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# Validation of Aura MLS $HO_x$ measurements with remote-sensing balloon instruments

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[1] Satellite measurements of OH and $HO_2$ obtained by the Aura MLS instrument are compared to the balloon-borne BOH and FIRS-2 instruments. All measurements are also compared with constrained photochemical model calculations. On average, both balloon measurements of OH agree with MLS within 17% over 25–40 km and the measurements agree with the model within 12%. The three measurements for column of OH above 40 km agree within 8% and the mean is 12% below the model. Measurements of $HO_2$ from FIRS-2 and MLS agree on average within 23% over 25–40 km and the differences are generally within the experimental precision. The $HO_2$ measurements agree with the model within 14%. Measurements of $HO_2$ for the column over 40–60 km agree within 16% and the mean measured column agrees with the model within the experimental precision. Our observations do not appear to indicate a "$HO_x$ dilemma." **Citation:** Pickett, H. M., B. J. Drouin, T. Canty, L. J. Kovalenko, R. J. Salawitch, N. J. Livesey, W. G. Read, J. W. Waters, K. W. Jucks, and W. A. Traub (2006), Validation of Aura MLS $HO_x$ measurements with remote-sensing balloon instruments, *Geophys. Res. Lett.*, *33*, L01808, doi:10.1029/2005GL024048.

## 1. Introduction

[2] The Aura satellite was launched on July 15, 2004 into a sun-synchronous near-polar orbit. The Microwave Limb Sounder (MLS) instrument on the Aura satellite has the capability to measure OH and $HO_2$ in both day and night [*Waters et al.*, 2006]. Further details on the THz module and the OH measurement and calibration are given by *Pickett* [2006]. Early validation of other molecules measured by MLS are given by *Froidevaux et al.* [2006]. Version 1.51 of the retrieval software was used in this paper.

[3] The Balloon OH instrument (BOH) and Far Infrared Spectrometer (FIRS-2) instrument were launched on a common balloon gondola on September 23, 2004, from Ft. Sumner, NM (latitude = 34.5° and longitude = −104°) and stayed aloft at ~38 km for nearly 24 hours. The BOH instrument is a heterodyne limb-viewing thermal emission instrument that is functionally identical to the THz module on MLS [*Pickett*, 2006] and only measures OH. The FIRS-2 instrument is a thermal emission far-infrared Fourier transform spectrometer developed at the Smithsonian Astrophysical Observatory [*Jucks et al.*, 1998]. It measures OH and $HO_2$ in the far infrared using multiple lines.

[4] Odd hydrogen ($HO_x$ = OH + $HO_2$ + H) chemistry dominates ozone destruction above 40 km and below 25 km. Previous observations of OH over 40–80 km from MAHRSI [*Conway et al.*, 2000] are not consistent with current chemical models, leading to the designation "$HO_x$ dilemma." However, previous FIRS-2 observations that are mostly sensitive to $HO_x$ below 40 km agree better with standard photochemistry [*Jucks et al.*, 1998].

## 2. Measurements and Model

[5] Since the Aura orbit is sun-synchronous, the local solar time (LST) and solar zenith angle (SZA) are nearly the same for each orbit on a given day. However, successive overpasses have a change in longitude of −24.7°. The MLS retrieval position at 34°N latitude on Sept. 23, 2004 had LST = 13.46 hr and SZA = 38.92° for the daytime ascending overpass. The night descending overpass has LST = 2.20 hr and SZA = 135.1°. MLS OH profiles are available at 1.5° intervals along each orbit for altitudes of 22–90 km. The MLS precision for $HO_2$ and for OH below 35 km is poorer. To obtain better precision, MLS $HO_2$ and OH profiles were zonally averaged over a latitude interval of 20° for a period of 15 days centered on the balloon-flight day.

[6] The BOH and FIRS-2 instruments can spend more time acquiring data because they are localized in latitude and longitude. The BOH instrument scans the limb in 15 seconds, and calibrated radiances averaged for 20–30 minutes are used for the retrieval. The FIRS-2 instrument takes 70 minutes to scan the limb. For direct comparisons with MLS, we use the scans closest to the overpass in LST. For altitudes less than that of the balloon, both balloon-borne instruments obtain profiles in limb-sounding mode with a height resolution of 3–6 km. Above the balloon altitude, both instruments lose the limb sounding path-length advantage. In addition, there is little spectral information in the radiance because the line shape is essentially Doppler-limited above 45 km. In addition, the OH lines are optically thick at noon and the foreground OH tends to mask the OH above 60 km. For both instruments we assume a profile for OH or $HO_2$ above the balloon and then scale it to bring the calculated radiance into agreement with observation. The FIRS-2 instrument uses a diurnally-varying model calculation and the BOH instrument uses a noon

