JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 112, D24S33, doi:10.1029/2007JD008720, 2007

Ground-based microwave ozone radiometer measurements compared with Aura-MLS v2.2 and other instruments at two Network for Detection of Atmospheric Composition Change sites

Ian S. Boyd,[1] Alan D. Parrish,[2] Lucien Froidevaux,[3] Thomas von Clarmann,[4] Erkki Kyrölä,[5] James M. Russell III,[6] and Joseph M. Zawodny[7]

Received 29 March 2007; revised 29 June 2007; accepted 30 July 2007; published 24 November 2007.

[1] Ozone measurements made by the Microwave Limb Sounder (MLS) on board the Earth Observing System (EOS) Aura Satellite are compared with measurements made by ground-based microwave radiometers (MWR) in the Network for Detection of Atmospheric Composition Change (NDACC) stations at Lauder, New Zealand (45°S, 169°E) and Mauna Loa, Hawaii (20°N, 204°E). The latter instruments measure ozone over the pressure range 56 to 0.03 hPa (about 20 to 72 km), allowing validation of ozone to the upper range of the MLS profiles. In addition, because they operate continuously, separate daytime and nighttime comparisons with MLS can be made to account for the large diurnal variations of ozone in the upper stratosphere and mesosphere. MLS-MWR comparisons show agreement generally within 5% between 24 and 0.04 hPa (about 26 to 70 km) and 5 to 13% elsewhere. To more thoroughly investigate ozone in the stratosphere and mesosphere and establish a consensus between different sets of measurements, comparisons, and analyses with other satellite-borne instruments, including the Stratospheric Aerosol and Gas Experiment II (SAGE-II), Halogen Occultation Experiment (HALOE), Global Ozone Monitoring by Occultation of Stars (GOMOS), and Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), are also made, using the ground-based microwave measurements as a reference. The resulting MLS-consensus difference profiles remove some of the features present in the MLS-MWR comparisons and indicate that the overall agreement between MLS and the correlative data, between 56 and 0.04 hPa, is mostly within 5% at both sites.

Citation: Boyd, I. S., A. D. Parrish, L. Froidevaux, T. von Clarmann, E. Kyrölä, J. M. Russell III, and J. M. Zawodny (2007), Ground-based microwave ozone radiometer measurements compared with Aura-MLS v2.2 and other instruments at two Network for Detection of Atmospheric Composition Change sites, *J. Geophys. Res.*, *112*, D24S33, doi:10.1029/2007JD008720.

1. Introduction

[2] The Aura satellite was launched on 15 July 2004 as part of a suite of environmental satellites flying in formation in space, including Terra, Aqua, CloudSat, PARASOL, and CALIPSO (collectively referred to as the A-Train). The four instruments aboard Aura are designed to provide measurements of 23 trace gases, temperature, height, and atmospheric aerosols, with an emphasis on air quality as well as stratospheric ozone and its depletion. These measurements address a wide range of concerns regarding the evolution of the composition and properties of Earth's atmosphere resulting from both human activities and natural causes and provide a means to improve predictions of the environmental consequences of the evolution.

[3] Together with other recently launched satellite-borne instruments, the Aura instruments provide near global coverage of simultaneous observations of temperature, ozone, and other trace gases over a wide altitude range. They will be important in providing accurate observational constraints to atmospheric models, as well as improving understanding of daily, short-term, and long-term variations in ozone and the coupling between the troposphere and stratosphere and the stratosphere and mesosphere. The Aura instruments also continue an extensive time series of satellite-borne ozone profile measurements, starting with the Stratospheric Aerosol and Gas Experiment (SAGE)-I and SAGE-II instruments. It is important that systematic differences between the calibration of new instruments and older

[1]NIWA Environmental Research Institute, University of Massachusetts, Amherst, Massachusetts, USA.
[2]Department of Astronomy, University of Massachusetts, Amherst, Massachusetts, USA.
[3]Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California, USA.
[4]Forschungszentrum Karlsruhe, Institut für Meteorologie und Klimaforschung, Karlsruhe, Germany.
[5]Earth Observation, Finnish Meteorological Institute, Helsinki, Finland.
[6]Center for Atmospheric Sciences, Hampton University, Hampton, Virginia, USA.
[7]NASA Langley Research Center, Hampton, Virginia, USA.

