

# Response to anonymous referee #1

Lorena Moreira

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  $1^\circ$  in latitude and  $8^\circ$  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  $1^\circ$  in latitude and  $8^\circ$  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  $(\mathbf{x}_{\text{MLS,low}} - \mathbf{x}_{\text{GROMOS}})/\mathbf{x}_{\text{GROMOS}}$  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  $\pm 10\%$  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<sub>3</sub> VMR (ppm) than MLS except for the lower stratosphere, where MLS measures more O<sub>3</sub> VMR (ppm) than GROMOS, both at daytime and nighttime. Nevertheless in the mesosphere GROMOS measures more O<sub>3</sub> VMR (ppm) than MLS, both at daytime and nighttime.

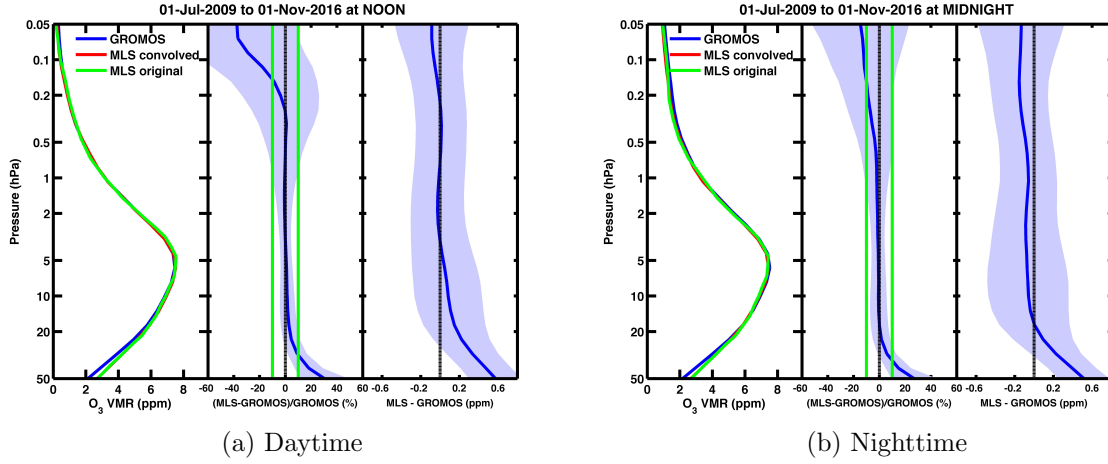


Figure 3: Mean ozone profiles recorded by GROMOS (blue line), MLS convolved (red line) and MLS original (green line) for the time interval between July 2009 and November 2016 are shown in the left panels of both daytime and nighttime Figures. The blue area (GROMOS) and the red area (MLS) are the standard 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  $\pm 10\%$  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 ...

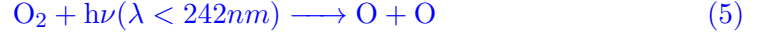
6. **Comments from the referee:** P2L23: Suggested wording, “source of odd-hydrogen, coupled with no decrease [or no change] in the production of odd-oxygen...”

**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.





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:

$$\Delta T'_b = 0.5 + \frac{\Delta T_b}{e^{-\tau}} \quad (8)$$

the error of the measured brightness temperature,  $\Delta T_b$ , is given by the radiometer equation:

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

The radiometer equation gives the resolution of the radiation measured, which is determined by the bandwidth of the individual spectrometer channels ( $\Delta f$ ), 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 ( $\Delta T_b$ ) 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.

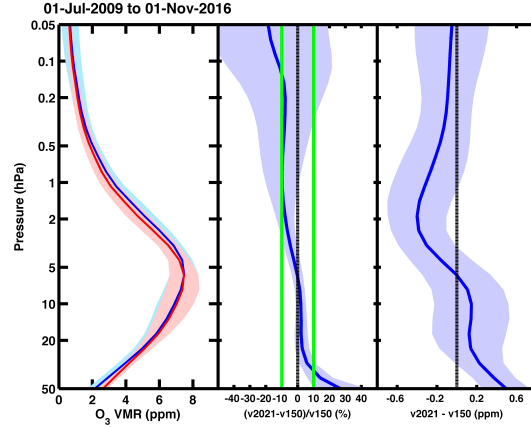


Figure 1: Mean ozone profiles retrieved by version 2021 (red line in the left panel) and by version 150 (blue line in the left panel) measured by GROMOS during the period from July 2009 to November 2016. The blue area (v150) and the red area (v2021) are the standard deviations of the ozone VMR. The mean 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  $\pm 10\%$  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 space-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 (blue 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.

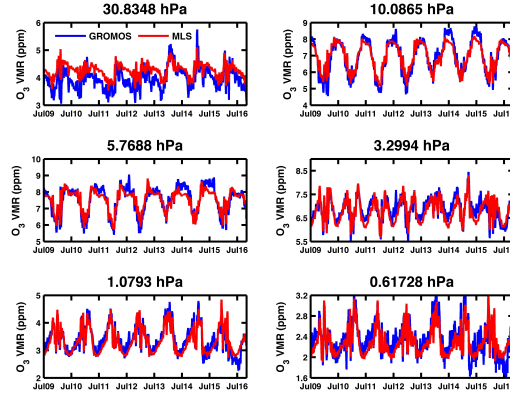


Figure 4: Time series of averaged daytime and nighttime  $O_3$  VMR measurements of GROMOS (blue line) and MLS (red line) for the period from July 2009 to November 2016 at different pressure levels. An averaging kernel smoothing has been applied to the series of the MLS measurements coincident in time 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.

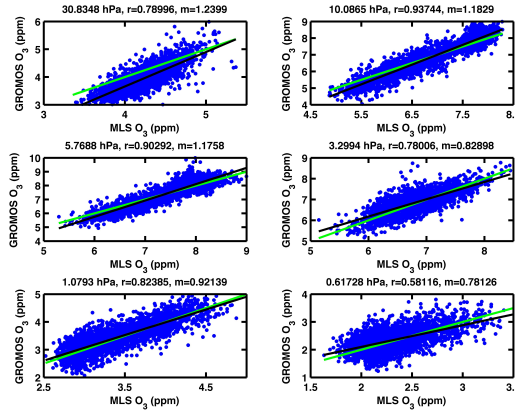


Figure 5: Scatter plots of coincident  $O_3$  VMR averaged over daytime and nighttime measurements of GROMOS and MLS for the period from July 2009 to November 2016 at different pressure levels. The black line is the linear fit of both time series. The green line indicates the case of identity,  $O_3(MLS)=O_3(GROMOS)$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

# Response to anonymous referee #2

Lorena Moreira

June 30, 2017

We are very thankful to the anonymous Referee #2 for the evaluation of our manuscript and for the valuable comments that helped significantly to improve the quality of the paper. We have revised the manuscript by following each one of your suggestions. Below we try to answer each comment. The changes in the manuscript are shown in blue and the text simply removed is crossed out in red.

## Specific comments

1. **Comments from the referee:** Pg. 1, Ln 13-15: This sentence presents a repetition that should be removed.

### Author's response:

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

**Author's changes in the manuscript:** Pg. 1, Ln 13-15: ~~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.~~

2. **Comments from the referee:** Pg. 2, Ln 8-10: If GROMOS data have been validated in the past what is the need of an additional comparison with Aura MLS? Differently, if the comparison with MLS serves as a validation of the new retrieval version, then a comparison of the new version with previous versions should also be present.

### Author's response:

We agree with the referee and we have performed a comparison between version 2021 and version 150 of the retrieval of GROMOS.

**Author's changes in the manuscript:** Pg. 3, Ln 12: ~~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:

$$\Delta T'_b = 0.5 + \frac{\Delta T_b}{e^{-\tau}} \quad (1)$$

the error of the measured brightness temperature,  $\Delta T_b$ , is given by the radiometer equation:

$$\Delta T_b = \frac{T_b + T_{rec}}{\sqrt{\Delta f \cdot t_{int}}} \quad (2)$$

The radiometer equation gives the resolution of the radiation measured, which is determined by the bandwidth of the individual spectrometer channels ( $\Delta f$ ), 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 ( $\Delta T_b$ ) 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.

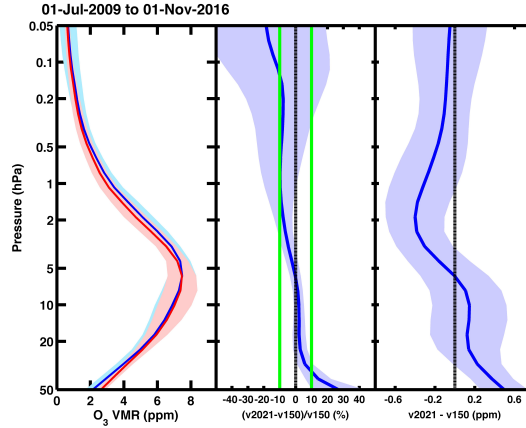


Figure 1: Mean ozone profiles retrieved by version 2021 (red line in the left panel) and by version 150 (blue line in the left panel) measured by GROMOS during the period from July 2009 to November 2016. The blue area (v150) and the red area (v2021) are the standard deviations of the ozone VMR. The mean 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  $\pm 10\%$  area. In the right panel is shown the VMR difference profile along with its standard deviation



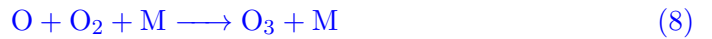
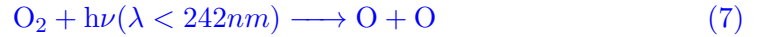
Pg. 4, Ln 1: In version 2021, the vertical resolution lies generally within 10–15 km in the stratosphere and increases with altitude to 20–25 km in the lower mesosphere. Between 20 to 52 km (50 to 0.5 hPa) the measurement response is higher than 0.8. For more details on version 2021 we refer to Moreira 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.

3. **Comments from the referee:** Pg. 2, Ln 23-24: Awkward sentence

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Pg. 2, Ln 20-26: 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 3) 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 4, 5 and 6), in conjunction with an unchanged rate of odd oxygen production (Reaction 7), leads to an increase in odd-oxygen. This results in higher ozone concentration because atomic oxygen recombination (Reaction 8) 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 9) rate with increasing solar zenith angle.



4. **Comments from the referee:** Pg. 2, Ln 27: I would remove this sentence, or place it elsewhere.

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Pg. 2, Ln 27: ~~This publication presents a new comparison between a ground-based instrument (GROMOS) and a spaced-based instrument (Aura/MLS).~~

5. **Comments from the referee:** Pg. 3, Ln 9: What a priori information are you referring to? Temperature and pressure profiles? What about the ozone a priori profile?

**Author's response:**

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

**Author's changes in the manuscript:** Pg. 3, Ln 9: The a priori profile of O<sub>3</sub> VMR required for the retrieval is taken from a monthly varying climatology from ECMWF reanalysis until available (70 km) and extended above by an Aura MLS climatology (2004 to 2011). The line shape used in the retrieval is the representation of the Voigt line profile from Kuntz (1997). Spectroscopic parameters to calculate the ozone absorption coefficients were taken from the JPL catalogue (Pickett et al., 1998) and the HITRAN spectroscopic database (Rothman et al., 1998). The atmospheric temperature and pressure profiles are taken from the 6 hourly of the European Centre for Medium-Range Weather Forecast (ECMWF) operational analysis data and are extended above 80 km by monthly mean temperatures of the CIRA-86 Atmosphere Model (Fleming et al., 1990).

6. **Comments from the referee:** Pg. 3, Ln 18: Why do you have a systematic bias in the spectral measurements?

**Author's response:**

We do have systematic biases in the spectral measurements due to spectroscopic errors and the water vapour continuum.

**Author's changes in the manuscript:** Pg. 3, Ln 18: In addition, 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.

