

Response to comments of Referee #1

We thank the referee for his or her thoughtful comments, which we have addressed as follows:

Referee comments

I would like to see a bit more discussion on the temporal and spatial availability of the measurements. Consistent (Zenith-sky and Global-Umkehr) naming conventions would be nice to avoid any confusion. I would also like to see some discussion of the local times that MLS passes over summit and seasonal change in the averaging kernel (if any).

Response

The new version of the manuscript now uses the terms “standard zenith-sky Umkehr method” and “Global-Umkehr method” consistently. Other issues noted in this general comment are discussed below.

Referee comment

Page 1, line 26: I would like to see a short explanation of what this measurement (the direct sun plus upper hemisphere) is typically used for.

Response:

The following was added to the manuscript:

“Such measurements were started by several groups in the early 1990s to monitor changes in UV radiation at the Earth’s surface. These activities were motivated by concerns that decreases in atmospheric ozone concentrations, which were caused by ozone depleting substances released by man into the atmosphere, could lead to increases in UV radiation with detrimental effects on human health, and terrestrial and aquatic ecosystems (e.g., Bais et al., 2015).”

The following reference was added:

Bais, A. F., McKenzie, R. L., Bernhard, G., Aucamp, P. J., Ilyas, M., Madronich, S., and Tourpali K.: Ozone depletion and climate change: impacts on UV radiation, *Photochem. Photobiol. Sci.*, 14(1), 19-52, 2015.

Referee comments

Considering that this paper may lead to retrieval of more historical ozone information in addition to what is already available (ozonesondes, Dobson Umkehr, etc.). I would like to see a bit more discussion on the temporal and spatial availability of the measurements.

and

Page 2, line 2 and line 21 and Section 2.2: You mention that there are other sites that have these UV detectors. It would be nice to have some general information about how many potential sites there are, their temporal measurement range, are there any Southern Hemisphere sites, and there many locations where there are not any Umkehr measurements, etc. It is briefly mentioned in the conclusions that there are many sites with time series of greater than 25 years, but this is not mentioned anywhere else.

Response

The majority of instruments that provide global spectral UV irradiance measurements suitable for the Global-Umkehr method are part of the UV monitoring networks mentioned in the introduction. While it is beyond the scope of this manuscript to assess the suitability of each of these systems, we estimate that about 25 instruments meet the accuracy requirements for the Global-Umkehr method and could potentially be utilized. The following was added to the manuscript:

“We estimate that about 25 spectroradiometers that are part of the various UV monitoring networks mentioned earlier provide data of sufficient quality for the Global-Umkehr method. Some of these instruments were established in the early 1990s at locations around the globe, including the Arctic, North America, Hawaii, Europe, New Zealand, Australia, and Antarctica.”

We also added a link to the European UV Database, which also includes quality-controlled spectral UV measurements that are potentially suitable for the Global-Umkehr method.

Referee comment

Page 3, line 4: Why not use the more recent ozone cross section studies?
<https://www.atmos-meas-tech.net/7/609/2014/>

Response

We note that Referee 2 had a similar comment.

We are aware that more accurate ozone absorption cross sections than those published by Bass and Paur (1985) are now available and recommended. Nonetheless, we decided to use the Bass and Paur (1985) data because OMI total ozone data are based on B&P. Using a different cross section would have complicated the validation of our results with OMI data. We added the following to the manuscript:

“While more accurate ozone absorption cross sections are now available (Gorshchev et al., 2014; Orphal et al., 2016), we used Bass and Paur (1985) data to facilitate validation with OMI total ozone column measurements, which are also based on Bass and Paur (1985).”