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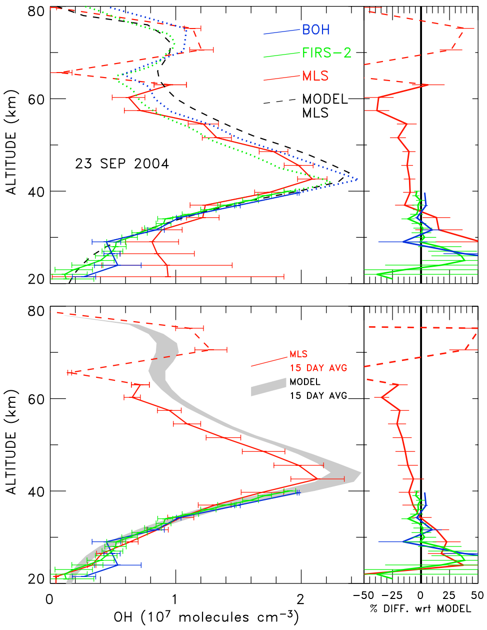

**Figure 1.** OH altitude profiles near 13.5 hr LST. The lower panel displays an average of MLS OH for 15 days centered on the balloon-flight day, over a 20° latitude range. For reference, balloon-borne measurements are duplicated in the lower panel. The model denoted by the dashed line uses MLS constraints for 23 Sept. 2004, while the shaded region denotes a model range for the MLS 15-day average.

MLS OH profile. Despite these differences, both instruments are sensitive to the column density above the balloon.

[7] The MLS and balloon-borne vertical resolution is approximately equal but there are significant differences in the horizontal footprint. In all three instruments, the horizontal resolution perpendicular to the boresight is comparable to the vertical resolution (~3 km). The horizontal footprint along the line of sight is based on the limb sounding geometry (400 km for 3 km height resolution). For MLS, the boresight at 34°N latitude has a horizontal heading of 350° for the ascending overpass. The balloon-borne instruments look out at a common azimuth angle (east at the time of the Aura daytime overpass). Therefore, the footprint is comparable in size, but the orientation can be quite different. In addition, the MLS footprint position does not change with altitude, while the balloon-instrument footprint position varies with height by as much as 470 km.

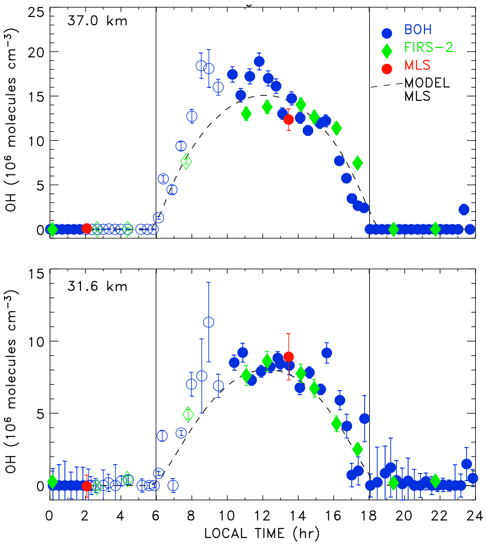

**Figure 2.** OH diurnal behavior: upper panel is number density for 37 km and lower panel is for 31.6 km. Data collected after the azimuth pointing system failure is indicated by open symbols.

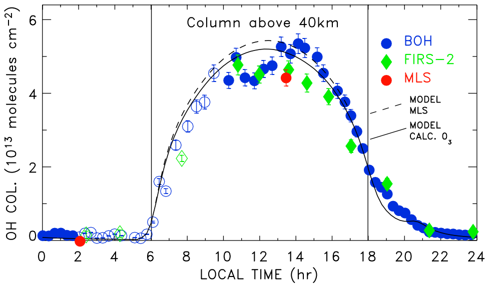

**Figure 3.** Same as Figure 2, for column from 40 to 80 km. Two model values are shown, as described in the text.

[8] The photochemical model employed here has been used previously to interpret data from aircraft, satellite, and balloon platforms [*Jucks et al.*, 1998, and references therein]. In the present analysis, the model is constrained by MLS measurements of [$O_3$], [$H_2O$], [$Cl_y$], [$NO_y$], [$N_2O$] (to infer [$CH_4$]), and temperature in the stratosphere and lower mesosphere (see auxiliary material[1]). Currently, MLS $H_2O$ profiles gradually revert to climatology above 60 km. Since there is diurnal variability of ozone, either daytime averages of [$O_3$] were used or [$O_3$] is unconstrained (see Figure 3). To improve the mesospheric capabilities of the model, Lyman-$\alpha$ photochemistry is sampled at 16 wavelengths over the solar Doppler profile [*Chabrillat and Kockarts*, 1997] using the O($^1$D) yields of *Lacoursiere et al.* [1999]. All other kinetics are from *Sander et al.* [2002]. During the 15-day averaging period needed for $HO_2$ and low-altitude OH, the $H_2O$ and $O_3$ concentrations were remarkably stable. The model was constrained to the average MLS values. The grey shading in the figures shows the range of concentrations that correspond to the 20° range of latitudes (and corresponding SZA values) used in the average.

