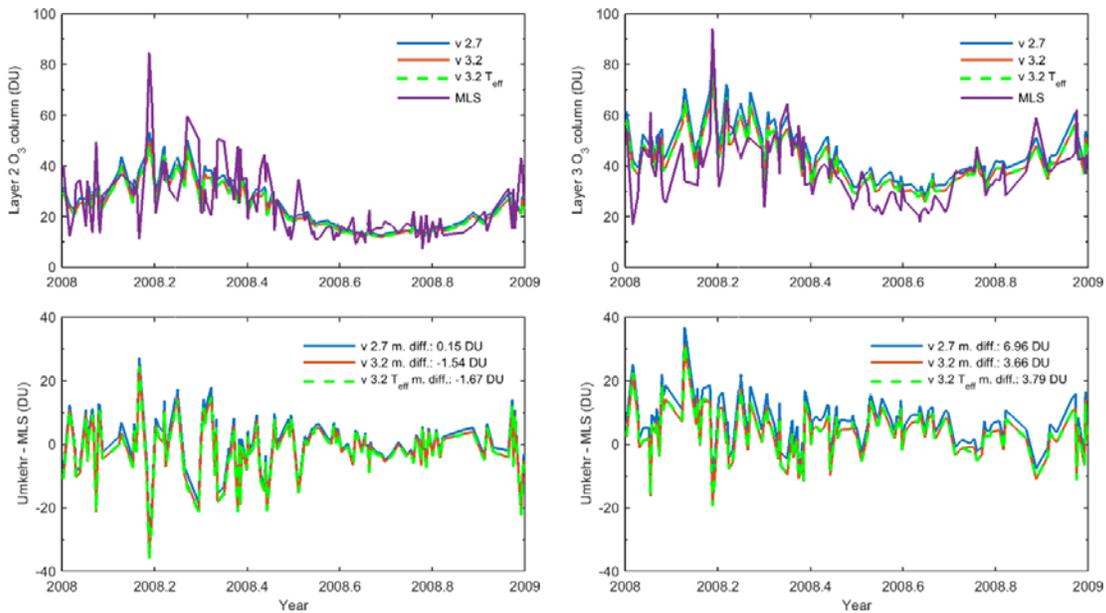


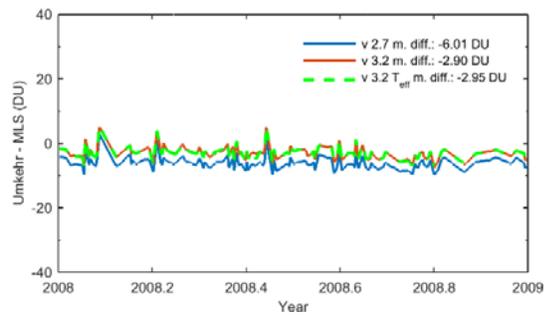
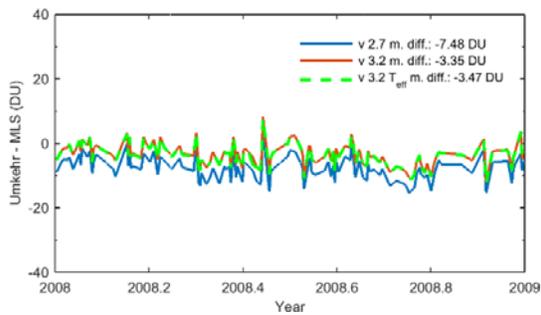
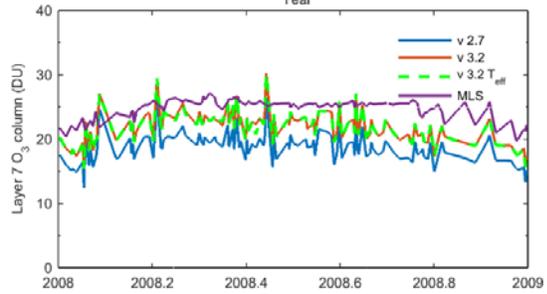
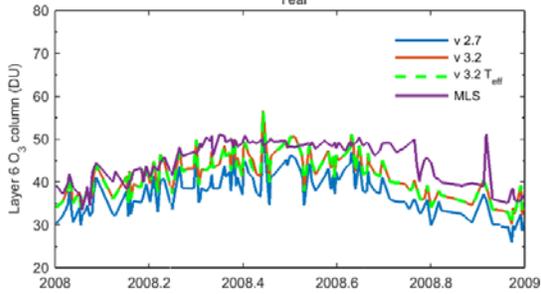
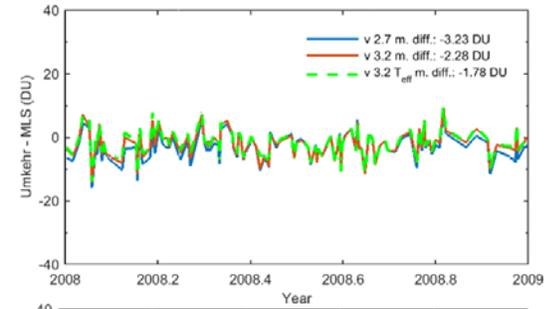
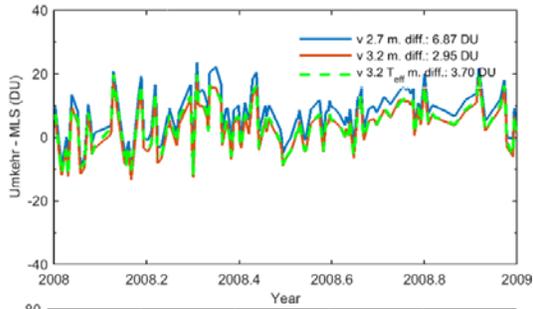
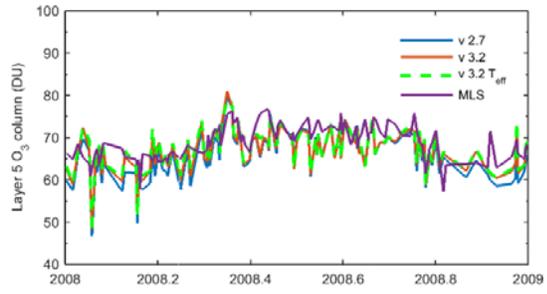
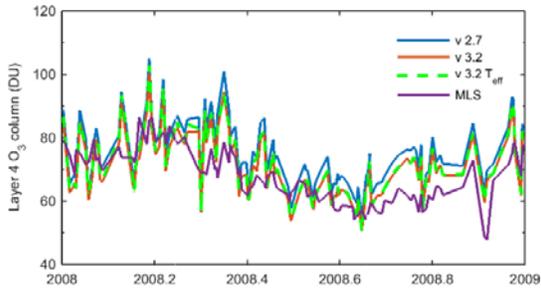
Figure 8: Yearly evolution of the ozone effective temperature derived from the Umkehr ozone profiles and climatological temperature profiles (blue) and calculated from the MLS ozone and temperature profiles (red).

The time series of the retrieved ozone column in each Umkehr layer, based on the different versions of the software, as well as the difference from the MLS column interpolated in the respective Umkehr layer, are shown in Figure 9. Although there is a mean bias in the retrieved Umkehr profiles in all layers, this bias does not show any seasonal dependence. The differences from the MLS show the highest variability in the lower stratosphere (layers 2 and 3, see also Figure 10). Possibly, these discrepancies could be improved if the comparison was performed by combining layers 2 and 3 in a single layer (2+3), as suggested by Petropavlovskikh et al. (2004; 2005), since they don't contain independent information. In addition, a further improvement should include the smoothing of the MLS profiles with the average kernels from the Umkehr algorithm. Moreover, it is evident that the stray light