7. **Comments from the referee:** Pg. 3, Ln 19: Even though the authors cite earlier papers describing in more details the technical aspects of the measurements, I think Figure 1 should still show an example of the spectrum measured and specify whether the 1-hour average spectrum is binned before deconvolving it. Are all channels binned in groups? Also those near the line center? This is critical for the high altitude comparison. Additionally, maybe a table similar to Table 1 of Moreira et al., 2015, would be a useful reminder of the main characteristics of GROMOS.

**Author's response:**

The referee is right to ask about an example of the spectrum measured and about a table of the GROMOS instrument specifications, yet we have not performed any instrumental change, therefore we can refer to Moreira et al. (2015) for these details.

The fast Fourier transform spectrometer (FFTS) has around 32768 channels and after the binning in frequency the number of points in frequency are 54 with high frequency resolution in the line centre compare to the line wings.

**Author's changes in the manuscript:** No changes.

8. **Comments from the referee:** Pg. 3, Ln 22: In figure 1, a priori and retrieved profiles are terribly close. I am aware that in the altitude region where the retrieval

algorithm is the most sensitive the a priori has a very small impact on the profile retrieved, yet it would be nice to see it. Most readers don't know and will wonder what's the point of the measurement if the climatology from other datasets already provides you with the true state.

**Author's response:**

In the middle panel of Figure 2 (former Figure 1) are shown the averaging kernels and the area of the averaging kernels, called measurement response. The averaging kernels are a key quantity for the characterisation of the retrieved profiles. It describes how the retrieval smoothes the true state and how sensitive it is to the a priori profile. The averaging kernel lines in the middle panel are shown in colour to help their interpretation, for instance, the green line shows the kernel line at 30 km and we can clearly see that the kernel actually peaks at 30 km. The measurement response is an indicator of the sensitive altitude range of the retrieved profile, it accounts for the amount of information from the true state of the retrieved profile at a given altitude. The measurement response (MR) is shown in red in the middle panel. It is considered a reliable altitude range of the retrieval when the true state dominates over the a priori information, i.e. where the measurement response is larger than 0.8 (an a priori contribution smaller than 20%). The measurement response shown in Figure 2 is around 1 from 18 to 70 km. Therefore, from this we can conclude the retrieved profile of GROMOS measurements is actually the true state of the atmosphere above Bern and not an a priori representation of the true state obtained from a climatology of other datasets.

**Author's changes in the manuscript:** No changes.

9. **Comments from the referee:** Pg. 4, Ln 1: How is this an improvement with respect to the older version? Again, a comparison with the previous retrieval version is necessary

**Author's response:**

As previously mentioned we have performed a comparison between version 2021 and version 150 of the retrieval of GROMOS. See comment 2 for details on the changes in the manuscript.

**Author's changes in the manuscript:** See **Comment from the referee 2** for details on the changes in the manuscript.

10. **Comments from the referee:** Pg. 4, Ln 19: Are these criteria consistent? The spatial requirement seems particularly generous compared to the temporal one. How far does a parcel of stratospheric air travel in one hour? A mesospheric one? Would a stricter spatial criterion improve your comparison results in the upper stratosphere/mesosphere? In other words, you should motivate your choices of coincident criteria.

**Author's response:**

In accordance with the referee wishes 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  $1^\circ$  in latitude and  $8^\circ$  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:** Pg. 1, Ln 10: 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.

Pg. 4, Ln 17: 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  $(x_{\text{MLS,low}} - x_{\text{GROMOS}})/x_{\text{GROMOS}}$  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  $\pm 10\%$  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<sub>3</sub> VMR (ppm) than MLS except for the lower stratosphere, where MLS measures more O<sub>3</sub> VMR (ppm) than GROMOS, both at daytime and nighttime. Nevertheless in the mesosphere GROMOS measures more O<sub>3</sub> VMR (ppm) than MLS, both at daytime and nighttime.

Pg. 6, Ln 24: 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).

11. **Comments from the referee:** Pg. 4, Ln 21: I suggest “to” instead of “with the compliance of”

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Pg. 4, Ln 21: ... and according to the spatial and ...

12. **Comments from the referee:** Pg. 5, Ln 1: I am not sure what this sentence implies. Are you suggesting that either the ground-based or the satellite-based data are inevitably faulty at high altitudes? Additionally, if I am not mistaken, the manuscripts you cite are either on SOMORA retrievals (which reach 55 km at the most) or GROMOS itself. Are you suggesting that the present relatively large

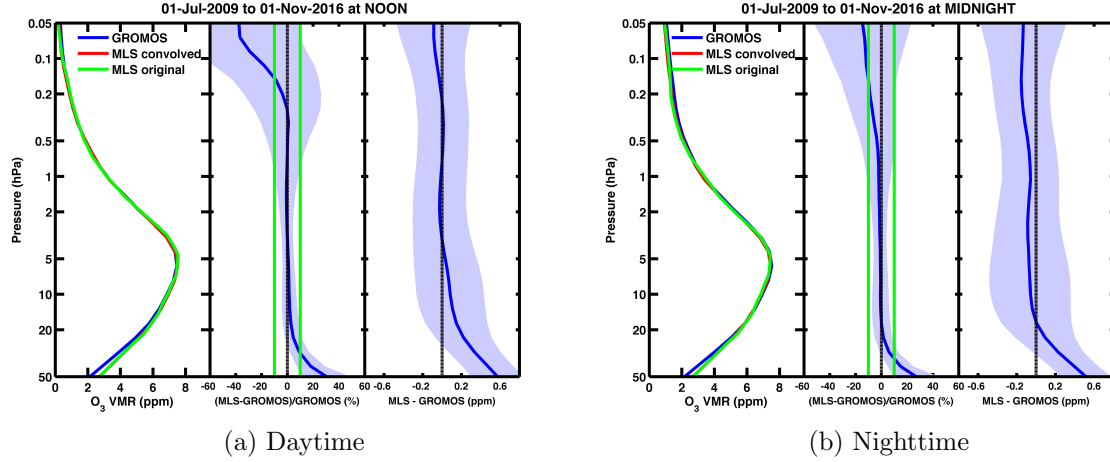


Figure 3: Mean ozone profiles recorded by GROMOS (blue line), MLS convolved (red line) and MLS original (green line) for the time interval between July 2009 and November 2016 are shown in the left panels of both daytime and nighttime Figures. The blue area (GROMOS) and the red area (MLS) are the standard 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  $\pm 10\%$  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

discrepancy in the GROMOS-MLS comparison at high altitude is likely to be due to GROMOS? If this is correct just say so.

**Author’s response:**

We agree on the referee’s comment and we have removed the sentence.

**Author’s changes in the manuscript:** Pg. 5, Ln 1: ~~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).~~

13. **Comments from the referee:** Pg. 5, Ln 4: I would write: “For an overview on the differences between coincident profiles, ...”

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Pg. 5, Ln 4: ~~For an overview on the differences between coincident profiles, ...~~

14. **Comments from the referee:** Pg. 5, Ln 11: I would quantify the “almost perfect” with the slope of the linear fit. Second to last sentence in Section 3 : Could this be due to the spatial coincidence criterion? Last sentence in Section 3: I would suggest to postpone this last sentence to the conclusions section.

**Author’s response:**

We agree on the referee’s comment therefore we have changed line 11 and we have removed the last sentence of Section 3.

**Author’s changes in the manuscript:**

Pg. 5, Ln1 1: The black lines, linear regression lines of the observations, are close to the green one to one lines,  $O_3(MLS)=O_3(GROMOS)$ .

Pg. 5, Ln 17–19: ~~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.~~

15. **Comments from the referee:** Pg. 6, Ln 2: This needs to be better explained. Specifically, what part of your results agree with the work of Sonnemann 2007 and what doesn’t. The fact that one dataset can peak at values that are twice as much as those of GROMOS seems an important difference. Do their data have a better vertical resolution? Retrievals that reach higher altitudes? Can you briefly address this difference?

**Author’s response:**

Our results on the annual variation of mesospheric ozone at Bern are in agreement with the ones observed at Lindau by Sonnemann et al. (2007). The result disagrees in the amplitudes of the annual variation however according to Sonnemann et al. (2007), *the MMM is an effect occurring at high latitudes close to the polar night terminator around 72 km altitude during nighttime in the winter half of the year and extends into middle latitudes with decreasing amplitude*. Sonnemann et al. (2007) show nighttime ozone mixing ratio at Lindau up to 80 km. The upper altitude limit for the retrieval of ozone at 142 GHz measured by GROMOS is approximately 75 km, due to the fact that height-resolved information cannot be retrieved in the Doppler broadening domain since the line width does not depend on altitude. We set our altitude limit up to 70 km where the measurement response is  $\sim 1$ , therefore we do not have contribution from the a priori.

**Author’s changes in the manuscript:** Pg. 6, Ln 1–2: Our results on the annual variation of mesospheric ozone at ...

Pg. 6, Ln 3: Disagreements appear in the amplitudes ...

16. **Comments from the referee:** Pg. 6, Ln 4–8: I would remove these two sentences as they were already stated in the introduction

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Pg. 6, Ln 4–8: ~~This maximum of mesospheric ozone during nighttime in winter is related to the middle mesospheric maximum of ozone (MMM) (e.g., Sonnemann et al., 2007; Hartogh et al., 2004) also known as the tertiary ozone maximum (e.g., Sofieva et al., 2009; Degenstein et al., 2005; Marsh et al., 2001). During winter, the photodissociation rate of water is reduced at high latitudes which leads to a decrease of catalytic ozone depletion by odd hydrogen.~~

17. **Comments from the referee:** Pg. 6, Ln 19: I would explicitly state what this anomaly is. Last two sentences in Section 4: It is not clear whether you ascribe the difference from Sonnemann et al. to the fact that Lindau is at higher latitudes. If this is the case, I would object that 5° latitude cannot make this large difference in mesospheric ozone values and that a latitude of 51.7 °N is not much higher than 47°N.

**Author's response:**

We acknowledge that “winter anomaly” is maybe not the best appellation so we have changed for “wintertime enhancement”.

According to Sonnemann et al. (2007), *the MMM is an effect occurring at high latitudes close to the polar night terminator around 72 km altitude during nighttime in the winter half of the year and extends into middle latitudes with decreasing amplitude*. The observed sharp decrease of the amplitude of the MMM of ozone is due to the strong latitudinal gradient between high and middle latitudes. In fact, it is surprising that we can observe the effect of MMM at our latitude. Therefore, the difference in latitude between Lindau and Bern may have such impact in the amplitudes of the annual variability of mesospheric ozone due to the MMM. However it could also be due to some other effects like for example, differences in the retrieval algorithms between Bern and Lindau, different instruments used to perform the measurements, different calculation methods...

**Author's changes in the manuscript:** Pg. 1, Ln 15: ~~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.~~

Pg. 6, Ln 19: ... the expected wintertime enhancement of the NDR

Pg. 6, Ln 32: Moreover, the wintertime enhancement of nighttime ...

Pg. 6, Ln 5: Nevertheless, our results are expected since this maximum of mesospheric ozone during nighttime in winter is related to the middle mesospheric maximum of ozone (MMM) and according to Sonnemann et al. (2007) its effect extends into midlatitudes with decreasing amplitude.

18. **Comments from the referee:** Pg. 6, Ln 27: Please, rephrase avoiding the repetition.

**Author's response:**

No comments.

**Author's changes in the manuscript:** Pg. 6, Ln 27: the diurnal variability and its amplitude, the night-to-day ratio (NDR).

19. **Comments from the referee:** Pg. 6, Ln 29: Together with the relative difference I would quote here also the absolute one, which is less than 0.2 ppmv, on average (if I read correctly from figure 2). Last sentence: I would specify what the anomaly is also here in the conclusions

**Author's response:**

No comments.

**Author's changes in the manuscript:** Pg. 6, Ln 29: 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).

Pg. 6, Ln 32: Moreover, the wintertime enhancement of nighttime ...

20. **Comments from the referee:** Figure 1:

- I would add a panel with the GROMOS 1-hour spectrum.
- I would enlarge, make it longer, the X-axis of the 3rd panel (maintaining the range 10-70 km).