[9] In Figures 1–5 the error bars for all measurements are 1$\sigma$ estimates of precision for the average displayed. In the lower panel of Figure 1, we inflate the MLS errors to 10% to account for uncertainties in instrument calibration (since the precision in the 15-day average above 35 km is very small). In Figures 1 and 4, the right panels are the percent difference of measurement minus model divided by the model. The balloon-borne instruments are only sensitive to a single column-like quantity above 40 km. Accordingly, the assumed profiles are shown as a dotted line without error bars.

[10] Figure 1 shows the OH altitude profiles in number density near 13.5 hr LST. The nearest FIRS-2 averages were at 13.6 hr LST. The nearest BOH profile had a mean LST =

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[1] Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2005GL024048.

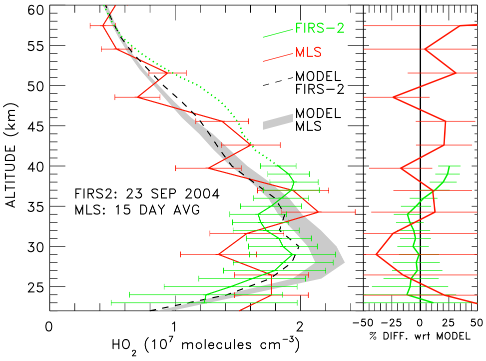

**Figure 4.** $HO_2$ profile near 13.5 hr LST. MLS data is an average of 15 days centered on the balloon-flight day. The model denoted by the dashed line uses FIRS-2 constraints and conditions, while the shaded region denotes a model range for the MLS 15-day average.

13.7 hr for altitudes above the balloon, decreasing to 13.4 hr at a tangent of 22 km. The averaging time for this BOH profile is 29 minutes. In the upper panel, MLS OH profiles were averaged over a 5° interval centered on 34N latitude and a longitude range of −122° to −72°. This average represents only 2 minutes of satellite observation time. The lower panel shows the effect of averaging for 15 days centered on the balloon day. The MLS OH values for the 15-day average are daytime values above 32 km and day–night differences below.

[11] The two panels in Figure 1 show the same balloon-borne data compared with a MLS local 1-day average (upper) and the 15-day average (lower). For the 1-day average, the overlap with the balloon instrument profiles is useful over 35–40 km and the differences between the balloon-borne instruments and MLS have an average absolute deviation (AAD) of 15%. Between 25–35 km, where MLS profiles must be zonally averaged over 15 days, the differences have an AAD of 19%. Combining the two ranges, the AAD over 25–40 km is 17%. The AAD between the measurement and the model over 25–40 km is 12%, and deviations are within measurement precision. The sharp dip at 65 km in the MLS data may be a retrieval artifact due to a change in scan speeds at this altitude. In addition, the MLS OH becomes negative above 80 km. The line marked 'MLS' in Figure 1 is changed to a dashed line above 64 km to highlight these potential systematic errors. We intend to have these retrieval issues resolved in the next version of the software, but the OH column above 60 km is not likely to change.

[12] Figure 2 shows the diurnal variability of OH density. The altitudes shown are MLS sample points near the mean balloon altitude and one scale height below. The plots show good agreement between model and experiment in the afternoon and night. On this balloon flight, the gondola azimuth pointing system suffered an electronics failure at 2.5 hr LST on 24 Sept. 2004 and thereafter the gondola rotated freely until flight termination at 10 hr LST. The data for this time period are indicated by open symbols and should be viewed with some caution.

[13] Figure 3 shows the diurnal variability of the column over 40–80 km. This column is an approximate measure of OH observed by FIRS-2 and BOH above the balloon altitude. The column near 13.5 hr LST shows a difference of 12% between BOH and MLS and a difference of 4.5% between FIRS-2 and MLS. These columns are an independent validation of MLS OH measurements above 40 km. The dashed line in Figure 3 indicates the standard model run in which $O_3$ is constrained to MLS daytime values (MODEL MLS), while the solid line indicates a model run in which $O_3$ is unconstrained (MODEL CALC. $O_3$). The difference between these curves indicates the sensitivity to $O_3$ above 40 km. The constrained model is 12% higher than the mean column for the three measurements.