The following references were added:

- Gorshchev V., Serdyuchenko A., Weber M., Chegade W., and Burrows J.P.: High spectral resolution ozone absorption cross-sections–Part 1: Measurements, data analysis and comparison with previous measurements around 293K, *Atmos. Meas. Tech.*, 7, 609-624, doi:10.5194/amt-7-609-2014, 2014.
- Orphal J., Staehelin J., Tamminen J., Braathen G., De Backer M. R., Bais A., Balis D., Barbe A., Bhartia P. K., Birk M., and Burkholder J. B.: Absorption cross-sections of ozone in the ultraviolet and visible spectral regions: Status report 2015, *J. Mol. Spectrosc.*, 327, 105-121, doi:10.1016/j.jms.2016.07.007, 2016.
-

Referee comment

Page 4, line 19: You mention on line 14 that a σ_a value of .1 is small and therefore very sensitive to the a priori. However, you go on to say on line 19 that $\sim .1$ is the standard deviation of the MLS profiles. I feel this needs clarification as you mention that σ_a is the anticipated variability (standard deviation) and therefore using a value higher than .1 (for example .4) means you are expecting a larger variability in the retrieval.

Response

As discussed in great detail in the manuscript, σ_a is an important parameter to optimize the solution. At the onset of the study, the optimal value for σ_a was not known, although a value of 0.1 is supported by the standard deviation of the MLS profiles. We therefore performed calculations with two settings, 0.1 and 0.4. We feel that the pros and cons of using either 0.1 or 0.4 are discussed in sufficient detail (in particular in the “Discussion” section), and think that lengthening the discussion further is not necessary. No change to manuscript.

Referee comments

I would also like to see some discussion of the local times that MLS passes over Summit.

and

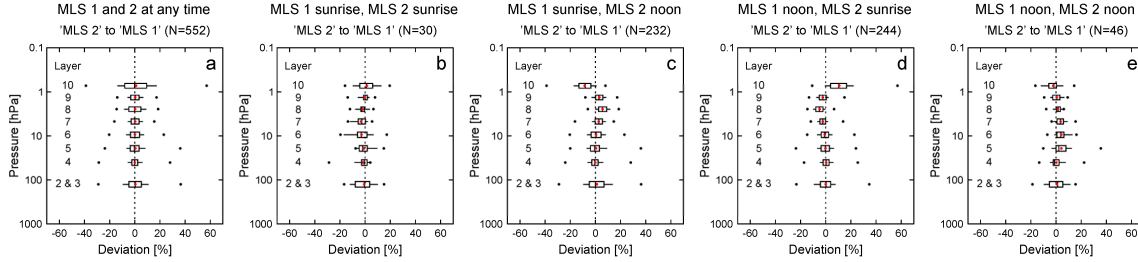
Page 10, line 1: What two times of day are MLS measurements taken at the latitude of Summit? Are there any inconsistencies here, diurnal effects, polarisation?, etc?

Response

MLS data that we have downloaded from http://avdc.gsfc.nasa.gov/pub/data/satellite/Aura/MLS/V04/L2GPOVP_Prof/O3/Summit/ are measured either between 5:28 and 6:26 UTC or between 14:11 and 15:10 UTC. These period concur roughly with sunrise and local solar noon at Summit, respectively. There is only one file per day provided by the Aura Validation Data Center, either from the earlier or later time range. There is no obvious difference in the timing between spring and fall. Of note, spectra used for the Global-Umkehr method were measured between 15:00 and 20:00 UTC.

MLS 1 data (i.e., MLS measurements taken before the period of Umkehr observations) and MLS 2 data (i.e., MLS data taken after this period) discussed in the manuscript were not filtered for time-of-day. The difference of MLS 2 and MLS 1 data showed virtually no bias (e.g., Figure 51 of manuscript), but there is some variability, which we attributed to several potential causes, including a real change in the ozone profile from one day to the next, variations in the horizontal distance between the locations of Summit and the MLS pixel from one day to the next, and random errors in MLS data.

Prompted by the referee’s comment, we have now filtered and analyzed MLS data also by time-of-day. Specifically, we have calculated the difference in MLS profiles from one day to the next in dependence of the time-of-day when MLS 1 and MLS 2 measurements take place. Results are shown in the figure below.