**Author's response:**

As we highlighted previously, we have not performed any instrumental change, therefore we can refer to Moreira et al. (2015) for these details.

With all due respect to the referee we do not understand the reason for enlarging the X-axis of the 3rd panel (maintaining the range 10-70 km).

**Author's changes in the manuscript:** No changes.

21. **Comments from the referee:** Figure 2:

- Would it be useful to show two separate averages, one for the daytime and one for the nighttime comparison?
- I would reduce the range of the X-axis of the middle plot to be from -60% to 60%
- I would use the same vertical unit (altitude or/and pressure) in all the figures or, even better, use both of them all the times. In figure 1 there's altitude, in figure 2 there's pressure.

**Author's response:**

We have calculated the mean relative difference profile and the VMR difference profile separating daytime and nighttime values.

In Figure 2 (former Figure 1) we use altitude units in order to help in the interpretation of what it is shown.

**Author's changes in the manuscript:** See the new Figure 3.

22. **Comments from the referee:** Figure 3:

- I would make these plots much larger, removing one or two pressure levels if necessary.
- Please specify in the caption the number of points involved in the moving average Figure 4

**Author's response:**

With all due respect to the referee we think that the plots are larger enough to be properly interpreted.

Former Figure 3 is now Figure 4 and the number of points involved in the moving average is 7 points.

**Author's changes in the manuscript:** Caption of Figure 4: Time series of averaged daytime and nighttime O<sub>3</sub> 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

23. **Comments from the referee:** Figure 4:

- Same comment as for Figure 3: I would make these plots much larger, removing one or two pressure levels if necessary.
- I would add the numbers  $m$  and  $q$  in the equation  $y=mx+q$  for each linear fit, or at least the slope  $m$ .
- I am surprised by the relatively low correlation value at 0.617 hPa. By looking at figure 3 I was expecting a better result. Any comment?

**Author's response:**

With all due respect to the referee we think that the plots are large enough to be properly interpreted.

In accordance with the referee wishes we add the slope of every linear fit in the titles of plots which form Figure 5 (former Figure 4).

In our opinion this “low” correlation value can be expected from the time series at 0.617 hPa shown in Figure 5 (former Figure 4) since GROMOS measures more  $O_3$  VMR (ppm) for most of the summers under assessment.

**Author's changes in the manuscript:** Figure 5

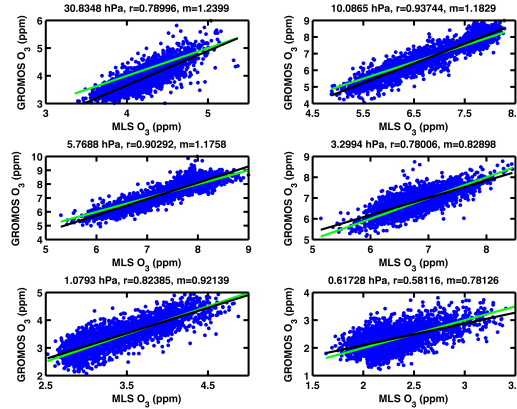


Figure 5: Scatter plots of coincident  $O_3$  VMR measurements of GROMOS and MLS for the period from July 2009 to November 2016 at different pressure levels. The black line is the linear fit of both time series, and  $m$  the slope of the linear fit. The green line indicates the case of identity,  $O_3(MLS)=O_3(GROMOS)$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

24. **Comments from the referee:** Figure 5:

- It would be useful to see a comparison of averaged nighttime vertical profiles, not just level 0.05 hPa, in order to establish, for example, whether the MLS  $O_3$  peak is at higher altitudes.
- As a matter of fact, it would be useful to see a comparison of GROMOS mesospheric profiles also with the averaged MLS original (not weighted with

GROMOS AVK) nighttime profiles, in order to understand the capabilities of GROMOS to spot the MMM with the “correct” intensity at the “correct” altitude.

- It would be best if line colors in the various figures were consistent, e.g., MLS always in red, GROMOS always in blue, and so on. In particular, maybe colors in Figure 5 could be changed (GROMOS in blue and cyan, MLS in red and orange?)
- Again, please in the caption state how many points are included in the average
- In the bottom plot I would add the standard deviation of the mean for both GROMOS and MLS.

**Author’s response:**

We have analysed the MMM at different altitudes, and for instance, at 0.1 hPa ( $\sim 63$  km) the results are pretty similar to the ones obtained at 0.05 hPa, although with smaller amplitudes.

In accordance with the referee wishes we have repeated the Figure. Former Figure 5 is now Figure 6.

With all due respect to the referee we think that our colours choice for this Figure 6 is rather intuitive.

Regarding, the addition of the standard deviation of the mean for both data records we think that this choice would make the Figure noisy. The standard deviation of the mean is  $\sim 0.3$  ppm for GROMOS,  $\sim 0.2$  ppm for MLS convolved and  $\sim 0.5$  ppm for MLS original.

**Author’s changes in the manuscript:** Pg. 5, Ln 27: The first panel of Figure 6 displays the  $O_3$  VMR measured at noon (GROMOS in red, [MLS convolved in orange and MLS original in magenta](#)) and at midnight (GROMOS in blue, [MLS convolved in cyan and MLS original in black](#)) at 0.05 hPa (70 km) for the already mentioned time period. [The original MLS data, i.e. not weighted with GROMOS AVKs, is shown in order to provide an insight of the observability of the effect of MMM at northern midlatitudes by GROMOS.](#)

25. **Comments from the referee:** Figure 6: Given that the daytime mesospheric ozone at 0.05 hPa is relatively constant, the night to day ratio provides more or less the same information already present in Figure 5. Maybe I am wrong, but then the authors should make an effort in discussing this figure a little more.

**Author’s response:**

We acknowledge that the night-to-day ratio (NDR) just provides information about the amplitude of the diurnal and seasonal variability of mesospheric ozone, nevertheless we want to keep it in the manuscript in order to be comparable with the study of Sonnemann et al. (2007).

**Author’s changes in the manuscript:** No changes.

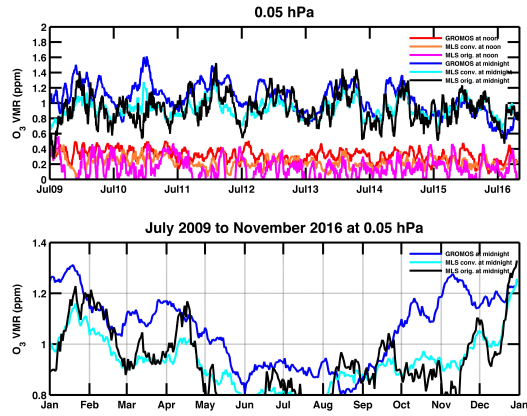


Figure 6: The first panel shows the diurnal variation of O<sub>3</sub> VMR measured at noon (GROMOS in red, MLS convolved in orange and MLS original in magenta) and at midnight (GROMOS in blue, MLS convolved in cyan and MLS original in black) at 0.05 hPa (70 km) and the second panel shows its evolution throughout the year averaged for the time interval under assessment (July 2009–November 2016). All time series are smoothed in time by a moving average over 15 points (1 week)

# Response to anonymous referee #3

Lorena Moreira

June 30, 2017

We are very grateful to Referee #3 for the useful and valuable comments which provided insights that helped significantly to improve the manuscript. All proposed objections and suggestions have been taken into account and discussed. Below we try to answer every comment. The changes in the manuscript are shown in blue and the text simply removed is crossed out in red.

## More specific comments

1. **Comments from the referee:** Page 1, line 4: “for the retrieval of” is odd wording: “A new version of the ozone profile retrievals...”

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 1, line 3–4: A new version of the ozone profile retrievals has been ...

2. **Comments from the referee:** Page 1, line 8: Shouldn’t it be “GROMOS and Aura MLS profiles agree within 3% on average for ...”, or “Average GROMOS and ...” or “On average, GROMOS and ...”?

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 1, line 8: On average, GROMOS

3. **Comments from the referee:** Page 1, lines 12/13: The sentence that spans these lines is poorly worded. “This behavior is related to ...” is probably better. Also “On the other hand” is an inappropriate way in which to begin the sentence that follows.

**Author’s response:**

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

**Author’s changes in the manuscript:** Page 1, lines 12/13: This behavior is related to ...

Page 1, lines 13/15: ~~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.~~

4. **Comments from the referee:** Page 1, line 19: “its” → “their”

**Author’s response:**

Thanks for spotting. We have corrected this.

**Author’s changes in the manuscript:** Page 1, line 19: information about [their](#) distribution ...

5. **Comments from the referee:** Page 1, line 22: The assertion that this family of measurements have been indispensable would benefit from some citations that back that point up.

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 1, line 22: Measurements of ozone performed by this technique have been indispensable in monitoring changes in the ozone layer and improving the comprehension of the processes that control ozone abundances ([e.g. Steinbrecht et al. 2009](#)).

6. **Comments from the referee:** Page 2, line 2: This sentence would also benefit from citations also ([e.g.](#), to some of the foundation documents for NDACC, or to GCOS [or similar] reports).

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 2, line 2: [Continuous](#) long-term monitoring of ozone is essential for the detection of long-term trends of the stratospheric ozone layer ([e.g. WMO, 2014](#)).

7. **Comments from the referee:** Page 2, line 10: “Furthermore” is inappropriate here. It’s generally used when introducing a third or greater point, not for a second point. I suggest “In addition, we have ...” or “We have also, ...”

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 2, line 10: We have [also](#) performed ...

8. **Comments from the referee:** Page 2, line 11: Badly constructed sentence. As written it sounds like there are two diurnal variations, one unspecified one, and one in mesospheric ozone, the amplitude of which you investigated.

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 2, line 11: We have [also](#) performed an analysis of the diurnal variation and [its amplitude \(night-to-day ratio\)](#) of middle mesospheric ozone, at 0.05 hPa (70 km).

9. **Comments from the referee:** Page 2, line 13/14. This explanation could be more complete, specifically, it would be good to give the timescale for the recombination. Presumably it’s  $\sim$  hours not  $\sim$  minutes, but needs to be made clear.

**Author’s response:**

We have changed the sentence.

**Author’s changes in the manuscript:** Page 2, line 13/14: Daytime production of atomic oxygen by photolysis of ozone (Reaction 7) and photolysis of molecular oxygen (Reaction 5) results in nighttime ozone production by recombination of atomic and molecular oxygen (Reaction 6).

10. **Comments from the referee:** Page 2, line 14: “Moreover” feels like the wrong word here. “In addition...” might be better.

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 2, line 14: In addition, we observe

11. **Comments from the referee:** Page 2, line 18: “an effect occurring at” → “a phenomenon that occurs at”

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 2, line 18: ... the MMM is a phenomenon that occurs at ...

12. **Comments from the referee:** Page 2, line 22: comma needed between “and” and “since”

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 2, line 22: ... 185 nm and, since photolysis ...

13. **Comments from the referee:** Page 2, lines 23/24: Badly worded sentence. Suggest: “The lack of odd-hydrogen needed for the catalytic depletion of odd-oxygen, in conjunction with an unchanged rate of odd oxygen production, leads to an increase in odd-oxygen”.

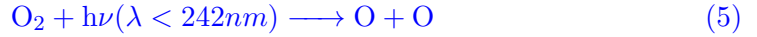
Regarding the discussion in this section of the paper, the more conventional way to frame it is to list some relevant reactions and then talk about the processes that give rise to maxima and diurnal cycles etc. in terms of those reactions. So we’d have sentences along the lines of “Lack of sunlight inhibits generation of odd hydrogen via reaction X, leading to enhancement in odd oxygen abundances due to continued production by reaction Y”, or something similar. The authors might want to consider taking that approach.