0148-0227/07/2007JD008720

ones be known in order to understand the evolution of the atmosphere over extended periods.

[4] The work presented here is limited to supporting validation of ozone measurements made with Microwave Limb Sounder (MLS) aboard Aura using two different techniques. We first compare the recently released MLS version 2.2 data set with ground-based microwave measurements made at Lauder, New Zealand (45°S, 169°E), and Mauna Loa, Hawaii (20°N, 204°E), which are Primary Network for Detection of Atmospheric Composition Change (NDACC) Stations. The microwave radiometers (henceforth referred to as MWR) are among the suite of instruments at these stations. We then use the MWR measurements as transfer standards, in a manner similar to that proposed by *Hocke et al.* [2007], to enable comparison of the MLS measurements to other satellite-based measurements that in most cases have little or no overlap in time with MLS. For this part of the work we have chosen instruments that have historically been used extensively for studies of ozone evolution as well as newer instruments that provide measurements over the full range of altitudes covered by the MLS measurements.

[5] Section 2 describes the instruments used in this study and their measurement techniques. These include Global Ozone Monitoring by Occultation of Stars (GOMOS) and Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) aboard Envisat and the SAGE-II and Halogen Occultation Experiment (HALOE) instruments. Section 3 describes the comparison between MLS and MWR measurements. Section 4 describes the comparison of MLS and other satellite-based measurements using the MWR measurements as transfer standards. Section 5 discusses some details regarding the interpretation of the comparison satellite measurements in the mesosphere that affect the interpretation of our results.

2. Instruments

[6] The MWRs have been in operation at Lauder and Mauna Loa since 1992 and 1995, respectively, and observe atmospheric thermal emission at 110.836 GHz. They consist of a cryogenically cooled heterodyne receiver and 120 channel spectrometer, and record the spectral line shape of an ozone rotational transition every 20 min at about 10 to 20 degrees elevation. A basic description of the instrument, observing technique, calibration, and profile retrieval is given by *Parrish et al.* [1992]. Additional engineering details are reported by *Parrish* [1994]. The ozone altitude distribution is retrieved from the pressure broadened line shape using the optimal estimation method of *Rodgers* [1976]. As described by *Connor et al.* [1995], the retrieved profile x_r is given by

$$x_r = A\, x_t + (I - A)x_a + e_r \tag{1}$$

where x_t is the true ozone profile, x_a is the a priori profile furnished to the retrieval algorithm, I is the identity matrix, and A is the averaging kernel matrix for the system (i.e., the instrument and its retrieval algorithm). The first term on the right-hand side of equation (1) shows how the true profile is smoothed by the averaging kernels. The a priori profile makes a small contribution to the retrieved profile, described by the second term. For the present version of the data, the magnitude of each element of this term is less than 15%, and mostly less than 7%, of the corresponding element of x_r between 20 and 64 km during the day. At night, the upper limit moves up to about 72 km owing to the increased mesospheric ozone signal. The a priori sensitivity increases rapidly outside these respective ranges. The third term, e_r, describes the effects of systematic and statistical errors in the measurement of the ozone spectrum, and spectral modeling errors, on the retrieved profile. These effects are discussed in detail in the preceding reference, so we will only give a brief summary of results below.

[7] MWR observations continue 24 hours a day, with selected spectra chosen based on the absolute tropospheric opacity and its variability, where limits for these values are set empirically at each site. This allows measurements in weather ranging from clear through to some overcast conditions. Good quality spectral data are routinely averaged over 4 to 6 hours to generate up to four ozone volume mixing ratio (VMR) profiles per day. The expected precision is 4 to 5% between 20 and 57 km and 7% at 64 km. The expected accuracy (i.e., combined random and systematic error) is 6 to 9% between 20 and 57 km and 11% at 64 km. The vertical resolution (defined as the full width to half maximum of the averaging kernels) is 6 to 10 km between 20 and 50 km and about 13 km at 64 km.