Panel a is identical with Figure 5l of the manuscript and shows statistics of the difference of MLS 2 and MLS 1 data irrespective of time-of-day. It can be seen that there is virtually no bias between the two datasets.

Panel b is based on a subset of these data where both MLS 1 and MLS 2 are from the sunrise period. For Panel c, MLS 1 data were from the sunrise and MLS 2 data from the noon period. For Panel d, MLS 1 data were from the noon period and MLS 2 data from the sunrise period. Finally, Panel e shows the difference where both MLS 1 and MLS 2 data were filtered for the noon period.

A comparison of Panels b – e indicates that there is a bias between MLS 1 and MLS 2 measurements depending on whether data from the sunrise or noon periods are used. We did not consider this bias when submitting the original version of the manuscript. This bias agrees qualitatively with the difference of day – night profiles measured independently by several instruments at Mauna Loa, Hawaii (Parrish et al., 2014), and Bern, Switzerland (Studer et al., 2014). Specifically, data from Mauna Loa indicate 2-3% *higher* ozone concentrations during the day for pressure levels between 2 and 10 hPa (~ Umkehr layers 6 – 8) and 5-10% *lower* concentrations during the day for pressure levels between 0.5 and 1 hPa (Umkehr layer 10). At Bern, ozone concentrations between 3 and 10 hPa are highest in the afternoon, exceeding midnight concentrations by 3-5%. Above 2 hPa (~43 km), the pattern reverses with ozone concentrations being lower during the day than at night. Differences observed at Mauna Loa and Bern are by and large consistent with those shown in Panel c above and small differences between the three datasets may be explained by the different latitudes of Summit, Mauna Loa, and Bern.

As a result of these new findings, the manuscript was changed as follows:

- In Section 2, we added:

“MLS measurements at Summit take place either between 5:28 and 6:26 UTC (period close to sunrise) or between 14:11 and 15:10 UTC (period close to local solar noon). There is only one data file per day in the NASA archive.”
- The following was added to the Discussion:

“Further analysis revealed that the difference between the MLS 1 and MLS 2 datasets depends also on the time when the daily MLS observation takes place. For example, when MLS 2 data are from the observation period close to local solar noon (14:11 to 15:10 UTC) and MLS 1 data are measured close to sunrise (5:28 to 6:26 UTC), MLS 2 data for Layers 7 – 9 are biased high by 3-6% relative to the MLS 1 dataset, while MLS 2 data for Layer 10 are biased low by 8%. This time-of-day dependency and its variation with altitude is by and large consistent with diurnal variations of the ozone profile measured by various instruments at Mauna Loa, Hawaii (Parrish et al., 2014), and by a microwave radiometer at Bern, Switzerland (Studer et al., 2014). This suggests that the

time-of-day effect observed at Summit is caused by actual diurnal changes of the ozone profile rather than potential time-dependent systematic errors in the MLS dataset. ”

The following references were added:

- Parrish, A., Boyd, I. S., Nedoluha, G. E., Bhartia, P. K., Frith, S. M., Kramarova, N. A., Connor, B. J., Bodeker, G. E., Froidevaux, L., Shiotani, M., and Sakazaki, T.: Diurnal variations of stratospheric ozone measured by ground-based microwave remote sensing at the Mauna Loa NDACC site: measurement validation and GEOSCCM model comparison, *Atmos. Chem. Phys.*, 14(14), 7,255-7,272, doi:10.5194/acp-14-7255-2014, 2014.
- Studer, S., Hocke, K., Schanz, A., Schmidt, H., and Kämpfer, N.: A climatology of the diurnal variations in stratospheric and mesospheric ozone over Bern, Switzerland, *Atmos. Chem. Phys.*, 14(12), 5,905-5,919, doi:10.5194/acp-14-5905-2014, 2014.
-

Referee comment

Page 15, Fig. 4: It would be interesting to see if the change in season (thus, the vertical structure of the ozone profile) modifies the structure of the relative averaging kernels, especially, as fall and spring statistics are compared later on in Table 2.