**Author’s response:**

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

**Author’s changes in the manuscript:** Page 2, lines 20/24: 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.



14. **Comments from the referee:** Page 3, Section 2.1. This section would benefit from having a few more details concerning the instrument. In particular, no information is given on the bandwidth of the observed spectrum, the spectral resolution, or the receiver noise temperature etc. These are all key parameters needed to get a sense of the measurement system. A plot showing a sample spectrum and associated error bars would be most welcome. For example, there's little point talking about adding 0.5K to the noise here or there without giving the reader a sense of how big the  $T_{rec}/\sqrt{B \tau}$  number is. At what altitude does Doppler broadening start to dominate over pressure broadening for this line? Also, presumably the retrievals need to assume a temperature (and height?) profile. Some information on where that is taken from, and the sensitivity of the result to it would be useful to give.

**Author's response:**

The referee is right to ask about more details concerning the instrument, yet for these details we refer to Moreira et al. (2015).

This 0.5 K added to the noise is due to spectroscopic errors and the water vapour continuum.

The Doppler broadening starts to dominate above 75 km, in case of ozone at 142 GHz.

We agree on the referee's comment about more information on the temperature and pressure profiles needed for the retrieval. The text has been modified according to it.

**Author's changes in the manuscript:** Page 3, line 18: In addition, 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](#).

Page 3, line 9: [The a priori profile required for the retrieval is taken from a monthly varying climatology from ECMWF reanalysis until available \(70 km\) and extended above by an Aura MLS climatology \(2004 to 2011\). The line shape used in the retrieval is the representation of the Voigt line profile from Kuntz \(1997\). Spectroscopic parameters to calculate the ozone absorption coefficients were taken from the JPL catalogue \(Pickett et al., 1998\) and the HITRAN spectroscopic database \(Rothman et al., 1998\) The atmospheric temperature and pressure](#)

profiles are taken from the 6 hourly of the European Centre for Medium-Range Weather Forecast (ECMWF) operational analysis data and are extended above 80 km by monthly mean temperatures of the CIRA-86 Atmosphere Model (Fleming et al., 1990).

15. **Comments from the referee:** Page 3, line 8: Is the ozone a priori really taken from the ECMWF analysis? How useful is that up to 70km, what is it based on. A reference would be good.

**Author’s response:**

Yes, it is. The a priori ozone profile does not play a role since the measurement response, area of the averaging kernels, is equal to unity for the altitude range from 18 to 70 km.

**Author’s changes in the manuscript:** No changes.

16. **Comments from the referee:** Page 3, line 13: You tell us that v150 has a constant a priori, but don’t say how it behaved in 2021, it would be useful to know.

**Author’s response:**

In version 2021, as diagonal elements of the a priori covariance matrix we assume a relative error around 35% at 100 hPa. The error decreases in the lower stratosphere up to 28%. Then it increases linearly from 35% in the upper stratosphere to 70% in the lower mesosphere. The off-diagonal elements exponentially decrease with a correlation length of 3 km.

We have performed a comparison between version 2021 and version 150 of the retrieval of GROMOS.

**Author’s changes in the manuscript:**

Page 3, line 12: 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:

$$\Delta T'_b = 0.5 + \frac{\Delta T_b}{e^{-\tau}} \quad (8)$$

the error of the measured brightness temperature,  $\Delta T_b$ , is given by the radiometer equation:

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

The radiometer equation gives the resolution of the radiation measured, which is determined by the bandwidth of the individual spectrometer channels ( $\Delta f$ ), 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 ( $\Delta T_b$ ) 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.

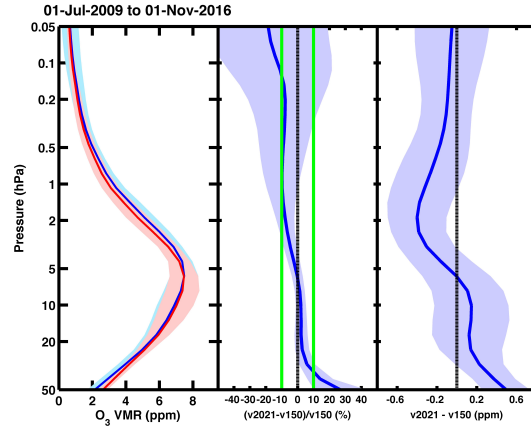


Figure 1: Mean ozone profiles retrieved by version 2021 (red line in the left panel) and by version 150 (blue line in the left panel) measured by GROMOS during the period from July 2009 to November 2016. The blue area (v150) and the red area (v2021) are the standard deviations of the ozone VMR. The mean 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  $\pm 10\%$  area. In the right panel is shown the VMR difference profile along with its standard deviation

Page 4, line 1: 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.

17. **Comments from the referee:** Page 3, line 13: “optimizing” in what sense, what

were you trying to optimize? The vertical range, resolution, what? [Or should you change the “and” on the same line to “by”?]

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 3, line 13: ..., thus optimizing the averaging kernels [by](#) improving ...

18. **Comments from the referee:** Page 3, line 15: This discussion is a little confusing. Earlier parts of the paper give the impression that this study of the diurnal cycle was, at least partly, enabled by the new GROMOS data version. However, here you talk about the new version being focused on improvements in the lower stratosphere. If there were improvements in the mesosphere, it would be best to be more specific about what they are and which of the changes (presumably among those discussed above) brought those improvements about.

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 3, line 15: ... the measurement response in the lower stratosphere [and in the mesosphere](#).

19. **Comments from the referee:** Page 3, lines 17/18: You need to define all of the terms in these equations, and give us the numbers for  $T_{rec}$ , B and tau.

**Author’s response:**

We have changed the sentence.

**Author’s changes in the manuscript:** Page 3, lines 17/18: [The error of the measured brightness temperature,  \$\Delta T\_b\$ , is due to noise fluctuations in the spectrum and is of the order of a few Kelvins in the line center and 0.5 K in the line wings of the spectrum.](#)

20. **Comments from the referee:** Page 3, line 23: “The AVKs are multiplied by 4 in figure 1 in order to ...”

**Author’s response:**

Thanks for spotting. We have corrected this. Former Figure 1 is now Figure 2.

**Author’s changes in the manuscript:** Page 3, line 23: The [AVKs](#) are multiplied by 4 [in Figure 2](#) in order ...

21. **Comments from the referee:** Page 3, line 24: AVK  $\rightarrow$  AVKs

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 3, line 24: [AVKs](#) ...

22. **Comments from the referee:** Page 4, line 5 (your numbers): “our location”  $\rightarrow$  “Bern” or “the GROMOS measurement location” or similar.

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 4, line 5: The satellite overpasses [the GROMOS measurement location](#) (at northern midlatitudes) twice a day

23. **Comments from the referee:** Page 4, Line 13: Suggest you make this a “displayed” equation rather than an “inline” one. Also, conventionally vectors are in lower case. If using LaTeX suggest `GROMOS` (`amsmath.sty`) rather than *GROMOS*, it give more suitable letter spacing (similarly for MLS).

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 4, Line 13: The smoothed profile of MLS adjusted to the vertical resolution of GROMOS is expressed as:

$$\mathbf{x}_{\text{MLS,low}} = \mathbf{x}_{\text{a,GROMOS}} + \mathbf{AVK}_{\text{GROMOS}} \cdot (\mathbf{x}_{\text{MLS,high}} - \mathbf{x}_{\text{a,GROMOS}}) \quad (10)$$

being  $\mathbf{AVK}_{\text{GROMOS}}$  is the averaging kernel matrix of GROMOS,  $\mathbf{x}_{\text{MLS,high}}$  is the measured Aura/MLS profile and  $\mathbf{x}_{\text{a,GROMOS}}$  is the a priori profile ...

24. **Comments from the referee:** Page 4, Line 15: Surely Tsou is not the first such reference. Cite others, or at least put “e.g.,” in front.

**Author’s response:**

No comments.

**Author’s changes in the manuscript:** Page 4, Line 15: by e.g. Tsou et al. (1995).

25. **Comments from the referee:** Page 4, line 19: More major point here.  $8^\circ/800$  km is a very large coincidence window, particularly given the  $\sim 165$  km along track spacing for MLS measurements. While you might need this on some days, when GROMOS falls in the gaps between the MLS orbits, on other days you’ll get  $\sim 5$  coincident observations. However, you do not tell us what you do in such circumstances. Do you compare your one GROMOS profile to all five? Do you pick the closest one? Do you average the five profiles together to give one comparison? What are the impacts of your choice on the subsequent analyses? More detail is needed here if readers are to be able to correctly interpret the results that follow.

**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  $1^\circ$  in latitude and  $8^\circ$  in longitude. Then, I have one profile of MLS to compare to one profile of GROMOS every time the temporal and spatial criteria is fulfilled. We define as nighttime (daytime) value the average between the values recorded within 2 hours around midnight (noon).

**Author’s changes in the manuscript:** Page 4, line 17: 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  $1^\circ$  in latitude and  $8^\circ$  in longitude.

26. **Comments from the referee:** Page 4, line 30: I’m a little bit wary of using the term absolute difference, more particularly in the caption for Figure 2, where you use the term “mean absolute difference”. It could be taken to mean the mean of the unsigned difference,  $|a-b|$ . Perhaps simply say “mixing ratio difference”?

**Author's response:**

We agree on the referee's comment.

**Author's changes in the manuscript:** We have changed mean absolute difference for [mean VMR difference](#) everywhere.

27. **Comments from the referee:** Page 5, lines 2 and 3 (counting from -2): At face value, the 30-day smoothing and 4-day filtering appear to be contradictory. If the 30 data points are for 30 days worth of observations, then surely such a smoothing is going to filter far more aggressively than 4 days? Are there more than 30 points per day? Is this related to the issue of having more multiple MLS matches to a single GROMOS measurement? If so, this needs to be made much clearer. Plus, the impact of this smoothing is going to vary quite significantly depending on how many points there are on a given day. Why not simply smooth on a daily rather than a point-by-point basis (average of all differences within an n-day window)? Again, all this needs to be much more clearly described.

**Author's response:**

As we have changed the spatial criteria of coincidence the number of coincident profiles has changed as well. Therefore, now we have performed moving average over 7 points which corresponds to around 1 week. Performing a daily smoothing in the time series will produce noisy and unclear Figures and hence difficulties to interpret them.

**Author's changes in the manuscript:** Page 5, lines 2 and 3: ~~Short temporal fluctuations (periods  $< 4$  days) are suppressed by a moving average over 30 data points of both time series.~~ All time series have been smoothed by a moving average over 7 points ( $\sim 1$  week).

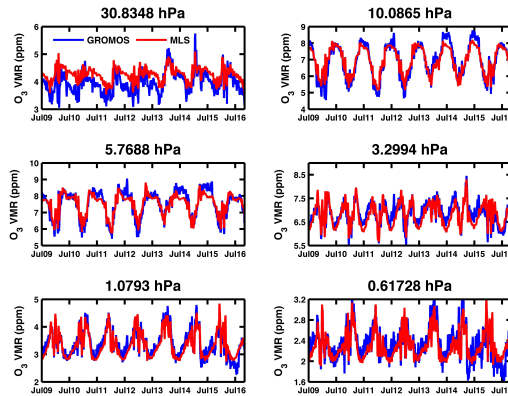


Figure 4: Time series of averaged daytime and nighttime  $O_3$  VMR measurements of GROMOS (blue line) and MLS (red line) for the period from July 2009 to November 2016 at different pressure levels. An averaging kernel smoothing has been applied to the series of the MLS measurements coincident in time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

28. **Comments from the referee:** Page 5, line 8: “almost perfect” is very much in the eye of the beholder, and in my eye your scatter plots are far from it. To me “almost perfect” is at the  $> 0.999$  level of correlation, where the points are all but indistinguishable from the 1:1 line, with perhaps just one or two strays. I suggest you use more measured language.

**Author’s response:**

No comment.

**Author’s changes in the manuscript:** Page 5, line 8: ~~An almost perfect agreement~~  
...

29. **Comments from the referee:** Page 5, line 9: Odd way to phrase it, simply say that the black line is close to the green one to one line.

**Author’s response:**

We agree with the referee.