[8] The MLS instrument uses heterodyne radiometers that observe the thermal emission from the atmospheric limb in broad spectral regions centered near 118, 190, 240, and 640 GHz and 2.5 THz. The primary ozone measurement is made with the 240 GHz radiometer, and additional measurements are also made in the 190 and 640 GHz and 2.5 THz bands. Ozone comparisons using the initial publicly released version 1.51 data set indicated that these data had a useful maximum upper limit of about 54 km [*Froidevaux et al.*, 2006], with significant differences between the 240 GHz and 190 GHz band retrievals in the mesosphere. For the version 2.2 data used here, better calibration of the narrow digital autocorrelator spectrometer channels provides more confidence in the mesospheric ozone values [*Froidevaux et al.*, 2007].

[9] The Aura orbit is Sun-synchronous at 705 km altitude with 98° inclination, crossing the equator (north-going) at 1345 Local Solar Time. The MLS fields-of-view are directed forward and vertically scan the limb in the orbit plane. Latitude coverage for each orbit is 82°S to 82°N, and about 3500 vertical profiles are obtained every 24 hours. The expected single profile (i.e., nonaveraged) accuracy ranges from 5 to 10% between 16 and 60 km, increasing to about 40% at 70 km. The vertical resolution is about 3 km between 12 to 60 km, increasing to about 6 km above this height [*Froidevaux et al.*, 2007]. Information on the MLS instrument and calibration can be found in the work of *Waters et al.* [2006], and the retrieval algorithms are described by *Livesey et al.* [2006].

[10] The SAGE-II instrument was launched in October 1984 aboard the Earth Radiation Budget Satellite and continued making measurements through to August 2005. It is a seven-channel solar photometer using ultraviolet and visible channels between 0.38 and 1.0 μm in solar occultation mode to retrieve atmospheric profiles of ozone, water vapor, nitrogen dioxide, and aerosol extinction

[*McCormick*, 1987]. In typical observation mode it took 15 sunrise and 15 sunset measurements per day over a latitude range of 80°S to 80°N. The precision of an individual SAGE-II profile, at 1-km resolution, is 5% between 24 and 36 km, degrading to 7% at 48 km. The accuracy of the measurements is estimated to be 6% above 25 km [*Cunnold et al.*, 1989].

[11] The HALOE instrument was launched aboard the Upper Atmosphere Research Satellite in 1991 and continued making measurements through to November 2005. The experiment uses solar occultation to measure vertical profiles of seven species at four infrared wavelengths [*Russell et al.*, 1993]. Latitudinal coverage was from 80°S to 80°N and the altitude range of the ozone measurements extends from about 15 to 90 km. The HALOE ozone vertical resolution is 2.2 km and the single profile accuracy ranges from 25% at cloud tops to 9% at 30 km. It remains at 9% until the ozone minimum region is reached at around 80 km where it is 20% [*Brühl et al.*, 1996].

[12] The GOMOS instrument is a medium-resolution star-occultation limb viewing spectrometer operating in the ultraviolet, visible, and near-infrared (UV-VIS-NIR) spectral ranges [*Bertaux et al.*, 2000; *Kyrölä et al.*, 2004]. It was launched aboard Envisat and became operational in August 2002. Ozone profiles are retrieved over the altitude range 15 to 100 km, with a vertical resolution of 2 to 3 km. While GOMOS measures continuously, scattered solar light during daytime and twilight hours mean retrieved profiles during those times suffer from additional noise, and only those ozone profiles measured in a dark atmospheric limb condition are used in this study. Error estimates are strongly dependent on the star being looked at. For example, on stars with temperatures above 7000 K, and magnitude less than 1.9, *Kyrölä et al.* [2006] show precision values ranging from 2 to 7% between 20 and 40 km, and mostly less than 2% up to 70 km for nighttime retrievals.

