Response to anonymous referee #1

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June 30, 2017

We would like to thank Referee #1 for the careful reading of our manuscript and for providing very constructive comments which certainly helped to improve the manuscript. This document includes all the referee's comments as well as our replies to every one of them. The changes in the manuscript are shown in blue and the text simply removed is crossed out in red.
As the **General Comments** from the Referee #1 are also mentioned in the **Smaller or more detailed comments** we will answer them separately in this section.

Smaller or more detailed comments

1. **Comments from the referee:** P1L10: The mean relative difference [singular] and its standard deviation increase with altitude up to 50% at 70 km. (I assume you mean that both the bias and the standard deviation are > 50%).

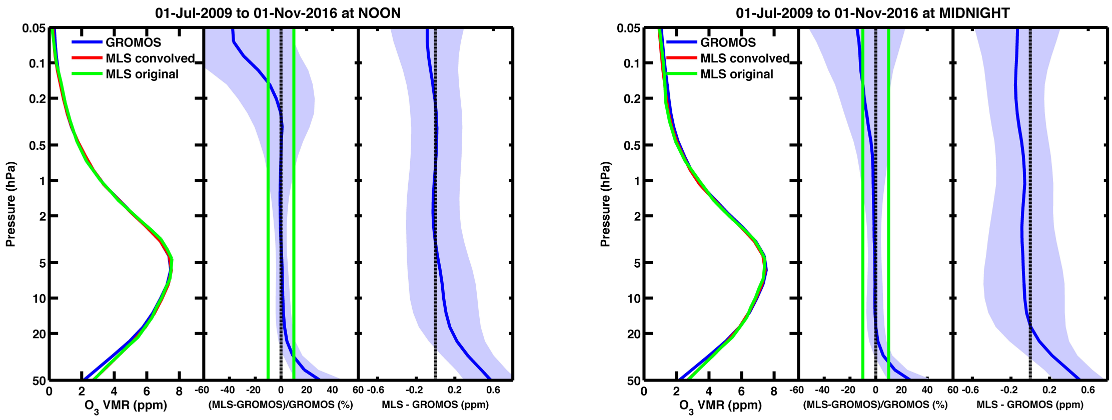
   Author's response:

   We have performed major changes in the comparison method. The criterion for spatial coincidence is now that horizontal distances between the sounding volumes of the satellite and the ground station have to be smaller than 1° in latitude and 8° in longitude. In addition, we have calculated the mean relative difference profile and the VMR difference profile separating daytime and nighttime values.

   Author's changes in the manuscript:

   P1L10: On average, GROMOS and MLS comparisons show agreement generally over 20% in the lower stratosphere and within 2% in the middle and upper stratosphere for both daytime and nighttime, whereas in the mesosphere the mean relative difference is below 40% at daytime and below 15% at nighttime.

   P4L17: The selected criterion for spatial coincidence is that horizontal distances between the sounding volumes of the satellite and the ground station have to be smaller than 1° in latitude and 8° in longitude. The present study extends over the period from July 2009 to November 2016 and covers the stratosphere and middle mesosphere from 50 to 0.05 hPa (from 21 to 70 km), and according to the spatial and temporal criteria, more than 2800 coincident profiles are available for the comparison. Figure 3a and Figure 3b show the mean ozone profiles of the collocated and coincident measurements of GROMOS (blue line), MLS convolved (red line) and MLS original (green line) at daytime and nighttime, respectively. The relative difference profile in percent given by $(\mathbf{x}_{\mathrm{MLS,low}} - \mathbf{x}_{\mathrm{GROMOS}})/\mathbf{x}_{\mathrm{GROMOS}}$ is displayed in the middle panel of both Figure 3a and Figure 3b along with the standard deviation of the differences (blue area). The green line delimits the ± 10% area. The mean profile of the VMR differences is shown in the right panel of both Figure 3. The mean relative differences and the VMR differences at daytime (nighttime) are over 20% or 0.5 ppm (15% or 0.4 ppm) in the lower stratosphere and decreasing with altitude up to 0.7% or 0.02 ppm (2% or 0.06 ppm) at the stratopause and increasing with altitude up to 38% or 0.085 ppm (15% or 0.12 ppm) at 0.05 hPa (70 km). We conclude from Figure 3 that during nighttime GROMOS measures more O_3 VMR (ppm) than MLS except for the lower stratosphere, where MLS measures more O_3 VMR (ppm) than GROMOS, both at daytime and nighttime. Nevertheless in the mesosphere GROMOS measures more O_3 VMR (ppm) than MLS, both at daytime and nighttime.



