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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.

Anonymous Referee #3

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Review of manuscript "Stratospheric and mesospheric HO2 observations from the Aura Microwave Limb Sounder" by Millán et al.

General comments:

This work presents a new HO2 dataset derived from Aura MLS measurements using an offline retrieval algorithm. The product, retrieved from averaged radiance profiles, presents several advantages over the standard v3.3 product, as the extended altitude range, the coverage of the polar regions, and also provides nighttime values for a wide altitude region. The manuscript describes the algorithm and the characterization of the retrieved quantity and assess the different error sources.

Comparisons with balloon-borne measurements and satellite measurements are also

presented as well as a comparison with a 3D chemistry climate model and a 1-D photochemical model. It is claimed that this dataset can be useful for a better understanding of the mesospheric O3 and HOx chemistry. In particular they found that the HOx partitioning in the retrieved HO2 and OH from MLS are compatible with our current understanding of the mesospheric chemistry. However, the absolute values of mesospheric HO2 are significantly underestimated by the models. Possible reasons for this underestimation are mentioned/listed although not really discussed or addressed.

I think that this new dataset of HO2 measurements add significant extra information to the standard product (e.g Fig. 2) and hence worth to be published. The result on the HOx partitioning of MLS products is also a significant contribution from the scientific (not only methodological) point of view. The other scientific result is just to point out to a models/MLS measurements disagreement which is not addressed. Then, it is not clear for me if the paper should be published in AMT or in ACP. I suggest that the authors give some more details and discussions on the possible causes of the disagreement (see below). This would make easier its publication in ACP.

Major comments:

Page 22913. Lines 21-24. I do not understand the meaning of the "retrieval numerics" error. My first guess would be that they are the "forward model" error, but this is considered in a separate contribution. Would that be what is normally called "smoothing" error? I.e., the effects of the regularization used in the retrieval? In the sentence "It is calculated as the retrieved value from the unperturbed radiances and the "truth" model atmosphere, i.e. that used for computing the synthetic radiance.", was that retrieval done with or without adding the noise to the synthetic radiance? I think it is important to clarify this error, since it is the major uncertainty in the region of HO2 maximum, above $\sim$0.1 hPa (Fig. 5). Related to this point, if they are actually the "smoothing" errors, they would be already taken into account when applying the AKs to the data to be compared and hence, the "bias" would not be as large as the 1 ppbv shown in Fig. 5 but significantly smaller. If this interpretation is correct, I would not consider this error

as a "bias" and would not mix with the other systematic (bias) errors. Related to this point, what is the "scatter" of the errors? What do they indicate?

In Secs. 4.3 and 4.4 the authors mention possible reasons for the discrepancies between the WACCM and 1-D models and MLS HO2 measurements. In particular they refer to our current uncertainty on the knowledge of the solar spectral irradiance measurements and/or its model representations, and the spectral resolution of the absorption cross sections of H2O and O2. Could the authors give some more details on what use the two models for these quantities? Do they have some hints on why they think they are possible causes or is it just speculation?

Other comments.

- Figures are very small and they have so many panels that are hardly readable in the printed version (I could read them only when zoomed out on the screen). In this sense, most of the figures are duplicated presenting the results in vmr and in number density. I cannot see any advantage of presenting additionally the number density figures. I think they could be removed and would help to make the other panels more readable.

- Fig. 1 and Page 22910 (lines 8 and 9). The text refers to 1K, 2K and 4K limb radiance precision. Is any of these that shown as the noise in Fig. 1? Why do you compare between these three precisions to say that the noise is large and averaging is needed? The signal at the top panel (band 28) for 4.6 hPa is much smaller (particularly at night) than the noise. However it looks as not affected by noise (very smoothed). Is it because the number of measurements averaged is very large? Would be useful to mention that number in the figure caption.

Page 22910. Lines 25 and ff. Just for curiosity, are the non-zero nighttime abundances positive, negative, both? You suggest to take the nighttime values as the "zero" for calculating the daytime values. However, the daytime and nighttime measurements are taken on different parts of the orbit (either ascending or descending). For other instruments the offset changes significantly along the orbit. Is that a good approach

for MLS or is the uncertainty in the correction of a similar magnitude that the correction itself?

Sec. 3. First full par. To be safer, I would consider as the daytime scans those with SZA< 85°. Would that make a significant change in the polar regions?

Near the end of this par., lines 10-13. "interpolated radiances". Apparently the sampling in altitude of MLS is ∼<1 km, and the vertical grid used here is ∼3km. Hence, it is also done some kind of "averaging" rather than "interpolation" in the radiances. Isn't it?

