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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Stratospheric and mesospheric HO_2 observations from the Aura Microwave Limb Sounder" *by* L. Millán et al.

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Response to reviewer 2: Stratospheric and mesospheric HO2 observations from the Aura Microwave Limb Sounder

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We sincerely thank reviewer 2 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 HO2 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 O2 and H2O cross section was deleted, the

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 cannot 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 O3 photolysis as well as H2 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 CO2 SD-WACCM simulations improving its agreement with satellite measurements. Such adjustment should also affect the H2O 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

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 HO2 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 HO2 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.

The manuscript “Stratospheric and Mesospheric HO2 Observations from the Aura Microwave Limb Sounder” by Millán et al. presents new HO2 data derived from the AURA/MLS. Compared to the standard MLS version, these new data provide significant improvements with in particular information at high altitudes (up to 0.003 hPa) covering the mesospheric peak of HO2. These data are important to address opened issues such as the underestimation by the models of middle atmospheric O3 and day-time HO2. The error analysis and the comparison with two other instruments (FIRS-2 and JEM/SMILES) demonstrate the good quality of the dataset. Also the comparisons with WACCM (3-D climate model) and the Caltech/JPL 1-D chemical model confirms the problems to reproduce HO2 mesospheric abundance with current chemistry models. They also discuss the origin of this problem and conclude that the issue is related to the source and sink of HOx and not its partitioning. I think the manuscript can be published in ACP but before I would like the authors to check an issue related to Figure 12:

For me, the kinetic-2 and MLS profiles (upper panels) are in very good agreement at about 0.02 hPa (HO2 peak). The agreement is much better than the value of 20% seen in the lower subplots. Also even considering that the upper panel profiles are not smoothed (is-it correct ?), I don't understand why the absolute and relatives differences show that the maximum difference is around 0.02 hPa. I may have misunderstood something, please check.
That's correct, the difference is due to the smoothing. We added the following sentence at the beginning of the results comparisons to make it clearer: In this section we compare the offline HO2 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.

I have also some minor comments that I have listed below:

P22909, Line 5: Are these data currently publicly available? Will they become part of the standard MLS dataset?
In the introduction we added this sentence: To date, this dataset provides ten years of data and, in the near future, it will be publicly available for download in a daily based hierarchical data format (HDF).

P 22911, Line 6: Are the selected radiances include bands 28 and 30? Are you

including other bands? Have you compared only band 28 vs bands 28+30 ? Using band 30 should increase the contamination from the O3 line and, hence, increases the sensitivity of the retrieval to uncertainties on the O3 VMR, temperature and spectroscopy.
We added: The best retrievals were found when doing a jointly band 28 and 30 retrieval as oppose to doing retrievals using only band 28 or only band 30, even despite the O3 line influencing band 30.

Are these errors taken into account in the measurement covariance matrix? A comment about this issue could be added in the paper.
In the Error assessment section, we added in the systematic uncertainties list: the contaminant species errors, such as the O3 line influencing band 30 ...

P 22912, Line 15: I understand that only HO2 VMR is retrieved and other relevant atmospheric parameters (e.g., temperature, O3) are fixed (based on the standard MLS products). Is-it correct ? Are any other parameters retrieved to correct instrumental baseline ?
We added: Furthermore, at each pressure level, a constant baseline is retrieved for each band to correct any instrument baselines as well as to take care of the water vapor continuum contribution.

P 22915, Line 5: Would it be possible to provide the order of magnitude of the differences between a single profile at the FIRS-2 position and the zonal mean profile? For instance, the authors could use a model like WACMM or the MLS water vapor profiles a proxy.
Done: Overall, the two instruments agree on the HO2 vertical structure, with increasing HO2 with height (in the VMR representation) however there seems to be an bias between them, with the MLS data in the lower bound. This might be

due to the differences between the FIRS-2 single profile and the MLS zonal mean profile. In SD-WACCM (see Sect. 4.3 for the model description), these differences (i.e. comparing a single profile versus a zonal mean profile at this location) are around 30%.

The smoothing of the FIRS-2 profile and the day-night difference MLS profile indicated in the caption of Fig. 5 could also be indicated in the text.
We added a paragraph at the beginning of the result section stating that all the comparisons used the MLS averaging kernels.

The night-time MLS profile could also be plotted in Fig. 5.
After careful consideration, we decided not to add the night MLS profile to avoid to confusion since this is a daytime comparison.

What is the highest altitude of the FIRS-2 profile (before smoothing)? Is the FIRS-2 profile extrapolated for the interpolation at the higher altitudes?
In the paragraph at the beginning of the result section we also stated that there are no altitude interpolation.

P 22915, Line 7: Is-it the monthly mean of the differences (SMILES-MLS) or the difference of the monthly mean profiles?
We added: Figures 7 and 8 show daytime and nighttime comparisons (monthly means and the differences of the monthly means), respectively.

P 22916, Line 8: How the SMILES profiles have been smoothed in the upper altitude range of the retrieval (need of altitude extrapolation) ?
In the paragraph at the beginning of the result section we also stated that there are no altitude interpolation.

It should be indicated that unlike MLS, SMILES data are not regularly distributed over a month. This could explain some of the differences seen between SMILES and MLS in the mesosphere since the HO2 mesospheric peak shows large month to month variability.
We stated: The retrieval top level differences will need to be explored further, to investigate if they are due to retrieval artifacts (both retrievals are more sensitive to the apriori at these levels), calibration uncertainties or sampling differences (unlike MLS, SMILES data are not regularly distributed); this will require a joint effort from the MLS and SMILES teams.

P 22918, Line 14. It should be also mentioned that the underestimation of daytime HO2 in the model is seen above 1 hPa at all latitudes. This is consistent with previous studies given in the introduction of the paper (Sandor et al., 1998 and Khosravi, 2013).
We added: Figure 9 also shows that SD-WACCM underpredicts HO2 by about 20–30% between 1 and 0.1 hPa. This result agrees with previous studies (Sandor et al., 1998; Khosravi et al., 2013) but contradicts the result of the study by Canty et al. (2006).

I haven't seen any comment on the large overestimation of the model near 0.2 hPa.
The apparent model overestimation near 0.2 hPa is probably related to the MLS change in vertical scan pattern near this pressure level.

P 22918, Line 15. I would rather consider the range 10 to 1 hPa since a quasi-systematic underestimation of the WACCM HO2 occurs above 1 hPa.
See P 22918, Line 14 response

P22919, Line 12. Does Fig. 12 shows the daytime MLS profiles? This information should be indicated in the caption or in the text.
Done

The size of the lowest subplots could be increased.
In the final paper, there will be bigger. No ACPD banner and one column (instead of two column) figures.

What is the meaning of the dashed lines?
In the caption we added: The dashed gray lines show the MLS precision as well as the 20 and 40% percentage regions

P22919. It would like to have a short comparison with the conclusions from previous studies. Are these results in contradiction with the study by Canty et al. who used the standard MLS data in the lower-mesosphere ?
See P 22918, Line 14 response