(a) Daytime (b) Nighttime

Figure 3: Mean ozone profiles recorded by GROMOS (blue line), MLS convolved (red line) and MLS original (green line) for the time interval between July 2009 and November 2016 are shown in the left panels of both daytime and nighttime Figures. The blue area (GROMOS) and the red area (MLS) are the standard deviations of the coincident measurements. The middle panels show the mean relative difference profile between data of both instruments, GROMOS as reference. The blue areas in the middle panels represent the standard deviation of the differences. The green lines in the middle panel delimit the ± 10% area. The mean VMR difference profile and its standard deviation (blue area) are displayed in the right panels of both daytime and nighttime, Figure 3a and Figure 3b, respectively

P6L24: The agreement between measurements coincident in space and time for both data records is within 2% (0.06 ppm) between 30 and 50 km (15–0.7 hPa) increasing up to 20% (0.5 ppm) at 20 km (50 hPa), for both daytime and nighttime. In the mesosphere the difference increases up to 38% (0.085 ppm) at daytime and up to 15% (0.12 ppm) at nighttime at 70 km (0.05 hPa).

2. **Comments from the referee:** P1L15: not sure what is meant by “anomaly” here (better to use words like “wintertime enhancement”).

Author’s response:

We agree on the referee’s comment and the text has been modified according to it.

We have decided to remove this line (P1L15).

Author's changes in the manuscript:

P1L15: ~~On the other hand, the amplitude of the diurnal variation, night-to-day ratio (NDR), is not as strong as the observed one at higher latitudes, nevertheless we observe the winter anomaly of the night-to-day ratio.~~

P6L16: ... the expected wintertime enhancement of the NDR

P6L29: Moreover, the wintertime enhancement of nighttime ...

3. **Comments from the referee:** P1L19/20: "... are its independence from solar irradiation and ..."

   Author's response:

   No comments.

   Author's changes in the manuscript: P1L19/20: ... are its independence from ...

4. **Comments from the referee:** P1L22/23: I suggest more concise wording, e.g. "Stratospheric ozone, in spite of its small abundance, plays a beneficial role by absorbing ..."

   Author's response:

   We agree on the referee's comment. The text has been modified according to it.

   Author's changes in the manuscript: P1L22/23: Stratospheric ozone, in spite of its small abundance, plays a beneficial role by absorbing ...

5. **Comments from the referee:** P2L1, I would delete "Thus" at the beginning of the sentence.

   Author's response:

   No comments.

   Author's changes in the manuscript: P2L1: ... of the atmosphere. Continuous ...

6. **Comments from the referee:** P2L23: Suggested wording, "source of odd-hydrogen, coupled with no decrease [or no change] in the production of odd-oxygen..."

   Author's response:

   We agree on the referee's comment. The text has been modified according to it.

   Author's changes in the manuscript: P2L20: Marsh et al. (2001) interpreted the tertiary peak by considering that in the middle mesosphere during winter, with solar zenith angle close to 90°, the atmosphere becomes optically thick to UV radiation at wavelengths below 185 nm and, since photolysis of water vapour (Reaction 1) is the primary source of odd-hydrogen, reduced UV radiation results in less odd-hydrogen. The lack of odd-hydrogen needed for the catalytic depletion of odd-oxygen (Reactions 2, 3 and 4), in conjunction with an unchanged rate of odd oxygen production (Reaction 5), leads to an increase in odd-oxygen. This results in higher ozone concentration because atomic oxygen recombination (Reaction 6) remains as a significant source of ozone in the mesosphere. Additionally, Hartogh et al. (2004) extended the interpretation by considering the very slow decrease of the ozone dissociation (Reaction 7) rate with increasing solar zenith angle.

$$\mathrm{H_2O} + \mathrm{h}\nu(\lambda < 185nm) \longrightarrow \mathrm{OH} + \mathrm{O} \qquad (1)$$

$$\mathrm{O + OH \longrightarrow O_2 + H} \tag{2}$$

$$\mathrm{H + O_2 + M \longrightarrow HO_2 + M} \tag{3}$$

$$\mathrm{O + HO_2 \longrightarrow O_2 + OH} \tag{4}$$

$$\mathrm{O_2 + h\nu(\lambda < 242 nm) \longrightarrow O + O} \tag{5}$$

$$\mathrm{O + O_2 + M \longrightarrow O_3 + M} \tag{6}$$

$$\mathrm{O_3 + h\nu \longrightarrow O_2 + O} \tag{7}$$

7. **Comments from the referee:** P2L29: a short discussion, and the conclusions are summarised in Section 5.