[13] The MIPAS instrument, also on board Envisat, is a Fourier transform spectrometer detecting the Earth's limb emission in the midinfrared [*Fischer and Oelhaf*, 1996; *European Space Agency*, 2000]. Its operating wavelength range is from 4.15 μm to 14.6 μm. Ozone profiles are retrieved during both daytime and nighttime hours. The nominal altitude range is 6 to 68 km [*Raspollini et al.*, 2006], although an Upper Atmosphere observation mode can give ozone retrievals up to 100 km [*Gil-Lopez et al.*, 2005]. An error analysis by the University of Oxford MIPAS group, available at http://www.atm.ox.ac.uk/group/mipas/err/, gives a precision of 6 to 11% and accuracy of 8 to 15% between 21 and 60 km, increasing to 27% and 47%, respectively, at 68 km, based on a global composite of results for five atmospheric conditions.

3. MLS-MWR Comparisons

[14] At the time of writing, the reprocessing of the MLS data set using the latest version 2.2 algorithm is incomplete. Geographical and temporal collocation criteria are therefore set to ensure an adequate number of comparison pairs, especially at the top of the retrievals. For the comparisons presented below, MLS measurements collocated with the MWRs are defined as being within 2.5° latitude and 12° longitude of the site and made within 6 hours of the MWR measurement. In addition, data screening guidelines for stratospheric and mesospheric measurements, described by *Froidevaux et al.* [2007], are also applied. In the event that there is more than one MLS measurement fitting these criteria, the one closest in time to the MWR measurement is chosen. Each pair consists of either daytime (defined as solar zenith angle less than 90°) or nighttime (defined as solar zenith angle greater than 105°) profiles.

[15] Each MLS measurement is convolved with the averaging kernels of the MWR measurement as described by *Connor et al.* [1995],

$$x_{hc} = A\, x_h + (I - A)x_a \quad (2)$$

where x_{hc} is the convolved profile, x_a is the MWR a priori profile, x_h is the MLS profile, and A is the averaging kernel matrix. Equation (2) has purposely been set up in the form of (1) to eliminate the a priori contribution to the microwave measurement when calculating MLS-MWR differences. We normalize these differences by the MWR profile so they can be expressed in fractional form

$$(x_{hc} - x_r)/x_r = A(x_h - x_t)/x_r - e_r/x_r \quad (3)$$

where e_r are the errors in the MWR measurement, as in (1). In (3) the MWR a priori dependence appears only in the normalizing x_r, where it is a negligible second-order effect. Equations (2) and (3) are approximate because MLS also has finite resolution; however, it is sufficiently better than the MWR resolution that the resulting errors are negligible. The MLS profiles are interpolated, as a function of log(pressure), to the fixed MWR pressure grid, which has spacing of 0.125 in $\log_{10}P$ (about 2 km spacing in altitude).

[16] The (MLS-MWR)/MWR differences, as determined using (3), are expressed as the percent difference of the averaged collocated VMR profiles. These are shown in Figure 1. Figure 1 also includes the mean ozone VMR profiles from the two instruments to provide a context for the difference profiles. For Mauna Loa, the daytime difference profile is based on 279 to 305 comparison pairs up to 0.1 hPa (about 64 km) and 203 pairs at 0.07 hPa (about 66 km). There are 374 pairs for the nighttime comparison up to 0.1 hPa, decreasing to 161 pairs by 0.04 hPa (about 70 km). For Lauder, the daytime difference profile is based on 146 to 163 comparison pairs up to 0.1 hPa, and 92 pairs at 0.07 hPa. There are 202 to 209 pairs for the nighttime comparison up to 0.1 hPa, decreasing to 71 pairs by 0.04 hPa. The drop in the number of comparison pairs above 0.1 hPa, especially with the daytime comparisons, is a result of the cutoff criteria applied to the MWR profiles. Error bars shown in the plots are two times the standard error of the mean (i.e., two times the standard deviation of the individual differences divided by the square root of the number of pairs). Because the random errors integrate down when large numbers of pairs are averaged together, they become small compared to the systematic measurement errors. We therefore include, in the plots, dashed lines indicating the root-sum-square of the combined MLS and MWR systematic errors.

