

## ***Interactive comment on “Stratospheric and mesospheric HO<sub>2</sub> observations from the Aura Microwave Limb Sounder” by L. Millán et al.***

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### Response to reviewer 1: Stratospheric and mesospheric HO<sub>2</sub> observations from the Aura Microwave Limb Sounder

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We sincerely thank reviewer 1 for his/her thoughtful comments on the previous draft, we hope this new version is more suitable for publication.

In doing the corrections of all reviewers we added the following 3 major changes:

(1) A paragraph at the beginning of the results sections explains that the averaging kernels were applied to all comparisons:

In this section we compare the offline HO<sub>2</sub> dataset with balloon-borne and other satellite measurements, as well as, with global climate and photochemical model simulations. In making these comparisons, i.e. when showing the absolute or percentage differences between the datasets, the MLS averaging kernels has been applied to properly compare them. Furthermore, when comparing the global climate or the photochemical model simulations, its high vertical resolution has been reduced to the MLS one using a least square fit as described by Livesey et al. (2011, Sect. 1.9). In these comparisons, no altitude extrapolation has been applied to any dataset.

(2) the discussion about the impact of the O<sub>2</sub> and H<sub>2</sub>O cross section was deleted, the

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discussion about the mesospheric discrepancies now reads:

These discrepancies might be due to a variety of reasons, for example: (1) our understanding of middle atmospheric chemistry may not be complete, (2) there might be due to differences between recent solar spectral irradiance (SSI) satellite measurements (Snow et al., 2005; Harder, 2010) and most parameterizations. These SSI measurements display a larger variability in solar UV irradiance which can-not be reconstructed with SSI models, including the model of Lean et al. (2005), used in this SD-WACCM run (Marsh et al., 2013). These SSI measurement-model differences have been proven to affect the HOx photochemistry (Haigh et al., 2010; Merkel et al., 2011; Ermolli et al., 2013); more UV irradiance leads to an enhancement of O<sub>3</sub> photolysis as well as H<sub>2</sub>O photodissociation, which leads to more HOx production through (Reactions R4 to R8). Further, Wang et al. (2013) showed that using a solar forcing derived from these SSI measurements the modeled OH variability agrees much better with observations. Lastly, (3) these discrepancies might be related to the WACCM representation of the mean meridional circulation which has been shown to have some deficiencies (Smith et al., 2011; Smith, 2012), suggesting that the gravity wave parametrization needs to be modified. In addition, Garcia et al. (2014) has shown that adjusting the Prandtl number, used to calculate the diffusivity due to gravity waves, significantly alters the CO<sub>2</sub> SD-WACCM simulations improving its agreement with satellite measurements. Such adjustment should also affect the H<sub>2</sub>O and hence the HOx chemistry.

(3) the photochemical model discussions now reads:

As shown in Fig. 12, in the upper mesosphere (pressures smaller than 0.1 hPa), the Kinetics 1 simulations do not reproduce the magnitude of the measured peak, underestimating it by as much as 60%. On the other hand, Kinetics 2 shows an improvement in the modeling of this peak, reducing the underestimation to less than 40%. These discrepancies coincide with the ones discussed in the previous section strongly suggesting that they are related to the model assumptions rather than to

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measurement errors. As with the SD-WACCM simulations, several factors could be the reason for this discrepancy: it might be due to limitations in our current understanding of middle atmospheric chemistry and/or due to the deficiencies in the model solar spectral irradiance used, in this case Rottman (1982). Also, considering that Kinetics 2 (the run testing the HOx partitioning) represents the measured HO<sub>2</sub> better, these simulations might suggest that, the modeling problems are related to the HOx production and loss balance rather than the HOx partitioning. In the upper stratosphere and lower mesosphere (between 1 and 0.1 hPa) for the most part the photochemical model underpredicts HO<sub>2</sub> by around 20% concurring with the SD-WACCM simulations as well as with previous studies (Sandor et al., 1998; Khosravi et al., 2013) but contradicting the result of the study by Canty et al. (2006).

Below are our responses to the reviewers comments in red.

General comments:

This manuscript presents algorithm and results for a new offline HO<sub>2</sub> retrieval from MLS/Aura limb observations. The main difference between this new retrieval and the MLS standard retrieval is that the retrieval is not applied to individual MLS limb measurements, but to zonally averaged data. The resulting noise reduction allows covering a significantly enhanced altitude range and a wider latitude range. I find the paper in general well written and relatively easy to follow. The paper is in my opinion suited for publication in ACP, but I ask the authors to consider the comments given below. My main criticism concerns two aspects:

a) The model-measurement comparison presented in section 4.4 does not really allow any conclusions to be drawn, as far as I can tell. Therefore, one may question the necessity of this section. In my opinion the section should at least be improved to better describe the assumptions made for the 2 model scenarios (see specific

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comment below) and by adding a more detailed discussion of the implications of the comparisons performed.

