Validation of Aura MLS HO_x Measurements with Remote-Sensing Balloon Instruments

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Measurements of OH and HO_2 obtained by the MLS instrument on the Aura satellite are compared with balloon-borne observations from the BOH and FIRS-2 instruments made on September 23-24, 2004 at a latitude of 34°N. All measurements are compared with model calculations constrained by MLS measurements of temperature, O_3, H_2O, and N_2O. Precision for MLS OH above 35 km allows comparisons at the same local solar time near the same latitude and longitude, but MLS precision for HO_2 and for OH at 20–35 km requires zonal means over 15 days. All three measurements of OH agree among themselves within 10% over 30–60 km and agree with a photochemical model within 25%. Measurements of HO_2 from FIRS-2 generally agree within 10% of the model. Agreement between model and MLS HO_2 is 25% and is limited by the experimental precision. For this latitude and season we do not observe any indication of a "HO_x dilemma."

1. Introduction

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 both OH and HO_2 simultaneously both in day and night [*Waters et al., submitted*]. Further details on the THz module and the OH measurement and calibration are given in *Pickett* [*submitted*]. Early validation of other molecules measured by MLS are given in *Froidevaux et al.* [*submitted*]. The retrieval software version used in this paper is 1.51.

The BOH and FIRS-2 instruments 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 and remained within 250 km of the launch site. The BOH instrument is a heterodyne limb-viewing thermal emission instrument that is functionally identical to the THz module on MLS [*Pickett, submitted*] 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.

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Odd hydrogen (HO_x = OH+HO_2+H) chemistry dominates ozone destruction above 40 km and below 25 km. Previous observations of OH between altitudes of 40 and 80 km from MAHRSI [*Conway et al.*, 2000] are not consistent with current chemical models, leading to the designation "HO_x dilemma." However, FIRS-2 observations that are mostly sensitive to HO_x below 40 km agree better with standard photochemistry [*Jucks et al.*, 1998]. It is therefore important to compare the MLS and balloon-borne measurements with model calculations.

2. Measurements and Model

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 September 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. In order to obtain better precision, MLS HO_2 and OH profiles were also zonally averaged over a latitude interval of 20° for a period of 15 days centered on the balloon-flight day.

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, like ground-based emission instruments. Unfortunately, above the balloon altitude there is little spectral information in the radiance because the line shape is essentially Doppler-limited above 45 km. For this reason, there is little information about the profile above the balloon. 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 MLS OH profile. Despite these differences, both instruments are sensitive to the column density above the balloon.

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 ($\sim$3 km). The horizontal footprint along the line of sight is based on the limb sounding geometry (200 km for 3 km height resolution). For MLS, the boresight at 34°N latitude has a horizontal heading of 350° for the ascending overpass. MLS uses a 2-dimensional retrieval so that the footprint is independent of altitude. The balloon-borne instruments look out at a common azimuth angle that is controlled by a gondola pointing system. At the time of the Aura daytime overpass the boresight was pointing east. The tangent at the balloon altitude is located at the position of the balloon, but a tangent at 20 km is located at a great circle distance of 4.2° (466 km), assuming a balloon altitude of 38 km. 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.

To help account for differences in scene averaging it is useful to compare both satellite and sub-orbital measurements to a model. 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 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-α photochemistry is sampled at 16 wavelengths over the solar Doppler profile [*Chabrillat and Kockarts*, 1997] using the $O(^1D)$ 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 Figures 1, 4, and 5 shows the range of concentrations that correspond to the 20° range of latitudes (and corresponding SZA values) used in the average.

In Figures 1–5, the error bars for all measurements are 1σ estimates of precision for the average displayed. In the lower panel of Figure 1, we inflate the 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. Model values are used in the denominator since these vary smoothly with altitude.

Figure 1 shows the OH altitude profiles in number density near 13.5 hr LST. The nearest FIRS-2 averages were at LST = 13.6 hr. The nearest BOH profile had an average LST = 13.7 hr for altitudes above the balloon, decreasing to 13.4 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 34° 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.

All of the measured OH profiles agree within 10% over 22–60 km. However, differences between the measurements above 40 km are not significant given that the balloon-borne instruments are only sensitive to a single column-like quantity above 40 km. Agreement with the model is excellent below 45 km. 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. We hope to have both these issues resolved in the next version of the retrieval software, but the OH column above 60 km is not likely to change.

Figure 2 shows several OH diurnal profiles of concentration. 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 LST = 2.5 hr on 24 September 2004 and thereafter the gondola rotated freely until flight termination at LST = 10 hr. The data for this time period are indicated by open symbols. Balloon data taken during this time should be viewed with some caution.