correction improves the agreement with the MLS profiles. This is more obvious in Figure 10 where the initial difference of about $\pm 20\%$ between the derived profiles and the MLS profiles, drops to less than $\pm 10\%$ when the stray light correction is applied. While the stray light correction effect is evident in all layers, it is less pronounced in layers 4 and 5 but shows a seasonal dependence that coincides with the layer where the bulk of ozone is present. For winter months the stray light correction has smaller effect for layer 4 at $\sim 20 - 25$ km, while for summer months it is smaller for layer 5 at $\sim 25 - 30$ km). These indications need further evaluation. For most months the bias in the lower layers (2, 3 and 4, from ~ 10 to ~ 25 km) is negative, while in the upper layers (5, 6 and 7, ~ 25 to ~ 40 km) is positive. The ozone columns in layers 5 and 6 show better agreement with the MLS profiles after the stray light correction is applied.

The correction for the temperature dependence of the ozone absorption coefficients with the “actual” effective temperature instead of the one based on the climatological profiles has a minor effect, as it appears from the comparisons of Figures 9 and 10. Of course, these results are valid for the Bass and Paur ozone cross sections and such an analysis could be performed in the future to examine the effect in different spectroscopic datasets.





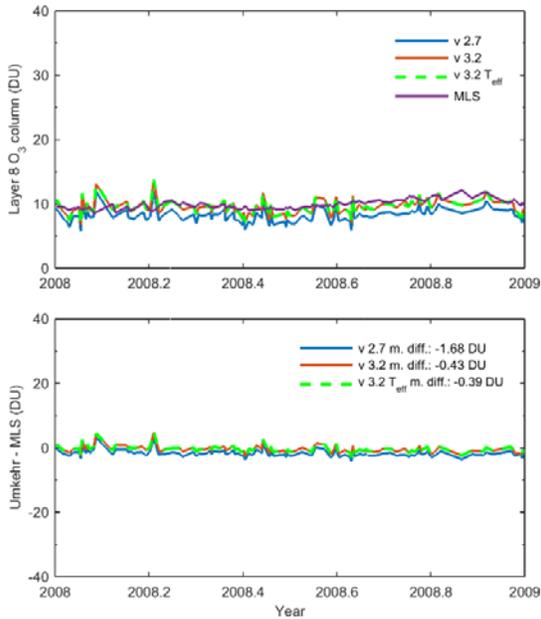
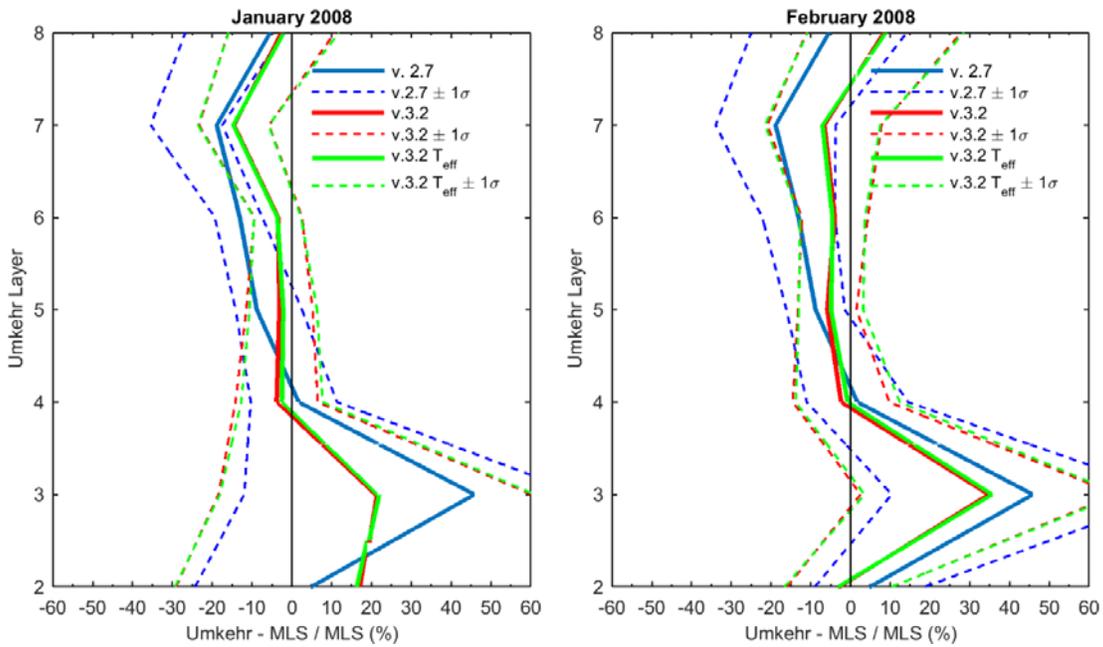
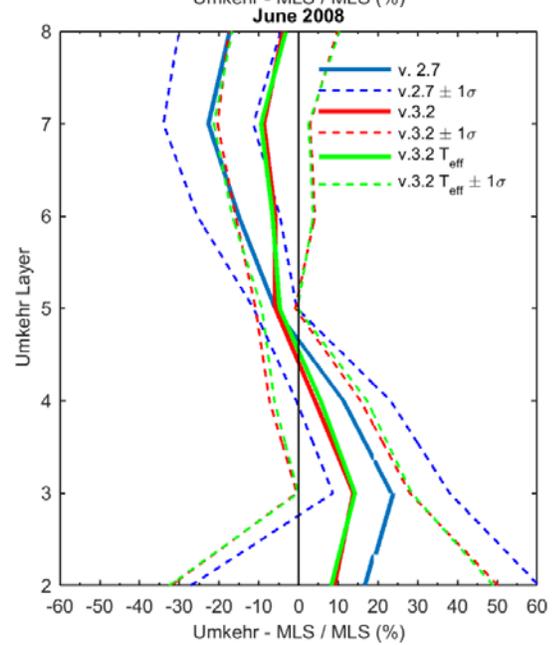
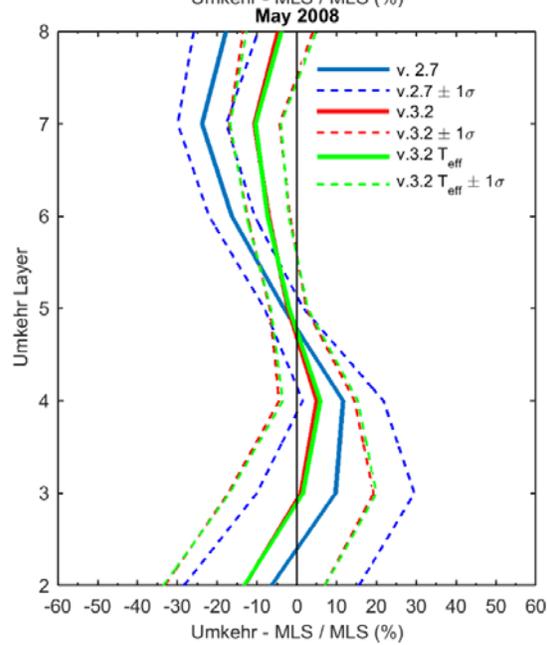
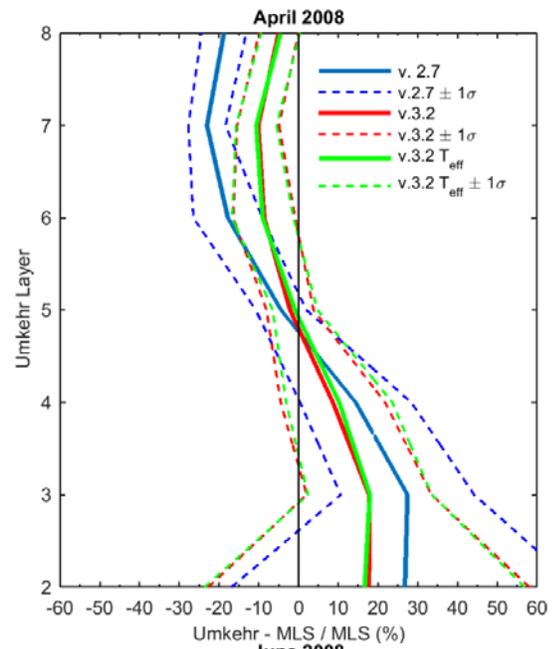
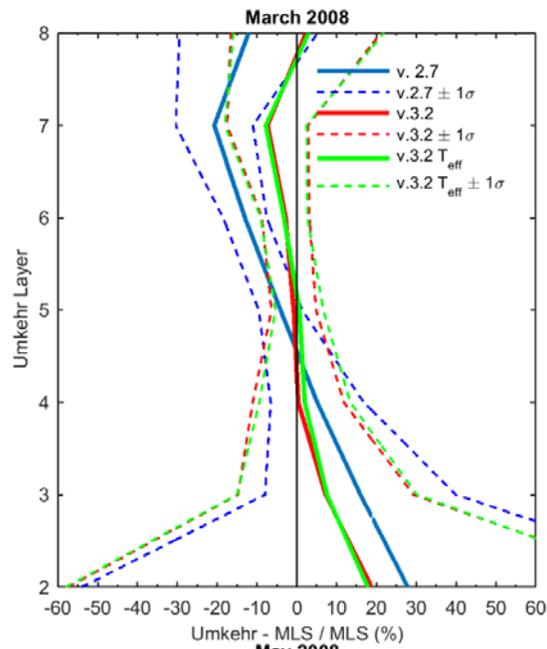
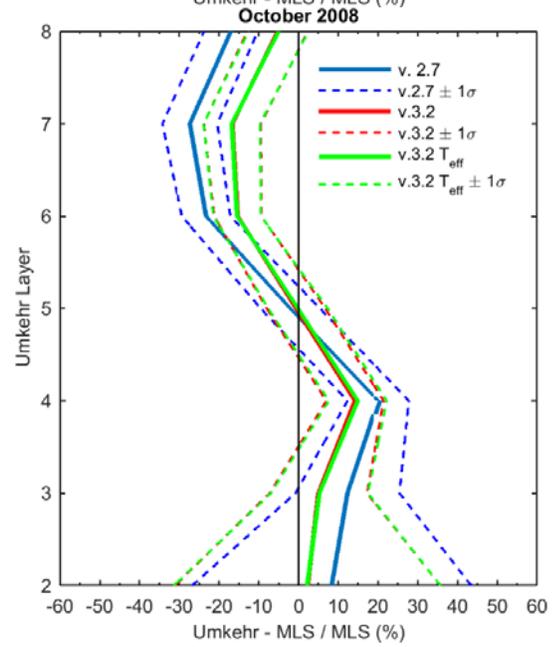
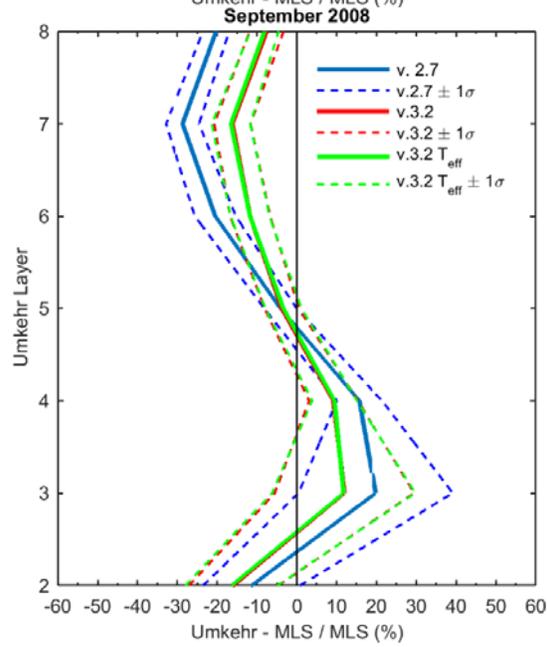
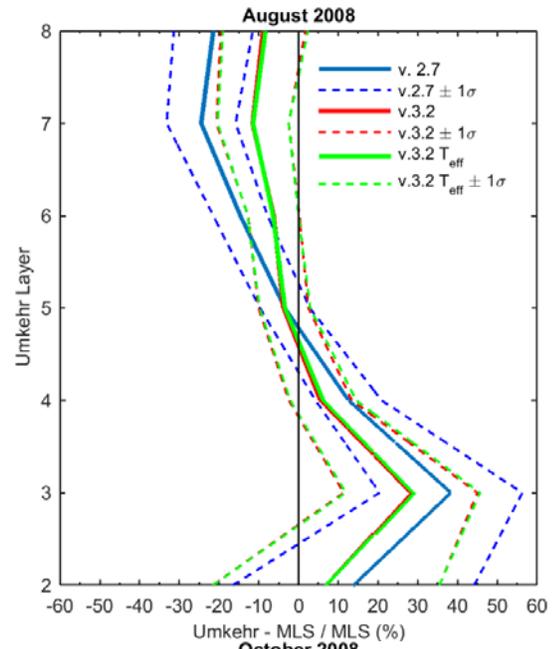
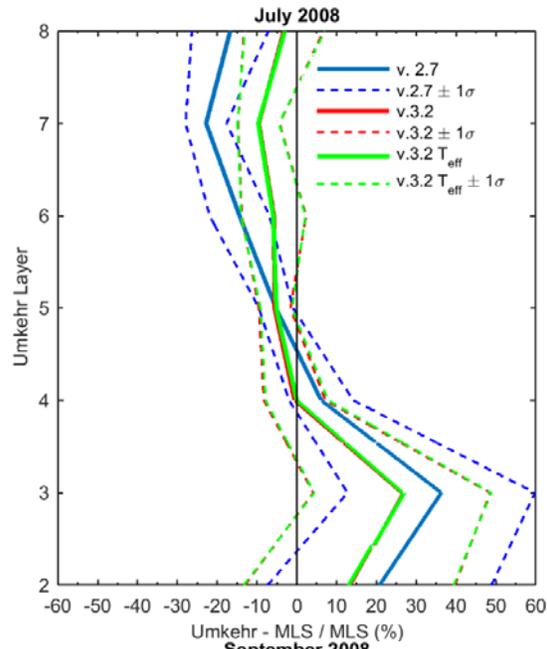