**Author’s changes in the manuscript:** Page 5, line 8–9: ~~The black lines, linear regression lines of the observations, are close to the green one to one lines,  $O_3(\text{MLS})=O_3(\text{GROMOS})$ .~~

30. **Comments from the referee:** Page 5, line 21: “variation is also expected”

**Author’s response:**

Thanks for spotting. We have corrected this.

**Author’s changes in the manuscript:** Page 5, line 21: ... therefore an annual variation is ~~also~~ expected

31. **Comments from the referee:** Page 6, lines -2 to 2: As discussed above, more discussion is needed here. Some more investigation is needed as to why the amplitudes of the cycles are so different. You don’t even tell us if we should be surprised by this level of disagreement. Note that the MLS averaging kernels imply not insignificant vertical smoothing at these altitudes for this instrument too. When taken in conjunction with the possible latitudinal gradient, are there plausible reasons to explain the differences based on sampling etc. alone, or is the only feasible explanation some instrumental/calibration difference? If nothing else, raise these questions and identify a route to answering them. Could the diurnal cycle in temperature (and thus the pressure/height relationship) play any role in this (from a measurement characteristics point of view rather than an atmospheric science one)? This manuscript would greatly benefit from an analysis, or at least an identification, of all the potential factors involved.

**Author’s response:**

According to Sonnemann et al. (2007), *the MMM is an effect occurring at high latitudes close to the polar night terminator around 72 km altitude during nighttime in the winter half of the year and extends into middle latitudes with decreasing amplitude*. The observed sharp decrease of the amplitude of the MMM of ozone is due to the strong latitudinal gradient between high and middle latitudes. In fact, it is surprising that we can observe the effect of MMM at our latitude. Therefore, the difference in latitude between Lindau and Bern may have such impact in the amplitudes of the annual variability of mesospheric ozone due to the MMM. However it could also be due to some other effects like for example, differences in

the retrieval algorithms between Bern and Lindau, different instruments used to perform the measurements, different calculation methods...

**Author's changes in the manuscript:** Page 6, line 4: Nevertheless, our results are the expected since this maximum of mesospheric ozone during nighttime in winter is related to the middle mesospheric maximum of ozone (MMM) and according to Sonnemann et al. (2007) its effect extends into midlatitudes with decreasing amplitude.

32. **Comments from the referee:** Page 6, lines 13-15: This discussion is unclear, at least to me. If the orange points are smoothed by 10 points, is that 10 days? How does this number related to the  $\sim 7$  years between 2009 and 2016. I don't get how the 10-point and 30-point smoothings are related.

**Author's response:**

We have repeated the comparison by changing the spatial criteria of coincidence and now the number of coincident profiles has changed. In the first panel of Figure 7 (former Figure 6) the moving average is over 30 points, roughly 1 month and in the second panel we used a moving average over 7 points which corresponds to around 1 week. The purpose of the smoothing is to help the interpretation of the results.

**Author's changes in the manuscript:** Page 5, line 30: All time series displayed in both panels of Figure 6 have been smoothed in time by a moving average over 15 data points ( $\sim 1$  week).

Page 6, lines 13-15: ...under assessment. Both time series were smoothed in time by a moving average over 30 points ( $\sim 1$  month). ... 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. ~~The Aura/MLS and the GROMOS series depicted in Figure 5 and Figure 6 have been smoothed in time by a moving average over 30 data points.~~

33. **Comments from the referee:** Figures: In general, all the figures use overly heavy line thicknesses. While it may be OK for the lines themselves (though rather on the heavy side), the line width used is far to heavy for the axes. Also the font should be slightly ( $\sim 20-50\%$ ) larger, and perhaps not bold, for greater clarity. Figure 2: Suggest "mean absolute difference"  $\rightarrow$  "mean mixing ratio difference". Also, how is 'its uncertainty' (last line) defined? Do you mean standard deviation?

**Author's response:**

We have calculated the mean relative difference profile and the VMR difference profile separating daytime and nighttime values, accordingly Figure 3 (former Figure 2) has changed.

**Author's changes in the manuscript:** Figure 3

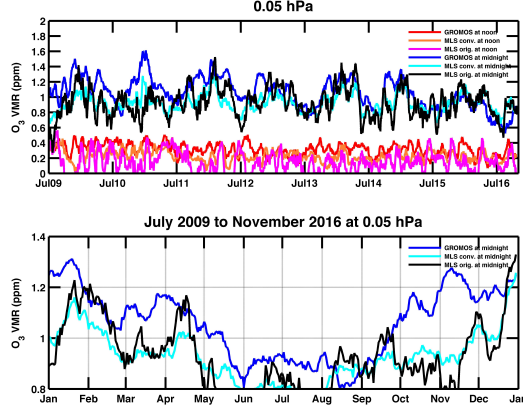


Figure 6: The first panel shows the diurnal variation of O<sub>3</sub> VMR measured at noon (GROMOS in red, MLS convolved in orange and MLS original in magenta) and at midnight (GROMOS in blue, MLS convolved in cyan and MLS original in black) at 0.05 hPa (70 km) and the second panel shows its evolution throughout the year averaged for the time interval under assessment (July 2009–November 2016). All time series are smoothed in time by a moving average over 15 points (1 week)

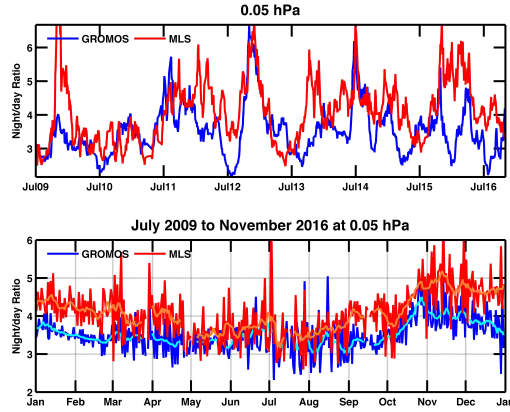


Figure 7: The first panel displays the night-to-day ratio (NDR) of GROMOS (blue line) and MLS (red line) at 0.05 hPa (70 km) for the time period from July 2009 to November 2016 and the second panel shows its evolution throughout the year averaged for this time period. The time series presented in the top panel are smoothed in time by a moving average over 30 data points (1 month) and the orange line (MLS) and the cyan line (GROMOS) shown in the second panel are averaged over 7 data points (1 week)

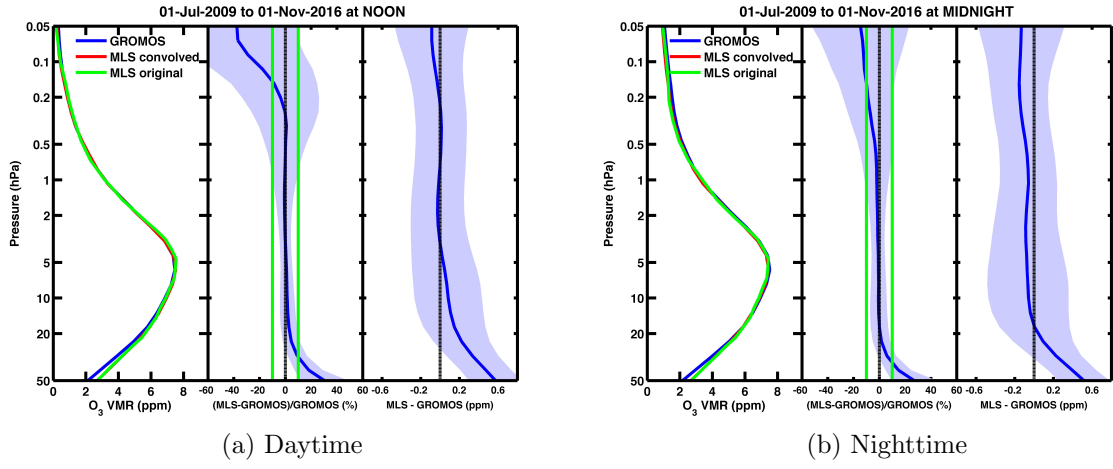


Figure 3: Mean ozone profiles recorded by GROMOS (blue line), MLS convolved (red line) and MLS original (green line) for the time interval between July 2009 and November 2016 are shown in the left panels of both daytime and nighttime Figures. The blue area (GROMOS) and the red area (MLS) are the standard 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  $\pm 10\%$  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



# Comparison of ozone profiles and influences from the tertiary ozone maximum in the night-to-day ratio above Switzerland

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**Abstract.** Stratospheric and middle mesospheric ozone profiles have been continually measured by the GROMOS (GROUND-based Millimeter-wave Ozone Spectrometer) microwave radiometer since 1994 above Bern, Switzerland (46.95°N, 7.44°E, 577 m). GROMOS is part of the Network for the Detection of Atmospheric Composition Change (NDACC). A new version ~~for the retrieval of ozone profiles of the~~ **ozone profile retrievals** has been developed with the aim to improve the altitude range of retrieval profiles. GROMOS profiles from this new retrieval version have been compared to coincident ozone profiles obtained by the satellite limb sounder Aura ~~/MLS~~. The study covers the stratosphere and middle mesosphere from 50 to 0.05 hPa (from 21 to 70 km) and extends over the period from July 2009 to November 2016, which results in more than ~~3500~~ **2800** coincident profiles available for the comparison. ~~GROMOS~~ **On average, GROMOS and MLS comparisons show agreement generally over 20% in the lower stratosphere and within 2% in the middle** and ~~Aura/MLS profiles agree within 3% for the altitude range from 25 to 55 km, with standard deviations of the mean relative differences around 5% from 30 to 40 km and tending to 10% towards the lower and upper stratosphere. Above the stratosphere, the mean relative differences and its standard deviations are increasing with altitude up to 50% at 70 km for both daytime and nighttime, whereas in the mesosphere the mean relative difference is below 40% at daytime and below 15% at nighttime.~~ In addition, we have observed the annual variation of nighttime ozone in the middle mesosphere, at 0.05 hPa (70 km), characterised by the enhancement of ozone during wintertime for both ground-based and space-based measurements. This behaviour is ~~explained by related to~~ **the middle mesospheric maximum of ozone (MMM). 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.**

## 1 Introduction

Passive millimeter wave radiometry is a well-established technique to monitor atmospheric constituents by detecting the radiation emitted by the rotational transitions of the molecules. It makes use of the spectral properties of the atmospheric species in order to derive information about ~~its~~ **their** distribution in the atmosphere. The main advantages of this technique are ~~the~~ **its** independence of solar irradiation and its insensitivity to weather conditions and aerosols. Additionally it offers a good temporal resolution of 1 hour. Measurements of ozone performed by this technique have been indispensable in monitoring changes in the ozone layer and improving the comprehension of the processes that control ozone abundances. ~~The ozonemolecule~~ **(e.g. Steinbrecht et al., 2009). Stratospheric ozone**, in spite of its small abundance ~~in the atmosphere, plays vital~~

~~roles. Stratospheric ozone plays~~, plays a beneficial role by absorbing most of the biologically harmful ultraviolet sunlight. The absorption of UV radiation by ozone creates a source of heat, therefore ozone plays a key role in the temperature structure of the Earth's atmosphere. Changes in the stratospheric ozone concentration alter the radiative balance of the atmosphere, the atmospheric composition and the dynamics of the atmosphere. ~~Thus, continuous~~ Continuous long-term monitoring of ozone is essential for the detection of long-term trends of the stratospheric ozone layer (e.g. WMO, 2014). The ground-based ozone radiometer GROMOS (GROUND-based Millimeter-wave Ozone Spectrometer) is part of the Network for the Detection of Atmospheric Composition Change (NDACC). In order to satisfy the requirements of accuracy and stability the validation of instruments is necessary. There have been a number of comparisons in the past, showing that GROMOS is a reliable tool to measure stratospheric and lower mesospheric ozone (WMO, 2014; Studer et al., 2013; van Gijssels et al., 2010; Keckhut et al., 2010; Dumitru et al., 2006).