[17] The oscillatory nature of the difference profiles is a noticeable feature at both sites, especially Mauna Loa. This feature is also seen in comparisons between MWR measure-

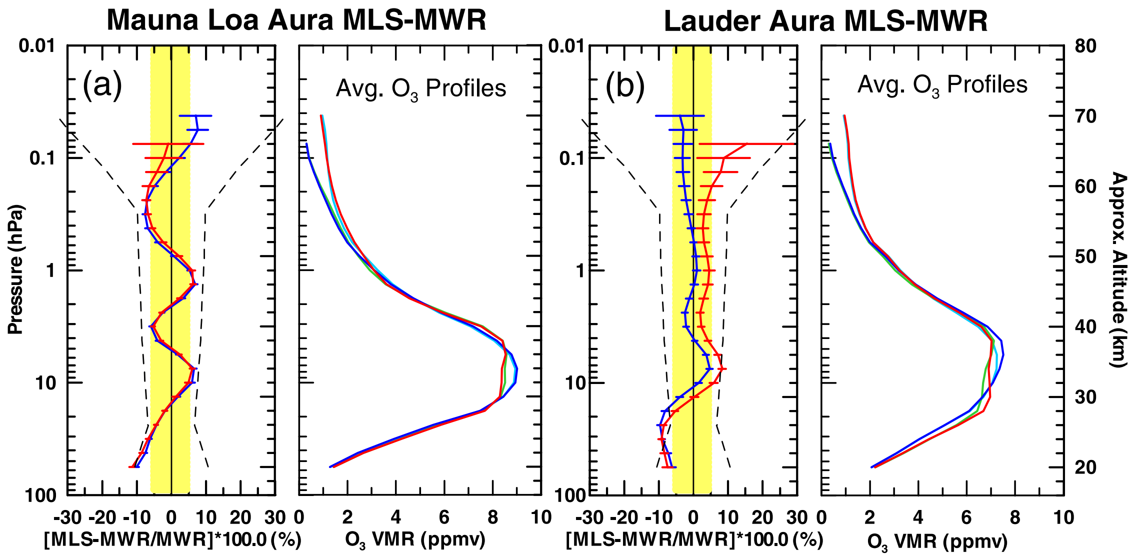

Figure 1. Microwave Limb Sounder (MLS)-microwave radiometer (MWR) ozone VMR difference profiles and mean ozone volume mixing ratio (VMR) profiles at (a) Mauna Loa and (b) Lauder. For the ozone VMR difference profiles: percentages are plotted against atmospheric pressure with MWR as the reference. Dashed lines are the root-sum-square of the combined MLS and MWR systematic errors. The right-hand abscissa gives approximate altitudes corresponding to the pressures. The red line shows the mean difference between daytime profiles and the blue line shows the mean difference between nighttime profiles. For the mean ozone VMR profiles, the dark blue line shows the MLS daytime profile, the cyan line shows the MLS nighttime profile, the green line shows the MWR daytime profile, and the red line shows the MWR nighttime profile.

ments and those made with instruments other than MLS, as shown in section 4, and can therefore be attributed to the MWR. Ground-based microwave measurements tend to produce retrievals with a small oscillatory component. Intuitively, this occurs because two ozone profiles, one of which has values that oscillate between being slightly greater than to slightly less than the values in the other will produce spectra that are so similar that they would be indistinguishable, given the noise and systematic errors contributed to measured spectra by imperfections in the instrument, and the fact that the instrument relies primarily on pressure broadening of the molecular emission to resolve altitude. More formally, *Connor et al.* [1995] calculate oscillatory error patterns that resemble those in Figure 1. The effects of systematic spectral measurement errors will propagate through the process of averaging multiple spectra and can produce artifacts in difference profiles such as those seen in Figure 1.