Response

As stated in the text (e.g., P13, L18) the relative averaging kernels do not depend much on season. For the sake of simplicity, we therefore decided not to show the RAKs in Figure 3 and 4 at the time of the initial submission. Prompted by the referee’s comment, we have now added the RAKs to the two figures.

Referee comment

Page 15, Fig 4: Also, why are the $\sigma_a = .4$ plots not shown in this figure? It would be interesting to see if the inversion agrees well in this case when it has more freedom due to a larger a priori covariance. If you have the results, they could also just be mentioned in the text.

Response

The effect of changing σ_a from 0.1 to 0.4 is very similar for spring and fall profiles. For sake of brevity, we therefore did not include results for $\sigma_a = .4$, and still feel that this is the appropriate decision. This is also supported by the small difference in the statistics for spring and fall shown in Table 2.

We added the following to Sect. 3.1.3:

“The effect of changing σ_a from 0.4 to 0.1 are similar for spring and fall profiles and results for $\sigma_a = 0.4$ were therefore omitted in Fig. 4.”

Referee comment

At the beginning of the paper, you define Umkehr to refer to the standard zenith sky Umkehr technique and Global-Umkehr to refer to direct sun plus upper hemisphere. However, throughout the text and especially in the discussion you refer to Global-Umkehr as just Umkehr which is confusing. I suggest keeping the naming conventions consistent throughout the text.

Response

The naming convention was homogenized throughout the manuscript.

Technical corrections

Page 1, line 11: Substitute ultraviolet for UV.
Changed.

Page 1, line 18: The OMI acronym does not need to be included here as it is not repeated in the abstract. It is redefined in the main text.
“OMI” deleted.

Page 2, line 4: Double closed bracket.
The second bracket belongs to “(e.g.,” of the preceding line. No change.

Page 7, line 2: Is the AFGL acronym defined (Air Force Geophysics Laboratory)?
“Air Force Geophysics Laboratory” included in text.

Page 14, line 11: suggest changing identical to virtually identical as there is a small difference of 1 DU as seen in Figure 4.
“virtually” included.

Page 15, line 7: Confusing sentence, suggest to change: ...they do not allow to assess the Global-Umkehr technique comprehensively. to something like they do not allow the comprehensive assessment of the Global-Umkehr technique.
Changed as suggested.

Page 5, line 10: Spaces seem to be present between all equations and symbols and full stops, commas. This can be misleading in some instances. For example, Page 5, line 10 may be interpreted as a dot product.
All equations will be reformatted by AMT before publication according to their guidelines. No change to manuscript.

Table 1. There are spaces on either side of the endashes which are not consistent with endash ranges throughout the text.
The entire manuscript, including the table, will be reformatted by AMT before publication according to their guidelines. No change to manuscript.

Page 10, line 1: typo - MLS measure(s) thermal... - remove "s"
Grammar corrected.

Page 10, line 15: space after second open bracket.
Space removed.

Page 10, line 24: Suggest remove therefore or move to the start of the sentence -
Therefore,...
“Therefore” moved to front.

Page 17, line 15: should N be in parenthesis?
N enclosed in parentheses.

Page 19, line 8: Change Table 2 allows to assess retrievals... to something like Table 2 allows the assessment of retrievals...
Changed as suggested.

Page 19, lines 12 and 13: change to to between -6 % to 4 % and to between -5 % to 2 %
“between” inserted.

Page 19, line 18: remove is
“is” removed.

Page 19, lines 19 and 21: insert a space after the equals sign
Spaces inserted.

Page 19, line 19: Change to but it is consistent
“it” included.

Page 19, line 20: remove comma after standard
comma removed.

Page 20, line 8: Is (/2) meant to be there?
Yes. This is the way the SBUV instruments are identified by Miyagawa et al., 2014.

Page 20, line 23: change resembles to "resemble
Grammar corrected.

Page 22, line 8: change to ...have to be...
“be” included.

Page 22, line 24: change to ...2–3 % of those... (use an endash?)
Corrected as suggested.