   Author's response:
   No comments.

   Author's changes in the manuscript: P2L29: a short discussion, and the conclusions ...

8. **Comments from the referee:** P3L26: Is the estimate of the a priori contribution not (more precisely) equal to 1 - the area, rather than the area itself? Then also, "We consider that the retrieval range is reliable where the true state dominates over the a priori information, ... I would note that this new retrieval characteristic is indeed quite different from past GROMOS papers, where it was not as well characterised near 0.05 hPa, but showing how the new and old retrieval compare, both in biases and in temporal behaviour, would be very useful in order for the reader to decide how these are different (and how different versus MLS also). It is not immediately clear what helps to provide the extra information at high altitudes that was not present in earlier retrievals (clarify please). Usually this can come if one adds spectral channels, for example, or if one changes the smoothing characteristics in the retrievals (obtaining noisier retrievals but with more vertical information). In this respect, you quote the vertical resolution of the new retrieval, so comparing that to the old version would be useful as well.

   Author's response:
   We agree with the referee, an estimation of the a priori contribution is 1 minus the area of the averaging kernels.
   In accordance with the referee wishes we have performed a comparison between version 2021 and version 150 of the retrieval of GROMOS.

   Author's changes in the manuscript: P3L26: AVKs are a representation of the weighting of information content of the retrieval parameters therefore an estimate of the a priori contribution to the retrieval can be obtained by 1 minus the area of the AVK (measurement response).
   P3L12: Recently, we have developed a new retrieval version (version 150) with the aim to optimise the averaging kernels. The differences with the former version (version 2021) are in the a priori covariance matrix, in the measurement error and in the integration time of the retrieval.
   In version 2021 the diagonal elements of the a priori covariance matrix are variable relative errors ranging from 35% at 100 hPa to 28% in the lower stratosphere and increasing with altitude from 35% in the upper stratosphere up to 70% in

the mesosphere. Meanwhile, in version 150 the a priori covariance matrix has a constant value for the diagonal elements of 2 ppm. For both retrieval versions the off-diagonal elements of the a priori covariance matrix exponentially decrease with a correlation length of 3 km.

Regarding the measurement noise, in version 2021 it is a constant error of 0.8 K whereas in version 150 we used a variable error depending on the tropospheric transmission:

$$\Delta T_b' = 0.5 + \frac{\Delta T_b}{e^{-\tau}} \tag{8}$$

the error of the measured brightness temperature, ΔT_b, is given by the radiometer equation:

$$\Delta T_b = \frac{T_b + T_{rec}}{\sqrt{\Delta f \cdot t_{int}}} \tag{9}$$

The radiometer equation gives the resolution of the radiation measured, which is determined by the bandwidth of the individual spectrometer channels (Δf), by the integration time (t_{int}) and by the total power measured by the spectrometer. A constant error of 0.5 K is considered as a systematic bias of the spectra, due to spectroscopic errors and the water vapour continuum. The error of the brightness temperature (ΔT_b) is of the order of a few Kelvins in the line centre and 0.5 K in the line wings of the spectrum. Therefore the measurement noise ($\Delta T_b'$) depends on the bandwidth of the spectrum and on the tropospheric transmittance. This is a more realistic approach for the retrieval than considering a constant measurement noise, resulting in an improvement in the retrieved ozone VMR in the lower stratosphere. The sampling time for version 150 is 1 hour and in case of version 2021 is 30 minutes. Longer integration time improves the retrieved ozone VMR at upper altitudes.

P4L1: In version 2021, the vertical resolution lies generally within 10–15 km in the stratosphere and increases with altitude to 20–25 km in the lower mesosphere. Between 20 to 52 km (50 to 0.5 hPa) the measurement response is higher than 0.8. For more details on version 2021 we refer to Moreira et al. (2015). Comparing the measurement response and the vertical resolution obtained by version 2021 and by version 150 we can conclude an improvement in the results retrieved by version 150. We assume that the changes performed in the a priori covariance matrix, in the measurement noise and in the integration time result in the improvement of the retrieval product, mainly observed in the lowermost and in the uppermost limit of the retrieved ozone VMR profile.

9. **Comments from the referee:** P4L4: For the heading, why not capitalise 'Microwave Limb Sounder" also? Proper documentation/reference for the MLS data should be included. For example, the MLS website points to Data Quality Documentation (Livesey et al.) for version 4 data (including how to properly screen the data), and there are past references for validation as well (including Boyd et al., JGR, 2007, mentioned here already).

Author's response:

No comments.