Figure 9: Time series of the column ozone in different Umkehr layers as derived from different versions of the retrieval algorithm and the MLS satellite for each Umkehr layer. The respective absolute difference of the Umkehr derived ozone from the MLS is shown underneath each panel. Lines in different colors correspond to Umkehr retrievals with (blue) and without (red) correction for stray light, and additionally with correction for the effective ozone temperature (green).







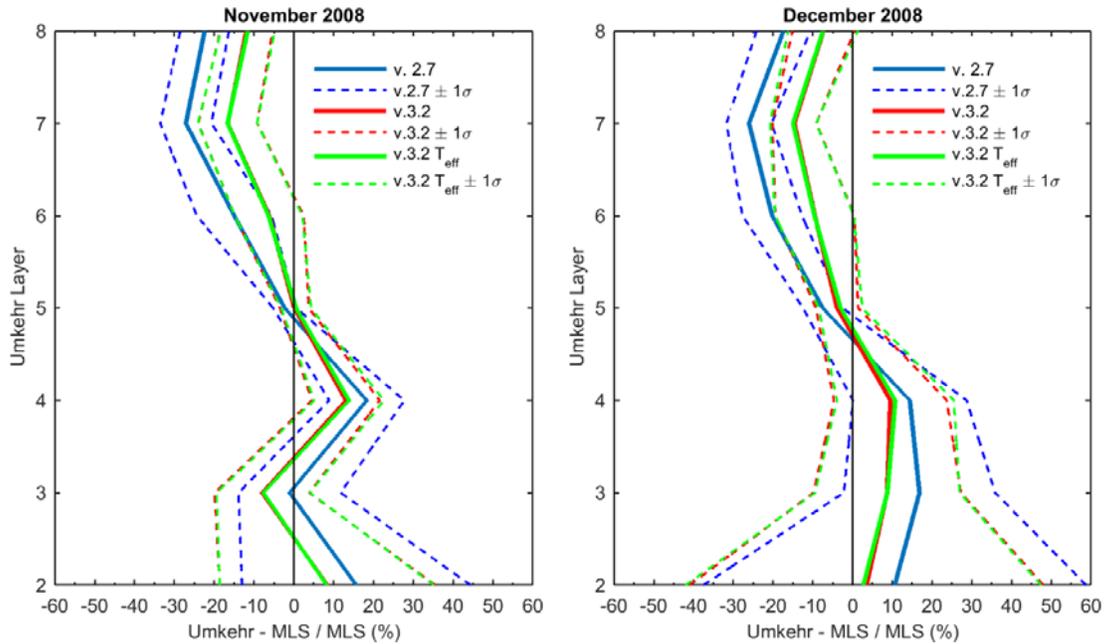


Figure 10: Monthly mean difference (%) between the derived ozone profiles and the MLS profiles with (blue) and without (red) including correction for the stray light, and additionally with correction for the effective ozone temperature (green). The dashed lines show the 1σ limits.

4. Main conclusions

During the specific STSM, the Umkehr record of a single-monochromator Brewer spectrophotometer operating in Thessaloniki was investigated. It was found that at specific SZAs during evening, when the sun is close to the horizon there are some “offset” points at the short Umkehr wavelength. An attempt was made to identify if such points are related to any instrumental issue, but this has not been proven. In addition such offset points were observed also in a collocated double monochromator Brewer, when these two instruments were operating in parallel. The origin of those points is still under investigation.

Furthermore, it was examined the impact of the stray light and the temperature dependence of the ozone absorption coefficients in the retrieved profiles. For this reason, the O3BUmkehr software was updated by Martin Stanek to include the stray light correction. The analysis confirmed earlier studies (Petropavlovskikh et al., 2011) showing that the effect of stray light is very high, while the ozone effective temperature that is used to correct the temperature dependence of the ozone absorption coefficients has a minor effect. The analysis will be continued at the home institute until the entire record of Umkehr measurements is processed.

The analysis of the Umkehr measurements of all instruments that participate in the Eubrewnet could be performed centrally, in batch mode, in the database. This requires that the station operators provide essential information for the software set up for each station and, especially, after determining the filters for the characterization of the “bad” (cloud affected) data. Nevertheless, to access the better quality Umkehr retrieval the manual

inspection of the retrievals is required, since 1-2 offset points may affect seriously the retrieval.

5. Future collaboration with the host institution

The collaboration with the host institution and particularly with Dr. Irina Petropavlovskikh will continue, as there is an ongoing process for the analysis of the complete Umkehr record of B005.

6. Foreseen publications to result from the STSM

The results of the STSM will be presented in the forthcoming COST1207 meeting in Azores. In addition, results of this STSM will be presented in the upcoming Quadrennial Ozone Symposium (QOS 2016) 4-9 September in Edinburgh, UK.

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