This manuscript presents a comparison between the data from the ground-based instrument GROMOS and the space-based instrument ~~Aura~~/MLS for the time interval from July 2009 to November 2016 covering the stratosphere and the middle mesosphere, which corresponds to the altitude range from 20 to 70 km (50 to 0.05 hPa). ~~Furthermore, we have~~ We have also performed an analysis of the diurnal variation and ~~the amplitude of the diurnal variation~~ its amplitude (night-to-day ratio) of middle mesospheric ozone, at 0.05 hPa (70 km). The diurnal variation of ozone in the lower and middle mesosphere is observed as an increase in ozone after sunset and a decrease after sunrise. ~~Atomic oxygen densities are comparable to, and even greater than those of ozone, so that the recombination reaction explains the daily cycle of ozone in the mesosphere (Brasseur and Solomon, 2005). Moreover~~ Daytime production of atomic oxygen by photolysis of ozone (Reaction R7) and photolysis of molecular oxygen (Reaction R5) results in nighttime ozone production by recombination of atomic and molecular oxygen (Reaction R6) (Brasseur and Solomon, 2005). In addition, we observe the annual variation of the nighttime mesospheric ozone with a maximum in wintertime and a minimum in summertime. This maximum of mesospheric ozone during nighttime in winter is related to the middle mesospheric maximum of ozone (MMM) (e.g., Sonnemann et al., 2007; Hartogh et al., 2004) also known as the tertiary ozone maximum (e.g., Sofieva et al., 2009; Degenstein et al., 2005; Marsh et al., 2001). Sonnemann et al. (2007) reported that the MMM is ~~an effect occurring a~~ a phenomenon that occurs at high latitudes close to the polar night terminator around 72 km altitude during nighttime in winter and extends into middle latitudes with decreasing amplitude. 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 R1) is the primary source of odd-hydrogen, reduced UV radiation results in less odd-hydrogen. The ~~shortage~~ lack of odd-hydrogen needed for the catalytic depletion of odd-oxygen ~~, and there is no decrease in the production of odd-oxygen, results in~~ (Reactions R2, R3 and R4), in conjunction with an unchanged rate of odd oxygen production (Reaction R5), leads to an increase in odd-oxygen. This results in higher ozone concentration because atomic oxygen recombination (Reaction R6) 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 R7) rate with increasing solar zenith angle.

~~This publication presents a new comparison between a ground-based instrument (GROMOS) and a space-based instrument~~

~~(Aura/MLS).~~



5



15 The next section describes briefly both instruments and measurement techniques. The results of the comparison are shown in Section 3. Section 4 analyses the night-to-day variability and provides a short discussion. ~~And finally,~~ and the conclusions are summarised in Section 5.

## 2 Instruments and measurement techniques

### 2.1 The ground-based microwave radiometer GROMOS

20 This study is based on stratospheric and mesospheric ozone volume mixing ratio (VMR) profiles observed by GROMOS. The ground-based millimeter wave ozone spectrometer has been operating in Bern, Switzerland (46.95°N, 7.44°E, 577 m) since November 1994 in the framework of the Network for the Detection of Atmospheric Composition Change (NDACC). The instrument measures the thermal microwave emission of the pressure broadened rotational transition of ozone at 142.175 GHz. The vertical distribution of ozone VMR can be retrieved from the measured spectral line since it contains information on the  
25 altitude distribution of the emitting molecule due to the pressure broadening. The retrieval procedure is performed through the

Atmospheric Radiative Transfer Simulator (ARTS2) (Eriksson et al., 2011) which is used as a forward model to simulate the atmospheric radiative transfer in a modelled atmosphere and so calculate the ozone spectrum of this modelled atmosphere. ~~A priori information is required in the inversion process and~~ The a priori profile of O<sub>3</sub> VMR required for the retrieval is taken from a monthly varying climatology from ECMWF reanalysis until available (70 km) and extended above by an Aura/MLS climatology (2004 to ~~2010~~)~~above (Studer et al., 2013)~~2011). The line shape used in the retrieval is the representation of the Voigt line profile from Kuntz (1997). Spectroscopic parameters to calculate the ozone absorption coefficients were taken from the JPL catalogue (Pickett et al., 1998) and the HITRAN spectroscopic database (Rothman et al., 1998). The atmospheric temperature and pressure profiles are taken from the 6 hourly of the European Centre for Medium-Range Weather Forecast (ECMWF) operational analysis data and are extended above 80 km by monthly mean temperatures of the CIRA-86 Atmosphere Model (Fleming et al., 1990). The accompanying Matlab package Qpack2 (Eriksson et al., 2005) compares the modelled spectrum with the measured spectrum and derives the best estimate of the vertical profile by using the optimal estimation method (OEM) (Rodgers, 1976). The OEM also provides a characterisation and formal analysis of the uncertainties (Rodgers, 1990).~~We have recently-~~

Recently, we have developed a new retrieval version ~~, version 150. The difference~~ (version 150) with the aim to optimise the averaging kernels. The differences with the former version ~~, (version 2021, is that the uncertainty of the a priori is kept constant with the altitude, thus optimizing the averaging kernels and improving the measurement response)~~ 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 ~~, The~~ 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~~and for~~. For both retrieval versions the off-diagonal elements ~~the values decay exponentially~~ of the a priori covariance matrix exponentially decrease with a correlation length of 3 km.~~This new retrieval version is performed with a~~ 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~~incorporated in the covariance matrix of~~  $\Delta T'_b = 0.5 + \frac{\Delta T_b}{e^{-\tau}}$ ,  $\Delta T_b = \frac{T_{rec}}{\sqrt{B \cdot t}}$ :

$$\Delta T'_b = 0.5 + \frac{\Delta T_b}{e^{-\tau}} \quad (1)$$

the error of the measured brightness temperature,  $\Delta T_b$ , is given by the radiometer ~~noise equation. In addition, a~~ equation:

$$\Delta T_b = \frac{T_b + T_{rec}}{\sqrt{\Delta f \cdot t_{int}}} \quad (2)$$

The radiometer equation gives the resolution of the radiation measured, which is determined by the bandwidth of the individual spectrometer channels ( $\Delta f$ ), 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.~~The inversion of the spectra is performed in case of transmission factor larger than~~, due to spectroscopic errors and the water vapour continuum. The error of the brightness temperature ( $\Delta T_b$ ) 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.

In Figure 1 is displayed a comparison between version 2021 and version 150 of ozone profiles measured by GROMOS for the time interval from July 2009 to November 2016. In the left panel are represented the mean ozone profiles retrieved by version 2021, in red, and by version 150, in blue. The standard deviation of the ozone VMR are shown by the coloured areas, red in case of v2021 and blue for v150. The mean relative differences (blue line in the middle panel) and the volume mixing ratio (VMR) differences (blue line in the right panel) are ranging from 30% (0.5 ppm) in the lowermost stratosphere to within 5% (0.2 ppm) in the middle stratosphere, and increasing to 10% (0.4 ppm) in the upper stratosphere and up to 18% (0.05 ppm) at 0.05 hPa (70 km). The blue areas in the middle and right panels represent the standard deviation of the differences, relative differences and VMR differences, respectively. We can conclude from Figure 1 that the differences between version 2021 and version 150 appear in the lower stratosphere and in the mesosphere.

Figure 2 displays an example of a GROMOS retrieval accomplished by the new retrieval version 150. The left panel show the a priori (green line) and the retrieved profile (blue line) measured in July 2013 at noon. In the middle panel are represented the averaging kernels (AVK) and the area of the averaging kernels (measurement response). The AVKs are multiplied by 4 in order to be displayed along with the measurement response (red line). The AVK-lines are grey except for some selected altitudes, which are shown in different colours to make the Figure 2 easier to interpret. 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). It is considered a reliable altitude range of the retrieval when the true state dominates over the a priori information, i.e. where the measurement response is larger than 0.8 (an a priori contribution smaller than 20%). The measurement response shown in Figure 2 is around 1 from 18 to 70 km. The magenta line in the right panel shows the altitude peak of the corresponding kernels and proves that the AVK peak at its nominal altitude for the considered altitude range. And finally, the cyan line displays the vertical resolution which is quantified by the full width at half maximum of the averaging kernels. The vertical resolution of this new retrieval version of GROMOS lies from 10 to 15 km below 40 km altitude and from 15 to 20 km below 70 km altitude. 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.

For technical details, measurement principle of the instrument, see for example Moreira et al. (2015) and Peter (1997) and references included therein.

## 2.2 The Aura ~~microwave limb sounder~~ Microwave Limb Sounder

The Microwave Limb Sounder (MLS) is a passive microwave limb-sounding radiometer onboard the NASA Aura satellite. The Aura spacecraft was launched in 2004 into a near polar, sun-synchronous orbit with a period of approximately 100 minutes. The satellite overpasses ~~our~~ the GROMOS measurement location (at northern ~~mid-latitudes~~ 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).

## 3 Comparison of ~~Aura~~/MLS and GROMOS

- 10 The vertical resolution of the ~~Aura~~/MLS is within 3.5 km in the stratosphere and up to 5.5 km in the middle mesosphere. Therefore in order to compare ozone profiles of GROMOS with ~~Aura~~/MLS, an averaging kernel smoothing is applied to the ozone profiles of the satellite data. The smoothed profile of ~~Aura~~/MLS adjusted to the vertical resolution of GROMOS is expressed as:  ~~$\mathbf{x}_{MLS,low} = \mathbf{x}_{a,GROMOS} + \mathbf{A}_{GROMOS} \cdot (\mathbf{x}_{MLS,high} - \mathbf{x}_{a,GROMOS}) \cdot \mathbf{A}_{GROMOS}^{-1}$~~

$$\mathbf{x}_{MLS,low} = \mathbf{x}_{a,GROMOS} + \mathbf{AVK}_{GROMOS} \cdot (\mathbf{x}_{MLS,high} - \mathbf{x}_{a,GROMOS}) \quad (3)$$

- 15 being  $\mathbf{AVK}_{GROMOS}$  is the averaging kernel matrix of GROMOS,  ~~$\mathbf{x}_{MLS,high}$~~   $\mathbf{x}_{MLS,high}$  is the measured ~~Aura~~/MLS profile and  ~~$\mathbf{x}_{a,GROMOS}$~~   $\mathbf{x}_{a,GROMOS}$  is the a priori profile used during the retrieval procedure of GROMOS. The application of averaging kernel smoothing for the comparison of profiles with different altitude resolutions has been introduced and described by e.g. Tsou et al. (1995).

- Every profile utilised in the comparison between ~~Aura~~/MLS and GROMOS should be coincident in time and space. The requirement of time coincidence is satisfied when both measurements are within 1 hour in time. 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 ~~81°~~ in latitude and ~~longitude ( $d < 800$  km)~~ 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 ~~with the compliance of the~~ to the spatial and temporal criteria, more than ~~3500-2800~~ coincident profiles are available for the comparison. ~~The~~ Figure 3a and Figure 3b show the mean ozone profiles ~~and its standard deviations~~ of the collocated and coincident measurements of ~~both instruments are shown in the right panel of Figure ??~~, GROMOS ~~blue line and blue area and~~  $\mathbf{x}_{GROMOS}$  (blue line), MLS convolved (red line) and MLS original (green line) at daytime and nighttime, respectively. The relative difference profile in percent given by  ~~$(\mathbf{x}_{MLS,low} - \mathbf{x}_{GROMOS}) / \mathbf{x}_{GROMOS}$~~   $(\mathbf{x}_{MLS,low} - \mathbf{x}_{GROMOS}) / \mathbf{x}_{GROMOS}$  is displayed in the middle panel of ~~Figure ??~~ both Figure 3a and Figure 3b along with the standard deviation of the differences (blue area). The green line delimits the  $\pm 10\%$  area. The mean ~~relative differences between GROMOS and Aura/MLS during this time period are within 3% between~~ 30 profile of the VMR differences is shown in the right panel of both Figure 3. The mean relative differences and ~~0.35~~ 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 (25 to 55 km) and progressively increasing to 50% at 70 km). We conclude from Figure 3 that during nighttime GROMOS measures more O<sub>3</sub> VMR (ppm) than MLS except for the lower stratosphere, where MLS measures more O<sub>3</sub> VMR (ppm) than GROMOS, both at daytime and nighttime. Nevertheless in the mesosphere GROMOS measures more O<sub>3</sub> VMR (ppm) than MLS, both at daytime and nighttime. The standard deviation of the mean relative differences are around 5% for the altitude range from 30 to 40 km and tending to roughly 10% toward the lower and upper stratosphere, and up to 50% in the middle mesosphere. The mean profile of the absolute differences is shown in the right panel of Figure ??.