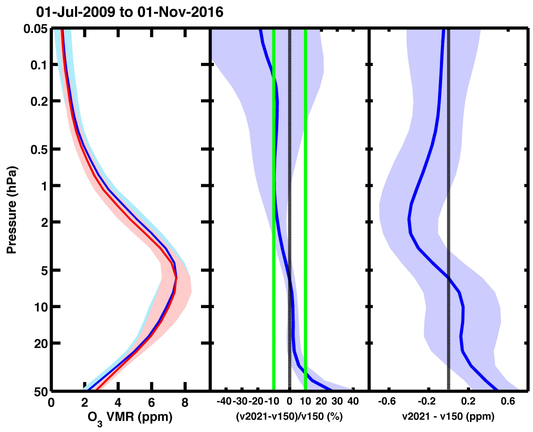


Figure 1: Mean ozone profiles retrieved by version 2021 (red line in the left panel) and by version 150 (blue line in the left panel) measured by GROMOS during the period from July 2009 to November 2016. The blue area (v150) and the red area (v2021) are the standard deviations of the ozone VMR. The mean relative difference profile (blue line) and the standard deviation of the differences (blue area) are represented in the middle panel, using the new version as reference. The green line delimits the ±10% area. In the right panel is shown the VMR difference profile along with its standard deviation

Author's changes in the manuscript:

P4L2: **The Aura Microwave Limb Sounder**

P4L5: The satellite overpasses the GROMOS measurement location (at northern midlatitudes) twice a day, approximately around noon and midnight. The standard product for ozone is derived from MLS radiance measurements near 240 GHz. The vertical resolution of the ozone profiles ranges from 3 km in the stratosphere to 6 km in the mesosphere (Schwartz et al., 2008). The present study has used ozone profiles from version 4.2. A summary of the quality of version 4.2 Aura MLS Level 2 data can be found in Livesey et al. (2016). Details about the Aura mission can be found in Waters et al. (2006).

10. **Comments from the referee:** P5L2: Change "relies" to "lies".

Author's response:

We have removed this sentence.

Author's changes in the manuscript: P5L1: ~~This result is in agreement with other comparisons performed between ground-based microwave radiometers and spaced-based instruments above Switzerland, where the bias among data sets relied within 5–10 % in the stratosphere and up to 50% towards the mesosphere (Studer et al., 2013; Barras et al., 2009; Hocke et al., 2007; Dumitru et al., 2006; Calisesi et al., 2005).~~

11. **Comments from the referee:** P5L13: Change altitudes to altitude. Also, the last sentence in section 3 does not convey anything new and could be easily deleted.

Author's response:

No comments.

Author's changes in the manuscript:

P5L13: ... for the altitude above ...

P5L14: ~~To sum up we can reiterate the fairly good agreement obtained for the comparison between ozone VMR profiles recorded by the ground-based instrument (GROMOS) and by the spaced-based instrument (Aura/MLS) during the time interval between July 2009 and November 2016 for the altitude range from 20 to 70 km.~~

12. **Comments from the referee:** P6L2: typo in 'Germany".

    Author's response:

    Thanks for spotting. We have corrected this.

    Author's changes in the manuscript: P6L2: ... (Germany, ...

13. **Comments from the referee:** P6L14: Change "shown" to "show"; delete "the" before 'Figure 6".

    Author's response:

    No comments.

    Author's changes in the manuscript: P6L14: ... the second panel of Figure 7 show a moving average over 7 data points (1 week) with the aim to clarify the understanding of Figure 7

14. **Comments from the referee:** P6L17: I suggest "although the latter data exhibit larger amplitudes".

    Author's response:

    No comments.

    Author's changes in the manuscript: P6L17: ..., although the latter data exhibit larger amplitudes.

15. **Comments from the referee:** P6L18: whereas at Lindau, winter-to-summer values vary by a factor of 2–3 ...

    Author's response:

    No comments.

    Author's changes in the manuscript: P6L18: ..., whereas at Lindau, winter-to-summer values vary by a factor of 2–3 at 70 km ...

16. **Comments from the referee:** P6L19: definition of the MMM being restricted to high latitudes, we can report its observation with a smaller amplitude at mid-latitudes.

    Author's response:

    No comments.

    Author's changes in the manuscript: P6L19: Thus, despite the definition of the MMM being restricted to high latitudes, we can report its observation with a smaller amplitude at mid-latitudes.

17. **Comments from the referee:** P6L23: Change "spaced-based" to "space-based".

Author's response:
Thanks for spotting. We have corrected this.