[14] Figure 4 shows the $HO_2$ density profiles near 13.5 hr LST. Figure 5 shows $HO_2$ diurnal variability at two altitudes. As described above, the MLS data are an average of 15 days centered on the flight day. The MLS data shown in both figures are a day–night difference. The dashed line indicates a model run constrained by FIRS-2 SZA and concentrations of $O_3$, $H_2O$, etc. (see auxiliary material), and the grey area indicates the range of model runs for the MLS latitude average. The plots show an AAD of 23% over 25–40 km for the differences between FIRS-2 and MLS, well within the combined experimental uncertainties. We are investigating whether the altitude oscillation seen in Figure 4 for MLS $HO_2$ is a retrieval artifact.

[15] The $HO_2$ columns over 40–60 km are shown in Table 1. Column $HO_2$ measured by MLS is 16 ± 12% (1 σ precision) smaller than that measured by FIRS-2. The MLS column is 4 ± 7% larger than its model, and the FIRS-2 column is 19 ± 10% larger than its model, a difference of less than $2\sigma$.

## 3. Conclusions

[16] Densities of OH obtained from the balloon-borne BOH and FIRS-2 instruments agree with MLS on average with an AAD of 17% over 25–40 km and agree with the model with an AAD 12%. Columns of OH above 40 km agree among themselves within 8% and are less than the model by 12%. Measurements of $HO_2$ below 40 km from FIRS-2 and MLS agree with an AAD of 23%. Agreement

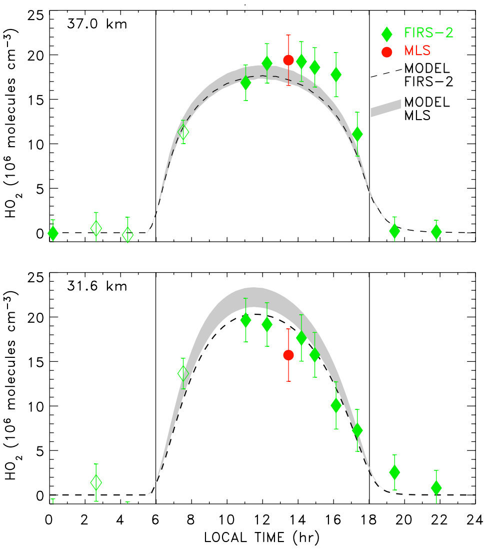

**Figure 5.** $HO_2$ diurnal behavior: See Figure 2.

**Table 1.** $HO_2$ Column Over 40–60 km Near 13.5 hours LST

| Source | Column, $10^{13}$ $cm^{-2}$ |
|---|---|
| MLS | 1.86 ± 0.13 |
| MLS model | 1.79 |
| FIRS-2 | 2.21 ± 0.18 |
| FIRS-2 model | 1.85 |

between model and measured $HO_2$ is 18% over 25–40 km and is limited by the experimental precision. The $HO_2$ column measurements over 40–60 km agree within 15%. MLS and FIRS-2 $HO_2$ columns agree with the model within experimental precision.

[17] Earlier measurements of OH from MAHRSI were shown to be 20% higher than calculated values near the OH peak at 45 km and about 25–35% lower than calculated values over 50–70 km [*Conway et al.*, 2000]. These differences were termed the "$HO_x$ dilemma" because certain changes in model kinetics, such as variations to the rate constants of the reactions O + OH, O + $HO_2$, and OH + $HO_2$ (within laboratory uncertainties), will lead to better agreement between measured and modeled OH for one altitude region (e.g., 50–75 km) but will worsen the agreement in the other height region (e.g., near 45 km) [*Conway et al.*, 2000]. Our calculations of OH are close to those reported by the MAHRSI group, for the same model constraints [*Jucks et al.*, 1998].

[18] In contrast to the MAHRSI measurements, MLS measurements of OH are about 10% lower than model values at the 45 km OH peak. The OH profile measured by MLS for 50–60 km is in better agreement with the model than found for similar comparisons using MAHRSI OH, but nonetheless MLS OH is systematically lower than model OH by 10–25%. The differences between MLS measured and modeled mesospheric OH below 64 km could likely be explained by variations in the rate constants for O + OH, O + $HO_2$, and/or OH + $HO_2$ that are well within the laboratory uncertainties [*Sander et al.*, 2002]. The differences between observed and modeled $HO_2$ place important additional constraints on such perturbations. Regardless, the MLS measurements of OH for Sept. 2004 at mid-latitudes do not present a "dilemma" in the sense of the MAHRSI OH observations because kinetics changes required to bring measured and modeled OH into closer agreement for the mesosphere will not prevent agreement, within measurement uncertainties, from also being obtained near 40 km altitude.

[19] **Acknowledgments.** We wish to thank all who helped make the Aura $HO_x$ measurements possible. We are grateful to NASA's Upper Atmosphere Research program for support of the balloon-borne instruments at SAO and JPL. We are grateful to the National Scientific Balloon Facility for launch services. Research at the Jet Propulsion Laboratory, California Institute of Technology, is performed under contract with the National Aeronautics and Space Administration. We thank the referees for their for their many helpful comments.

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