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; Hoeke et al., 2007; Dumitru et al., 2006; Calisesi et al., 2005).

For an overview of the 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 Aura/MLS (red line) are displayed in Figure ??-4 for different pressure levels. Short temporal fluctuations (periods < 4 days) are suppressed. All time series have been smoothed by a moving average over 30 data points of both time series 7 data points (~ 1 week). The agreement between both ground-based and satellite-based instruments depends upon altitude and time. A negative deviation of GROMOS series with respect to Aura/MLS occurs in the lower stratosphere. On the other hand, a positive deviation of GROMOS with respect to Aura/MLS is observed in the middle stratosphere for summers 2011, 2012, 2014 and 2015. Further, we notice a negative bias of GROMOS during summer 2016 from the stratopause towards the mesosphere. In Figure ??-5 are shown the scatter plots of GROMOS and Aura/averaged daytime and nighttime O<sub>3</sub> VMR measurements of GROMOS and MLS at the same pressure levels as Figure ??. An almost perfect agreement between the 4. The black lines, linear regression lines of the observations (black lines) and the identity of both data sets (, are close to the green one to one lines, O<sub>3</sub>(Aura/MLS)=O<sub>3</sub>(GROMOS)–green lines) is observed, except for the lower stratosphere where we find the negative deviation of GROMOS with respect to Aura/MLS. The linear fit deviates from the identity where there is less ozone in the case of GROMOS during winter in the middle to upper stratosphere as we also observe in Figure ??4, along with the positive deviation of GROMOS with respect to Aura/MLS during some summers. The calculation of the correlation coefficients also reveals good agreement with  $r > 0.75$  for all altitudes levels except for the altitudes altitude above 50 km where  $r$  is around 0.55. 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.

#### 4 Analysis of the night-to-day ratio

The diurnal variation of mesospheric ozone is characterised by an increase at the beginning of the nighttime and by a decrease after sunrise. This effect is explained by the recombination of atomic and molecular oxygen (e.g., Brasseur and Solomon,



2005). Because the ozone distribution in the mesosphere is mainly controlled by photochemistry, it depends strongly on the solar zenith angle (Nagahama et al., 2003), therefore an annual variation is also expected in mesospheric ozone. Figure ??-6 shows both the diurnal variation of mesospheric ozone and the annual variation of nighttime mesospheric ozone. To analyse the variability of mesospheric ozone we have used ozone VMR measurements coincident in space and in time recorded by GROMOS and by ~~Aura/MLS~~ for the time period from July 2009 to November 2016. The first panel of Figure ??-6 displays the O<sub>3</sub> VMR measured at noon (GROMOS in red, ~~Aura/MLS in orange~~MLS convolved in orange and MLS original in magenta) and at midnight (GROMOS in blue, ~~Aura/MLS in cyan~~MLS convolved in cyan and MLS original in black) at 0.05 hPa (70 km) for the already mentioned time period. The original MLS data, i.e. not weighted with GROMOS AVKs, is shown in order to provide an insight of the observability of the effect of MMM at northern midlatitudes by GROMOS. We define as midnight (noon) value the average between the values recorded within 2 hours around midnight (noon). The daytime mesospheric ozone does not show any distinct annual variation. On the other hand, the annual variation of nighttime mesospheric ozone is characterised by a maximum in wintertime and a minimum in summertime. The second panel of Figure ??-6 shows the evolution of the midnight mesospheric ozone throughout the year averaged for the time interval from July 2009 to November 2016. All time series displayed in both panels of Figure 6 have been smoothed in time by a moving average over 15 data points (~1 week). A closer observation shows that the annual variation of the nighttime ozone exhibits a primary maximum over wintertime and a secondary maximum around springtime. Our results on the annual variation of mesospheric ozone at Bern (Switzerland, 46.95°N, 7.44°E) are in agreement with the ones observed at Lindau (~~Gernamy~~Germany, 51.66°N, 10.13°E) by Sonnemann et al. (2007). ~~Differences occur~~Disagreements appear in the amplitudes where the maximum values of GROMOS and MLS original do not exceed 1.5 ppm, 1.2 ppm in the case of ~~Aura/MLS~~MLS convolved, whereas at Lindau the maximum values exceed 3 ppm at 70 km. ~~This~~Nevertheless, our results are expected since this maximum of mesospheric ozone during nighttime in winter is related to the middle mesospheric maximum of ozone (MMM) (e.g., Sonnemann et al., 2007; Hartogh et al., 2004) also known as the tertiary ozone maximum (e.g., Sofieva et al., 2009; Degenstein et al., 2005; Marsh et al., 2001). During winter, the photodissociation rate of water is reduced at high latitudes which leads to a decrease of catalytic ozone depletion by odd hydrogen and according to Sonnemann et al. (2007) its effect extends into midlatitudes with decreasing amplitude. Furthermore, we have analysed the amplitude of the diurnal variation, the night-to-day-ratio (NDR). The NDR is closely related to the MMM, but it is also related to the change of the diurnal variation from winter to summer (Sonnemann et al., 2007). The annual variation of the NDR is modulated by oscillations of planetary time scale (Sonnemann et al., 2007). Sofieva et al. (2009) reported that during a sudden stratospheric warming event the tertiary ozone maximum can decrease significantly or can even be completely destroyed. Hocke (2017) has shown the loss of the tertiary ozone layer in the polar mesosphere due to the solar proton event in November 2004.

The first panel of Figure ??-7 displays the NDR of GROMOS (blue line) and ~~Aura/MLS~~ (red line) at 0.05 hPa (70 km) for the time interval from July 2009 to November 2016 while the second panel shows its evolution throughout the year averaged for the time interval under assessment. Both time series were smoothed in time by a moving average over 30 points (~1 month). The orange line (~~Aura/MLS~~) and the cyan line (GROMOS) depicted in the second panel of Figure ??-7 show a moving average over ~~10 data points~~7 data points (~1 week) with the aim to clarify the understanding of the Figure ??-7. The ~~Aura/MLS~~



and the GROMOS series depicted in Figure ?? and Figure ?? have been smoothed in time by a moving average over 30 data points. Figure 7. Both the ground-based and the satellite-based instruments confirm the expected winter anomaly enhancement of the NDR, also observed at Lindau by Sonnemann et al. (2007), although with smaller the latter data exhibit larger amplitudes. We observe winter-to-summer values of a factor of one to two, whereas at Lindau it is shown, winter-to-summer values of vary by a factor of two to three 2–3 at 70 km (Sonnemann et al., 2007). Thus, despite the definition of the MMM being restricted to high latitudes, we can report its observation with a smaller amplitude at mid-latitudes.

## 5 Conclusions

Stratospheric and middle mesospheric ozone profiles for the period from July 2009 to November 2016 recorded by the ground-based instrument GROMOS and by the spaced-based instrument Aura/space-based instrument MLS were used to perform a comparison and to evaluate the diurnal variability and the amplitude of the diurnal variability its amplitude, night-to-day ratio (NDR). The agreement between measurements coincident in space and time of both data sets is within 3% between 25 to 55 km. At upper altitudes (up to 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 the mean relative ozone differences are within a range of 50% (0.05 hPa). In general terms we can report a good agreement among, we report good agreement between the new retrieval version (v150) of GROMOS and the version 4.2 of Aura/MLS. Further MLS. Furthermore, we observe extensions of the middle mesospheric maximum of ozone (MMM) during winter towards northern mid-latitudes. This effect is smaller in amplitude at mid-latitudes compared to high latitudes. Moreover, the winter anomaly enhancement of nighttime mesospheric ozone is observed by GROMOS and Aura/MLS above Bern.

## 6 Code availability

Routines for data analysis are available upon request by Lorena Moreira.

## 7 Data availability

The data from the GROMOS microwave radiometer is available via <http://ftp.cpc.ncep.noaa.gov/ndacc/station/bern/hdf/mwave>. MLS v4.2 data are available from the NASA Goddard Space Flight Center Earth Sciences Data and Information Services Center (GES DISC), <http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/MLS/index.shtml>.

*Author contributions.* Klemens Hocke performed the retrieval of the GROMOS measurements. Lorena Moreira carried out the data analysis and prepared the manuscript. Niklaus Kämpfer is the principal investigator of the radiometry project. All authors have contributed to the interpretation of the results.

*Competing interests.* All authors declare that there are no conflicts of interest in the current version of the manuscript.

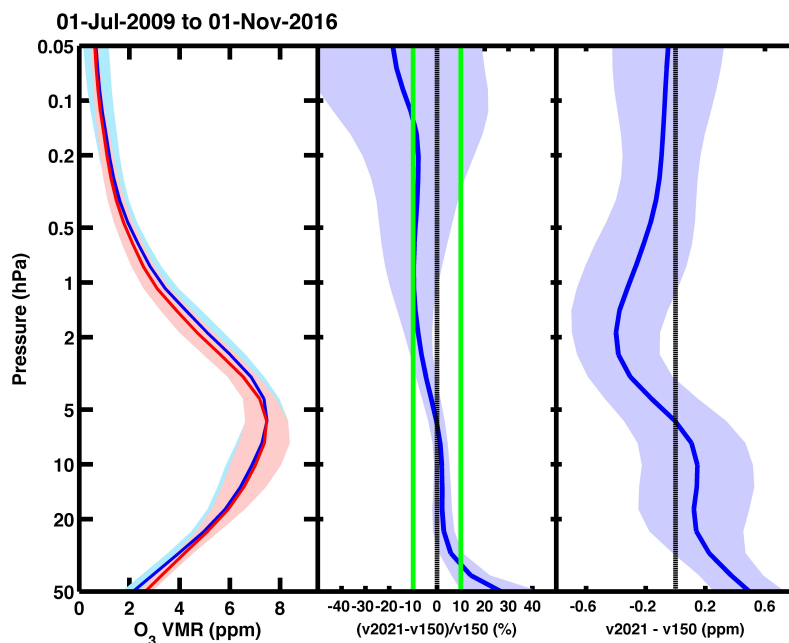
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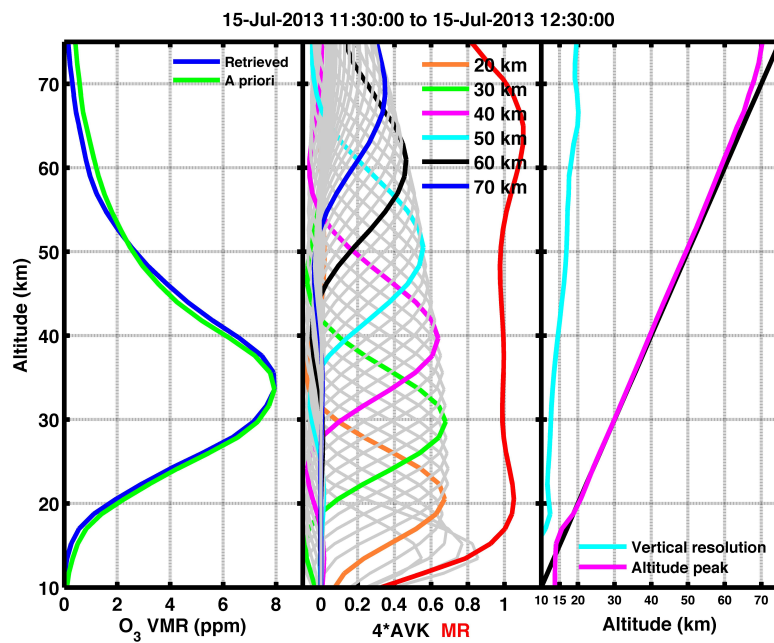
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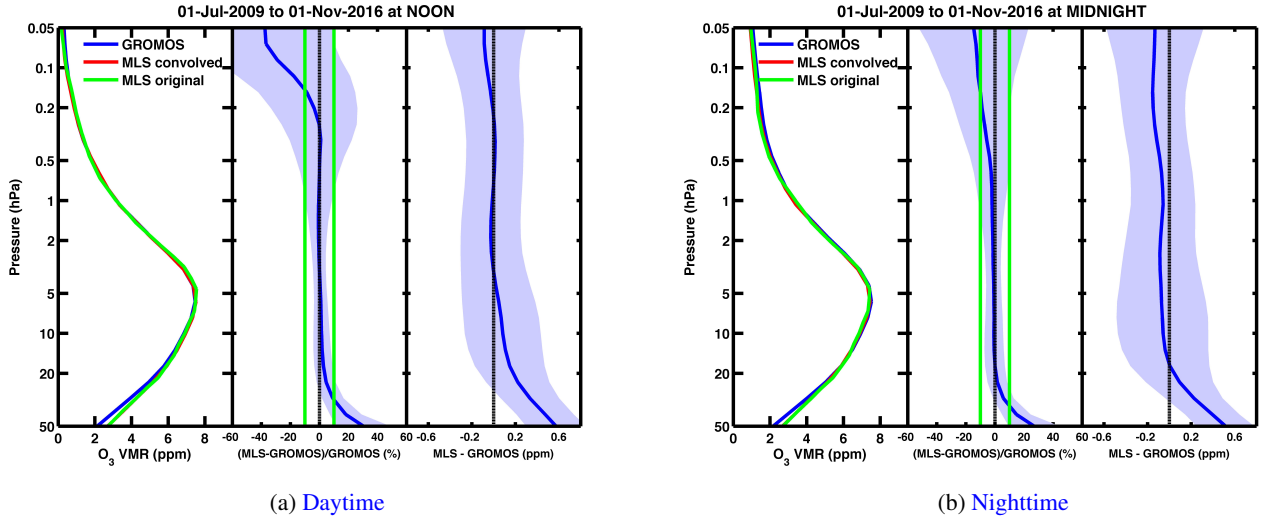