[18] Even with the oscillations, MLS-MWR agreement is generally within 5% at Mauna Loa between 30 and 0.04 hPa (about 24 to 70 km) and elsewhere within about 10%. Agreement is also mostly within 5% at Lauder, with exceptions being below 18 hPa, and, for the daytime comparison, above 0.15 hPa. Below 18 hPa, MLS ozone measurements are consistently low compared with the MWRs at both sites, but the discrepancy is larger at Lauder than at Mauna Loa in the 18 to 30 hPa region. A systematic error that contributes to the discrepancy at Lauder is discussed in more detail in the next section.

[19] The standard MLS pressure grid on which the ozone profile is retrieved has a level at 0.1 hPa, and the following level at 0.046 hPa. For daytime retrievals the errors reported along with the MLS ozone VMR values are increasing rapidly in the mesosphere because of the low ozone amounts in this region during daytime. They are generally greater, and often considerably greater, than 100% at 0.046 hPa. When converting to the MWR fixed pressure grid used here, MLS ozone VMRs at levels above 0.1 hPa are interpolated from results at 0.1 and 0.046 hPa and thus strongly influenced by the variability and large errors on the latter, as well as the reduced number of daytime MWR measurements available above 0.1 hPa.

[20] For the nighttime retrievals, absolute ozone amounts are higher and the corresponding MLS errors are generally lower at 0.046 hPa. At both Lauder and Mauna Loa, comparisons with the MWRs are close to or within 5% at the gridded pressure level of 0.042 hPa, and support the hypothesis that MLS nighttime measurements are suitable for scientific purposes up to at least 0.046 hPa. Above this, there are considerably fewer nighttime MWR measurements available for comparison, and more MLS measurements would be needed to provide reliable information on the accuracy of the retrievals.

4. Using the MWR Measurements as Transfer Standards

[21] In this section we compare a set of historical and current satellite-borne data sets to the MWR measurements and, by using the MWRs as transfer standards, look at the agreement between MLS and a consensus of the other instruments at Mauna Loa and Lauder. The selection of instruments is mostly limited to those that make daytime and/or nighttime measurements into the mesosphere to

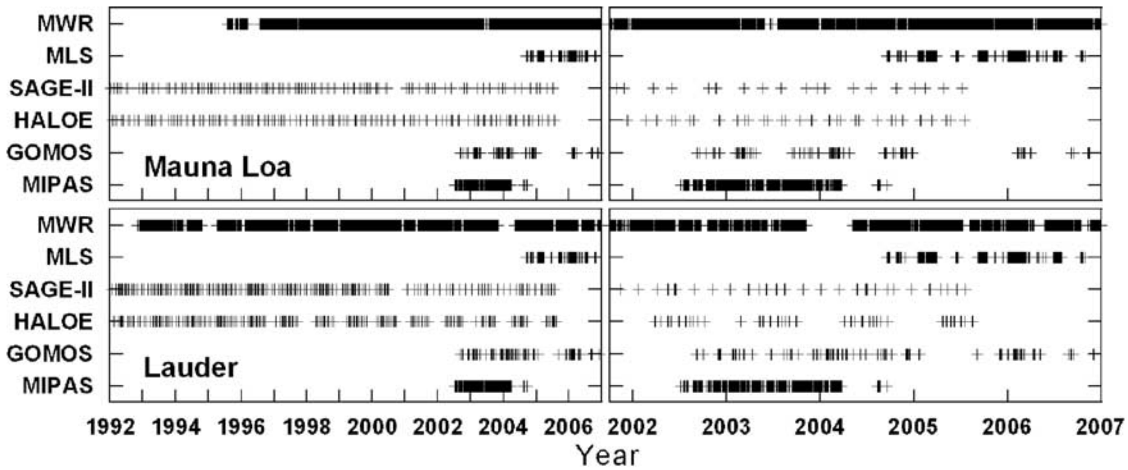

Figure 2. Times at which ozone measurements were made at Mauna Loa and Lauder with the instruments indicated at the left, with one cross per measurement. On the right is displayed the same result as those on the left but with an expanded timescale.