This section reinforces the results shown in the previous section (the WACCM section) as well as suggest further that the model discrepancies are real model errors rather than measurement errors. This has been stated in the new manuscript. See below.

b) Figure 6 shows a comparison between MLS offline and FIRS-2 balloon HO<sub>2</sub> observations for the 1 to 10 hPa pressure range. The MLS profile is a daytime/nighttime average, despite the fact that earlier in the paper it was stated that the retrievals between 1 and 10 hPa are affected by systematic biases. For this reason the daytime measurements presented for this pressure range are differences between daytime and nighttime measurements. If there are known biases, a comparison of daytime/nighttime averages to FIRS data does not appear to be a valid comparison.

See below

Specific comments:

Page 22907, line 10: "in the Lyman-alpha and the Schumann-Runge bands"

This phrase implies that the Lyman-alpha signature is also a band, which is not the case.

changed to: in the Lyman–Alpha region and the Schumann–Runge bands

Page 22907, line 22: "a problem known as the HO<sub>x</sub> dilemma" From the following description of differences between observations and model simulations it's not fully clear what the "HO<sub>x</sub> dilemma" is. Is it the low bias of OH measurements compared to model simulations reported by Summers et al. (1997) or is it the general disagreement between models and measurements, with the latter being sometimes higher and sometimes lower compared to the models?

The text was rearranged to make it clear that it was the general disagreement between

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models and measurements. See text

Page 22908, line 2: "lower that the values" – "lower than the values"

Done

Page 22908, line 11: "Furthermore, models have consistently under-predicted the amounts of O<sub>3</sub> at such altitudes, an issue known as the O<sub>3</sub> deficit problem" Some of the references cited are 3 decades old. The most recent one is already 10 years old. I'm wondering, whether more recent studies find better agreement between modelled and observed O<sub>3</sub>?

I added the Siskind (2013) - Comparison of a photochemical model with observations of mesospheric hydroxyl and ozone, reference that still shows the deficit.

Page 22909, line 17: "It covers between 82S and 82N" - "It covers latitudes between 82S and 82N"

Done

Page 22910, line 13: "in (Livesey et al., 2006)" - "in Livesey et al. (2006)"

Done

Page 22910, line 19: "indecipherable" - "indistinguishable" ?

Done

Page 22910, line 21: "with a 10deg latitude typical precision" I'm not entirely sure what you mean here. Probably the typical precision for measurements zonally averaged and binned in 10 deg latitude bins? I suggest stating this more explicitly.

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Done, changed to: typical precisions varying from 0.15 ppbv ( $52 \times 10^6 \text{ cm}^{-3}$ ) at 10 hPa to 3 ppbv ( $5 \times 10^6 \text{ cm}^{-3}$ ) at 0.046 hPa for measurements zonally averaged and binned in a  $10^\circ$  latitude bin.

Page 22911, line 20 “with day-night differences used as a measure of daytime HO<sub>2</sub> for pressures between 10 and 1 hPa where the nighttime values exhibit non-zero values indicative of biases.” I’m wondering, whether this special treatment between 1 and 10 hPa leads to discontinuities at the 1hPa level? It would be good to provide a quantitative estimate on the jump or discontinuity at 1hPa – or an upper threshold. In Figure 2 such a discontinuity is not visible, but this may just be because of the finite width of the vmr bins.

We added in the offline retrieval section: Note that, a visual inspection of the 10o bin monthly average profiles have shown, overall, no signs of a discontinuity at 1 hPa when using this approach.

Page 22913, line 6: “this retrieval” Suggest to replace this by “the retrieval presented in this study”, to avoid confusion with the standard retrieval, which is also mentioned in the previous sentence.

Done

Page 22913, line 22: ““truth” model atmosphere” - ““true” model atmosphere” ?

Done

Page 22914, line 13: “The MLS HO<sub>2</sub> profiles is a 20 deg latitude bin” This statement is certainly not correct, a profile is not a latitude bin. Suggest replacing by, e.g.: “The MLS HO<sub>2</sub> profile corresponds to a 20 deg latitude bin”

Done

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Page 22916, line 4: “within half an hour of the MLS measurements” You mean half an hour in terms of local time, not UT, right? I suggest mentioning this explicitly.

Changed to: Note that only SMILES measurements made within half an hour local time of the MLS measurements were used in this comparison.

Page 22917, line 5: ‘a strong zonal latitudinal gradient’ I don’t quite understand what you mean by ‘zonal latitudinal gradient’. Please clarify.

Deleted: strong zonal latitudinal

Page 22917, same sentence: ‘gradient from the summer pole towards the winter pole’ The “gradient” generally points from low values to high values, i.e. if you speak of the gradient’s direction (and not just the fact that there is a gradient), there’s a gradient from the winter (NH) pole to the summer (SH) pole and not vice versa. I suggest omitting the statement on the direction of the gradient and just state that there is a gradient.

Changed to: As can be seen, both display similar VMR structures with a gradient from the winter pole towards the summer pole.

Page 22917, line 11: ‘zonal latitudinal gradient’ Please change (see comment above)

Deleted: strong zonal latitudinal

Page 22917, line 20: ‘eg.’ - ‘e.g.’