**Figure 1.** Mean ozone profiles retrieved by version 2021 (red line in the left panel) and by version 150 (blue line in the left panel) measured by GROMOS during the period from July 2009 to November 2016. The blue area (v150) and the red area (v2021) are the standard deviations of the ozone VMR. The 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  $\pm 10\%$  area. In the right panel is shown the VMR difference profile along with its standard deviation

Mean ozone profiles recorded by GROMOS (blue line) and by Aura/MLS (red line) for the time interval between July 2009 and November 2016 are shown in the left panel. The blue area (GROMOS) and the red area (Aura/MLS) are the standard deviations of the coincident measurements. In the middle panel is represented the mean relative difference profile between data of both instruments, GROMOS as reference. The blue area represents the standard deviation of the differences. The green line delimits the  $\pm 10\%$  area. The mean absolute difference profile and its uncertainty (blue area) is displayed in the right panel

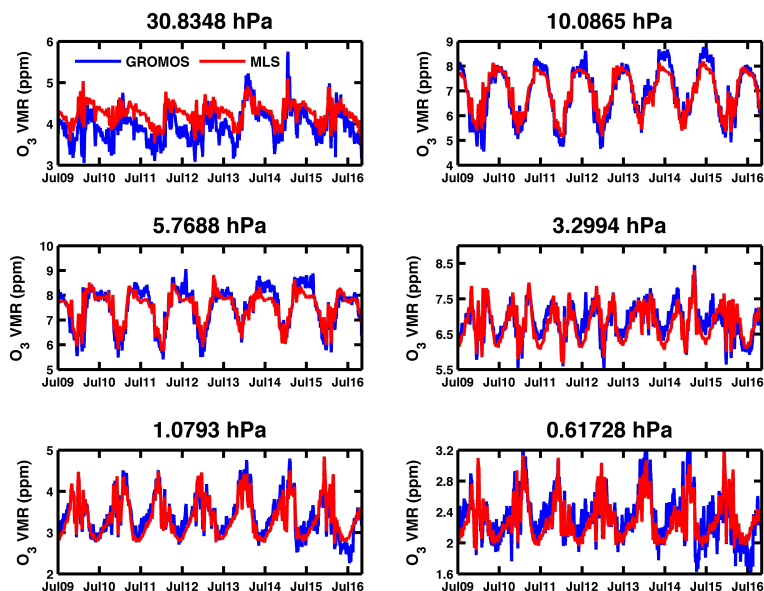


**Figure 2.** Example of an a priori profile and a retrieved ozone profile (green and blue lines in the left panel, respectively), averaging kernels (grey and colour lines in the middle panel), the measurement response (red line in the panel), vertical resolution (cyan line in the right panel) and altitude peak (magenta line in the right panel) of the GROMOS retrieval version 150 for July 15, 2013 with an integration time of 1 hour

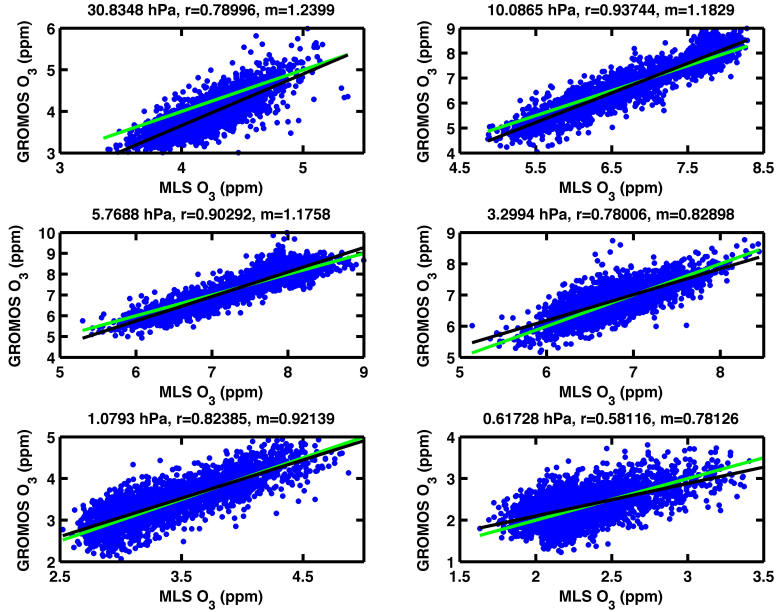


**Figure 3.** Mean ozone profiles recorded by GROMOS (blue line), MLS convolved (red line) and MLS original (green line) for the time interval between July 2009 and November 2016 are shown in the left panels of both daytime and nighttime Figures. The blue area (GROMOS) and the red area (MLS) are the 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  $\pm 10\%$  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

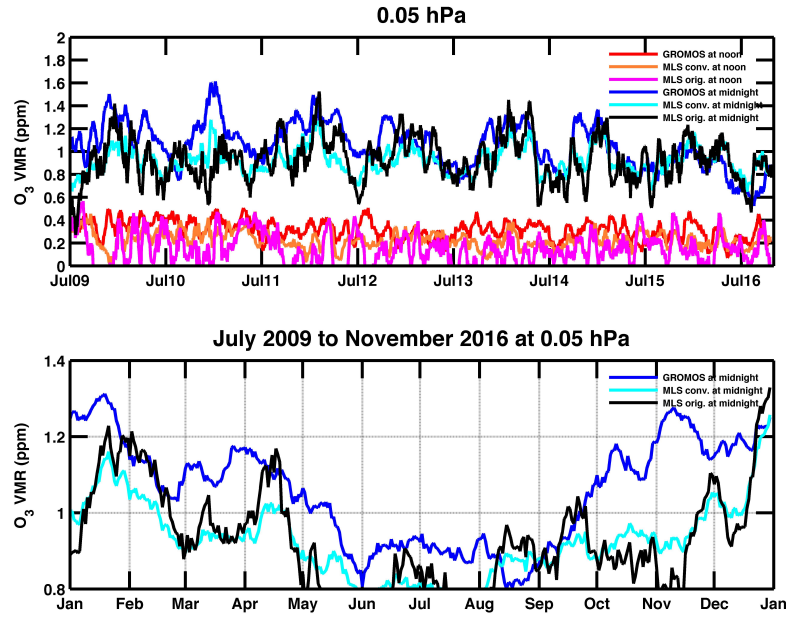




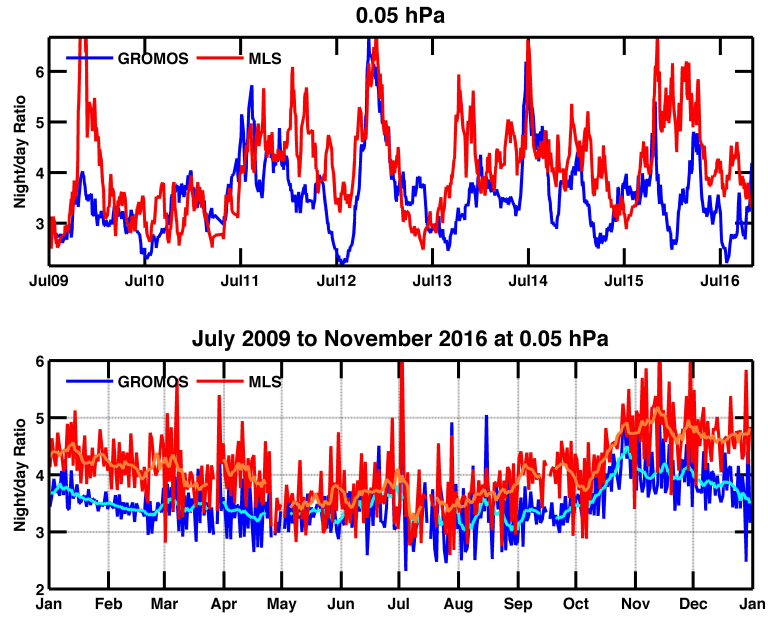
**Figure 4.** Time series of [averaged daytime and nighttime](#)  $O_3$  VMR measurements of GROMOS (blue line) and [Aura](#)/MLS (red line) for the period from July 2009 to November 2016 at different pressure levels. An averaging kernel smoothing has been applied to the series of the [Aura](#)/MLS measurements 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



**Figure 5.** Scatter plots of coincident  $O_3$  VMR measurements of GROMOS and **Aura**/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, and  $m$  the slope of the linear fit. The green line indicates the case of identity,  $O_3(\text{Aura/MLS}) = O_3(\text{GROMOS})$ .  $r$  values are correlation coefficients of the **Aura**/MLS and GROMOS time series



**Figure 6.** The first panel shows the diurnal variation of O<sub>3</sub> VMR measured at noon (GROMOS in red, ~~Aura~~/MLS convolved in orange and MLS original in magenta) and at midnight (GROMOS in blue, ~~Aura~~/MLS convolved in cyan and MLS original in black) at 0.05 hPa (70 km) ~~ans~~ and the second panel shows its evolution throughout the year averaged for the time interval under assessment (July 2009–November 2016). All time series are smoothed in time by a moving average over 15 points (~1 week)



**Figure 7.** The first panel displays the night-to-day ratio (NDR) of GROMOS (blue line) and ~~Aura~~/MLS (red line) at 0.05 hPa (70 km) for the time period from July 2009 to November 2016 and the second panel shows its evolution throughout the year averaged for this time period. ~~All~~The time series presented in the top panel are smoothed in time by a moving average over 30 data points ( $\sim 1$  month) and the orange line (~~Aura~~/MLS) and the cyan line (GROMOS) shown in the second panel are averaged over ~~10~~7 data points ( $\sim 1$  week)