Figure 3 shows the diurnal profile of the column above 40 km. This column is an approximate measure of OH observed by FIRS-2 and BOH above the balloon altitude. The plots show good agreement between model and the three experiments during daytime. 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 decay of OH in Figure 3 just after sunset observed by BOH is not well reproduced by the model.

Figure 4 shows the HO_2 altitude profiles in number density near 13.5 hr LST and Figure 5 shows two HO_2 diurnal profiles. As described above, the MLS data is an average of 15 days centered on the flight day. The MLS data shown in both figures is a day–night difference. The dashed line indicates a model run constrained by FIRS-2 concentrations and SZA of O_3, H_2O, etc. (see auxiliary material[1]), and the grey area indicates the range of model runs for the MLS latitude average. The plots show 25% agreement between FIRS-2 and MLS within the combined experimental uncertainties. The mid-day HO_2 profile measured by FIRS-2, as well as the diurnal variation at 31.6 and 37.0 km, all exhibit excellent agreement with the model calculations with differences generally less than 10%. The agreement between the model and MLS is 25% and is limited by the measurement precision. We are investigating whether the oscillation observed for MLS HO_2 is a retrieval artifact.

3. Conclusions

Concentrations of OH obtained from the balloon-borne BOH and FIRS-2 instruments agree among themselves within 10% over 30–60 km and agree with a photochemical model within 25%. Measurements of HO_2 from FIRS-2 generally agree within 10% of the model. Agreement between model and MLS HO_2 is 25% and is limited by the experimental precision.

Earlier measurements of OH from MAHRSI were shown to be ∼25% higher than calculated values near the OH peak at 45 km and about 40% lower than calculated values over a broad altitude region in the mesosphere [*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].

In contrast to the MAHRSI measurements, MLS measurements of OH are about 10% lower than model values at the 45 km OH peak and 30% higher than the model at 70 km. The OH profile measured by MLS for 50–60 km is in better agreement with the model (differences typically 10 to 25%) than are found for comparisons using MAHRSI OH. The differences between MLS measured and modeled mesospheric OH 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 excellent agreement between FIRS-2 HO_2 and modeled HO_2 places 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.

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Notes

1. Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2005GLXXXXX.

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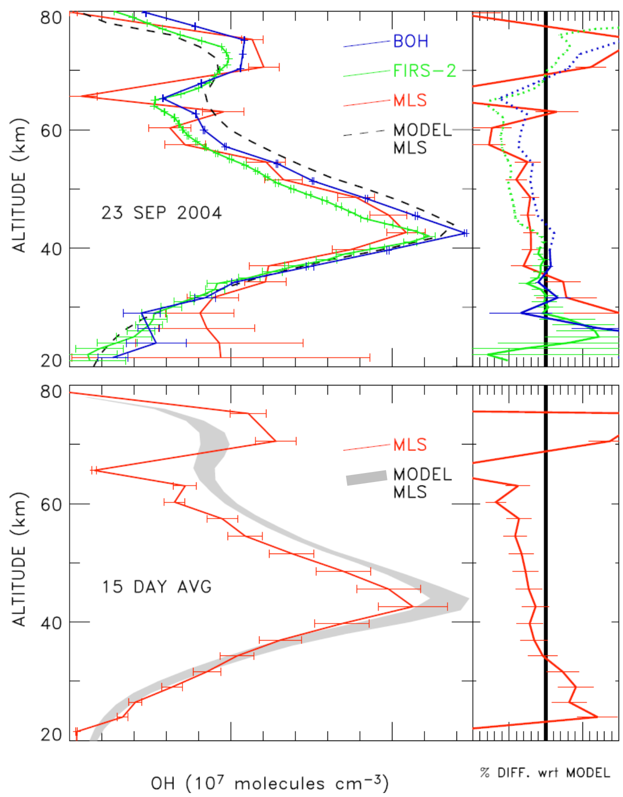

Figure 1. OH altitude profiles near 13:30 local solar time. Lower panel is an average of 15 days centered on the balloon-flight day, over a 20° latitude range. The model denoted by the dashed line uses MLS constraints for 23 Sept. 2004, while the shaded region denotes a model range for latitudes included in the MLS 15-day average.