Done

Page 22918, line 23: ‘as well as a strong zonal latitudinal gradient from the summer to

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the winter pole' See comments above

Changed to: as well as a gradient from the winter to the summer pole.

Page 22918, line 26: 'the H available is the one generated at sunlit latitudes, transported at high altitudes poleward, where it descends and reacts with O<sub>2</sub> at night.' I suggestion mentioning explicitly that this applies to the winter, i.e., the northern hemisphere in this case.

The sentence now reads: however in this case, due to the lack of photodissociation of H<sub>2</sub>O, the H available is the one generated at sunlit latitudes (in this case, in the northern hemisphere), transported at high altitudes towards the winter pole where it descends and reacts with O<sub>2</sub> at night (in this case, in the southern hemisphere) (Pickett et al., 2006)

Page 22918, line 27: 'Pickett et al. (2006)' - '(Pickett et al., 2006)'

Done

Page 22919, line 17: 'which adds a constraint to MLS OH to mostly ..' don't understand what this means. What kind of 'constraint' is that. Does it simply mean that you use MLS OH profiles? Please clarify.

expanded to: in addition, constrains the model using MLS OH measurements

Section 4.4: I think this section is the weakest part of the paper, because the implications of the model-measurement comparisons are not clear. If there are no conclusions to be drawn from this comparison, one may question, why section 4.4 is necessary at all. If this section remains in the paper, the implications of the differences between the model runs need to be explained better and in more detail, in my opinion. What exactly do we learn from the fact that the agreement to measured HO<sub>2</sub> is improved if OH is

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also taken from MLS measurements?

We learn that the error is more related to the HO<sub>x</sub> production than the HO<sub>x</sub> partitioning as said in the text. Also, we added when talking about the mesospheric differences: These discrepancies coincide with the ones discussed in the previous section (WACCM section) strongly suggesting that they are related to the model assumptions rather than to measurement errors.

Can robust conclusions be drawn if the uncertainties of the MLS data products are considered?

The errors are plotted in the figure and the differences are bigger.

Page 22920, line 12: 'These offline HO<sub>2</sub> has' - 'This offline HO<sub>2</sub> dataset has'

Done

Page 22920, line 19: 'from 10 to 0.0032' - 'from 10 to 0.0032 hPa' Done

Next line: 'from 1 to 0.0032' - 'from 0 to 0.0032 hPa'

Done

Page 22921, line 3: 'in the low side' - 'on the low side' ?

Done

Page 22921, line 11: 'as much as 60% but probably' - 'as much as 60%, which is probably' ?

Done

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Figure 4, caption, line 1/2: 'daily, weekly .. yearly 10 deg latitude bin' I find this phrase odd, because the latitude bin is not a 'daily, weekly etc.' latitude bin

Changed to: Expected precision for a daily (D), weekly (W), monthly (M) and yearly (Y) MLS HO2 offline data averaged over a 10 latitude bin.

Figure 5, caption, line 3: 'Magenta lines' There are no magenta lines on my screen (nor on the printout). To me it looks more like violet.

Changed to: Purple lines

Figure 6, caption, line 2: 'The MLS data correspond to the daytime-nighttime average of the 15 to the 25 September ..' Earlier in the paper you wrote about possible biases affecting both daytime and night time measurements at altitudes below the 1 hPa level. Because of the bias you reported differences between daytime and nighttime HO2 between 1 and 10 hPa. For the FIRS-1 comparison you use the daytime-nighttime average, which leads to the conclusion that the comparison shown in Fig. 6 is not a valid comparison. Can you use FIRS daytime measurements only to compare to your bias-corrected daytime measurements? This issue needs to be addressed.

We did use FIRS daytime data only as explained in the text. We also added in the caption: The FIRS profile corresponds to the one with the closest SZA to the MLS (daytime only) data.

Even though this is not the best comparison, since we are taking the day-night difference and an averaging over a latitude bin to compare against a single profile; this is the best we can do with noisy products. See for example, Kovalenko (2007) Validation of Aura Microwave Limb Sounder BrO observations in the stratosphere or Stachnik (2013) Stratospheric BrO abundance measured by a balloon-borne submillimeterwave radiometer or Pickett (2008) Validation of Aura Microwave Limb Sounder OH and HO2 measurements

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Figure 6, caption, line 5: Suggest changing 'The differences shown are' to 'The differences shown in the bottom panels are'

Done

Figure 8, caption, line 2: 'shown on April' - 'shown for April'

Done

Figure 10: this figure also shows daytime data, right? This should be mentioned in the caption.

Done

Figure 12: the dashed lines in row 2 and 3 are barely visible.

Changed to a darker gray

Figure 12, caption, line 5: 'has been use to' - 'has been used to'

Done

Baron et al. (2009) reference: All last names end with a 'k'. There's something wrong. Also the second 'Urban' should be 'Murtagh', right?

Corrected

Kikuchi et al. reference, line 3: Is 'Susukik' correct? This should probably read 'Suzuki'?

Corrected

Snow et al. (2005) reference: 'Mcclintock' - 'McClintock'

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Corrected

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