Lines 19-23. You mention here that "... for pressures between 10 and 1 hPa where the nighttime values exhibit non-zero values indicative of biases." However, Fig. 5 shows that the biases are not particularly large at those pressure levels; actually they are larger at lower pressures (higher altitudes). Shouldn't daytime values be calculated in a similar way above around 0.1 hPa, where the bias is also large?

Fig. 3. Are the results shown here for a daytime case? Please, state that, if so.

Fig. 4. I would remove the number density plot and would use a log scale for the errors. The caption refers to a "This profile". Is it the solid black line?

Last par. in Sec. 3.2, lines 6-8. "For pressures smaller than 0.1 hPa, the main source of bias and scatter are retrieval numerics, which, although unsatisfactory, is understandable given the 14 km vertical resolution in this region." This suggests to me that you are talking about a "smooth" error (see above). Correct?

Fig. 5 caption. families of systematic errors -> sources(?) of systematic errors As before, I suggest to remove the panels with the errors in the density. Idem for Fig. 6.

Sec. 4.1 Comparisons with FIRS-2. How many FIRS-2 profiles are available for that day? Just that used? If there are more but taken at other SZA's, and if SZA is very important, they could be corrected with a photochemical model. I think the statistics should be increased. BTW, in the figure caption is not mention that it is just one FIRS-2

profile.

Page 22916, lines 7-8, "The retrieval top level differences will need to be explored further,..." Given that there are so few HO2 measurements, and the importance of these measurements for the mesospheric chemistry (next sections), should not this be explored further in this work? It is important to clearly state that the models/MLS measurements comparison in the next section is not caused by a bias in MLS HO2 data.

Sec. 4.3. It is known that WACCM does not reproduce very well the measured temperature and O3 fields and even the meridional circulation (e.g. Smith, 2012; Smith et al., 2011; 2013). Could these be possible reasons to explain the HO2 WACCM-MLS differences? Furthermore, Garcia et al. (2014) has found that the parameterization of the gravity waves (GW), done through the change of the Prandtl number, significantly changes the CO distribution in the upper mesosphere. This might also impact H2O and hence HO2. Has this been explored?

In connection with this and the possible reason mentioned in the manuscript about possible inaccuracies in the representation of the absorption cross sections of H2O and O2 around the Lyman–Alpha region and the Schumann–Runge bands, Garcia et al. (2014) has found that an overestimation of the O2 cross-section in the 105–121 nm wavelength range was causing a too low CO concentration in the upper mesosphere. The large O2 cross-section assumed in the standard WACCM absorbed the UV radiation at high altitudes, preventing its penetration into lower altitudes and hence the CO production from CO2 photolysis. Although this spectral range is just at the edge of the Lyman-alpha, which affects H2O, this might be a reason for the WACCM/MLS discrepancy. With the reduced O2 cross-section, radiation will penetrates deeper, H2O will be more strongly photodissociated and hence producing more OH and more HO2. It might worth to explore this point.

Page 22917, par. at lines 17-21. Since the feature discussed in not shown in the

presented figures I cannot see the reason for its discussion. I suggest to remove it.

Page 22918, lines 10-15. It would be useful to mention which solar flux data is used in WACCM and how other data would change (at least qualitatively) the results. The same applies to the 1-D model described in Sec. 4.4 and it is extensive to the parameterization of the cross-sections (see major comment above).

Page 22918, lines 18-20. "For pressure levels smaller than 0.1 hPa, the lack of a clear second peak in the SD-WACCM dataset reflects the smaller mesospheric concentrations in 20 this dataset." To which "second peak" do you refer? I cannot see it (just see the peak at $\sim$0.02 hPa).

Page 22918, line 27. "poleward" -> "towards the winter pole"?

Page 22919, lines 1-2. "and overestimating by as much as 50 % over the polar winter regions." This might be right but I would not conclude that when comparing second (WACCM with AKs) and third (MLS) panels in the left column of Fig. 11. They both appear with the same light green color. BTW, in the percentage differences panels, which WACCM is being used, with or without the applied AK's? This comment is extensive to all figures where the AKs are applied (e.g. Figs. 7, 8, 9, and 10 as well).

page 22920, line 13, Typo, extended?

Page 22291, line 2, I would call this a "bias" rather than an "offset".

Page 22291, line 10-14. "In the upper mesosphere, we found an underestimation by the model by as much as 60% but probably in part due to the low spectral resolution of the absorption cross sections in the Lyman–Alpha region and Schumann–Runge bands." This reason has just been mentioned in the text as a possible explanation but has not been studied in this work. The reader might be mis-led. It should be re-written.

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