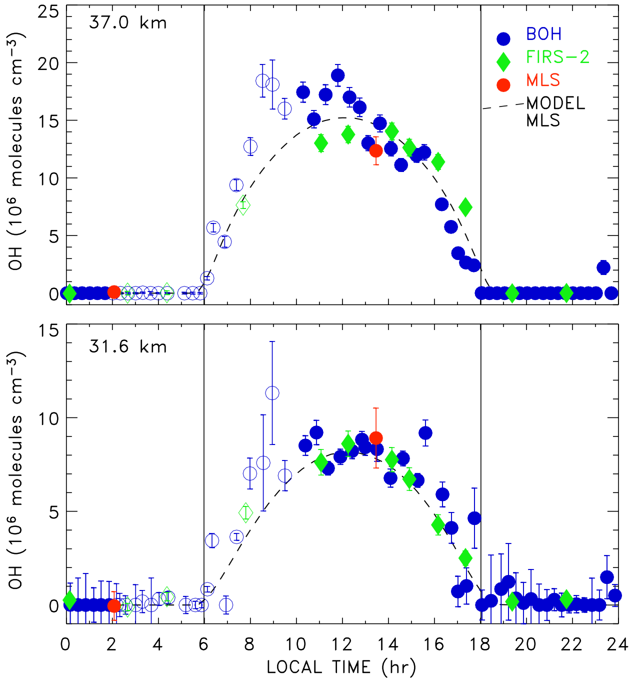

Figure 2. OH diurnal behavior: upper panel is number density for 37 km and lower panel is for 31.6 km. Data collected after the azimuth pointing system failure is indicated by open symbols.

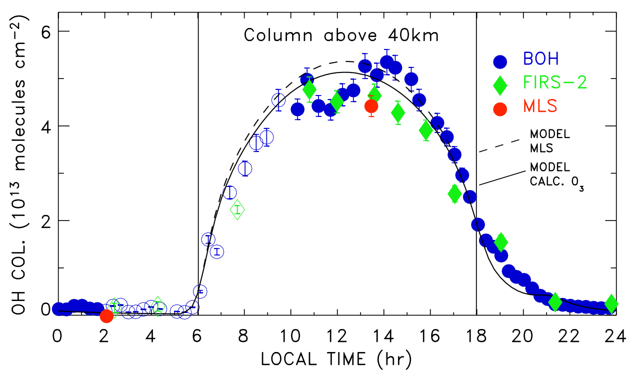

Figure 3. Same as Figure 2, for column above 40 km. Two model values are shown, as described in the text.

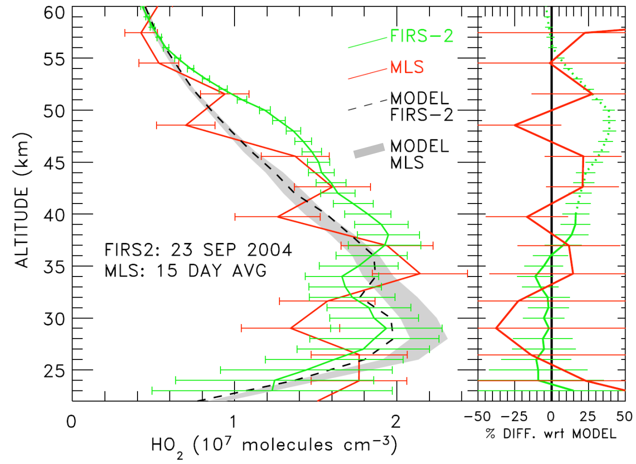

Figure 4. HO_2 profile near 13:30 local solar time. MLS data is an average of 15 days centered on the balloon-flight day. The model denoted by the dashed line uses FIRS-2 constraints and conditions, while the shaded region denotes a model range for latitudes included in the MLS 15-day average.

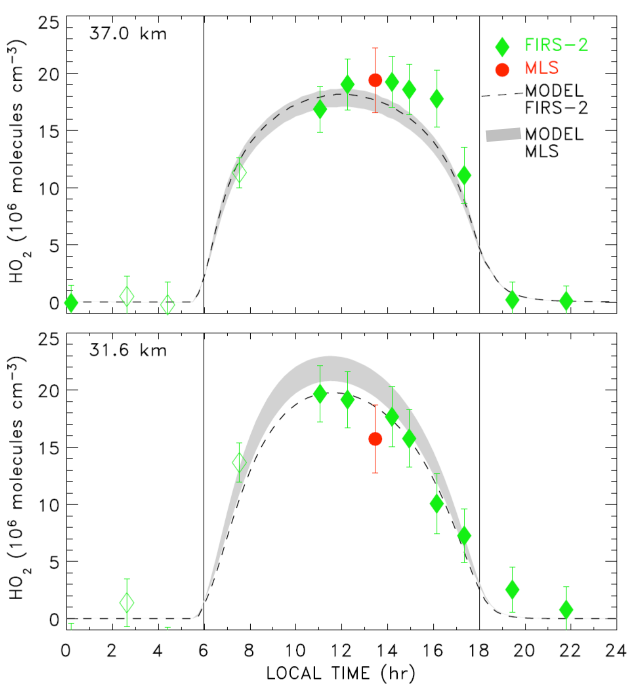

Figure 5. HO_2 diurnal behavior: upper panel is number density for 37 km and lower panel is for 31.6 km. Model designations same as Figure 4. Data collected after the azimuth pointing system failure is indicated by open symbols.