Author's changes in the manuscript: P6L23: ... by the space-based ...

18. **Comments from the referee:** P6L26: "we report good agreement between the new retrieval..."

    Author's response:
    No comments.

    Author's changes in the manuscript: P6L26: In general terms, we report good agreement between the new retrieval ...

19. **Comments from the referee:** P6L27: Change "Further" to "Furthermore".

    Author's response:
    No comments.

    Author's changes in the manuscript: P6L27: Furthermore, we observe

20. **Comments from the referee:** Fig 2. I would say "The middle panel shows the mean relative difference..." Also, The mean absolute difference and its uncertainty (blu area) are displayed in the right panel. [with a period after the last word in the Fig. captions]. By the way, more needs to be clarified here: is this for daytime or nighttime (presumably not) or for an average of day and night? The red line could be made thinner to allow one to see the blue line below it, or make the red line dashed maybe.

    Author's response:
    A new Figure 3 is displayed in the first comment. In this new Figure 3 the comparison between GROMOS and MLS was performed by separating daytime (Figure 3a) and nighttime (Figure 3b) values.

    Author's changes in the manuscript: Figure 3

21. **Comments from the referee:** Fig. 3: Is this for nighttime data only or both averaged (it may not matter too much at these lower altitudes but still worth clarifying)?

    Author's response:
    Former Figure 3 is now Figure 4, and in both the data represented is the average between daytime and nighttime data.

    Author's changes in the manuscript: P5L4: For an overview on the differences between coincident profiles, the average over daytime and nighttime values of the ozone VMR (ppm) time series of GROMOS (blue line) and MLS (red line) are displayed in Figure 4 for different pressure levels.

22. **Comments from the referee:** Fig. 4: Same question as for Fig. 3 (same answer presumably).

    Author's response:
    Former Figure 4 is now Figure 5 and in both the data represented is the average between daytime and nighttime data.the data represented is the average between daytime and nighttime data.

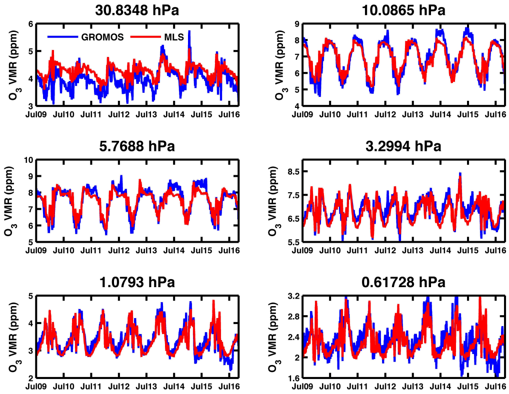


Figure 4: Time series of averaged daytime and nighttime O_3 VMR measurements of GROMOS (blue line) and MLS (red line) for the period from July 2009 to November 2016 at different pressure levels. An averaging kernel smoothing has been applied to the series of the MLS measurements coincident in time and space with the GROMOS measurements. Both time series are smoothed over 7 points or 1 week in time by a moving average

Author's changes in the manuscript: P5L10: In Figure 5 are shown the scatter plots of averaged daytime and nighttime O_3 VMR measurements of GROMOS and MLS at the same pressure levels as Figure 4.

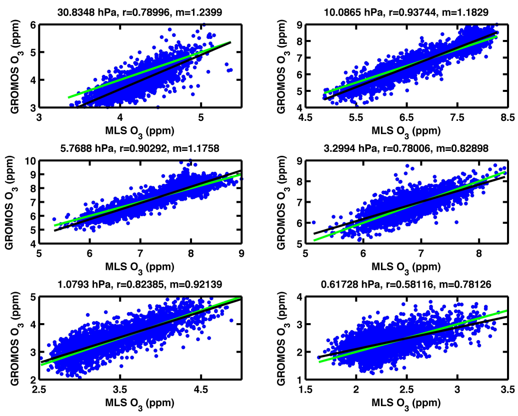


Figure 5: Scatter plots of coincident O_3 VMR averaged over daytime and nighttime measurements of GROMOS and MLS for the period from July 2009 to November 2016 at different pressure levels. The black line is the linear fit of both time series. The green line indicates the case of identity, O_3(MLS)=O_3(GROMOS). r values are correlation coefficients of the MLS and GROMOS time series

23. **Comments from the referee:** Fig. 5: Change "ans the second panel" to "and the second panel".

Author's response:

Thank for spotting. We have corrected this. Former Figure 5 is now Figure 6.

Author's changes in the manuscript: Caption of Figure 6: ... and the second panel ...