enable testing of the full upper range of the MLS measurements. The exceptions are SAGE-II and HALOE, which have records covering the periods 1984–2005 and 1991–2005, respectively, and made measurements at sunrise and sunset. Diurnal ozone variations in the upper stratosphere and mesosphere must be considered when comparing their measurements with the others. The HALOE retrieval incorporates a photochemical model intended to account for diurnal variation of ozone along the instrument's line of sight at sunrise and sunset, which is briefly described by *Natarajan et al.* [2005]. In section 5 we consider modeled and measured diurnal variations of ozone and conclude that the HALOE data can be used with caution in comparisons with other daytime measurements up into the mesosphere, and we do so here. Corrections are not made for diurnal variations along the line of sight in the SAGE-II ozone retrievals, and they are excluded from the comparisons above 1.0 hPa (about 48 km).

[22] The MWRs at Mauna Loa and Lauder have been measuring ozone, more or less continuously, for 12 and 15 a, respectively. The long periods of operation, and the stability of these measurements make the MWR data sets valuable, first, in contributing to the monitoring of regional and global long-term ozone trends [e.g., *Steinbrecht et al.*, 2006], and second, as a means of validating other ozone measurements. Because of the high temporal resolution of the MWRs, they can be used effectively as transfer standards, to allow comparison of data sets that would otherwise have limited or no coincident or collocated measurements.

[23] Figure 2 displays a time series of available collocated measurements for the instruments used in this study for (left) the period from 1992 to 2007 and (right) the period from 2002 to 2007, when the GOMOS, MIPAS, and MLS instruments came online. Several features are evident in Figure 2. As previously noted, the density of MWR measurements is high. The data gap in 1996 at Mauna Loa and the one from late 2003 to early 2004 at Lauder were caused by receiver failures. The Lauder instrument was removed from the site for use in an experiment elsewhere from late 1994 to April 1995, causing the gap at that time. While MLS generally records one or more daily observations close to both sites, only a limited number of MLS version 2.2 profiles are presently available. Collocated HALOE measurements at Lauder are seasonal in nature, more so after July 1996 when a battery failure aboard the satellite resulted in a reduced measurement schedule. While there are fewer HALOE measurements at Mauna Loa, they are more evenly spaced through the year. The density of the SAGE-II measurements was affected after July 2000, when a problem with the azimuth gimbal system occurred. This was corrected and operations resumed at a lower measurement frequency in November 2000. It can be seen, however, that there are few coincident measurements between MLS, HALOE and SAGE-II over both sites and none after mid-2005. The density of MIPAS measurements is high until the instrument was switched off in March 2004 owing to degradation of the interferometer. It was switched on for a short period in August and September 2004, with measurements made at a reduced spectral resolution, and started taking regular measurements again from January 2005 with reduced spectral resolution and a reduced measurement schedule. The measurements made since January 2005 are not yet publicly available, so there is no overlap between MIPAS and MLS measurements. The GOMOS instrument was offline from January to August 2005 owing to an instrument anomaly that caused it to go into a standby mode, resulting in a substantial gap in the overlap between GOMOS and MLS measurements. Since coming back online, it has been operated with a reduced azimuth field of view.

[24] To provide an indication of the stability of the MWR instruments and their suitability as transfer standards, Figure 3 displays, as a function of pressure, long-term trends in the MWR measurements relative to four other collocated or coincident data sets at each site. The measurements used are those from the National Institute for Public Health and the Environment (RIVM) lidar [*Brinksma et al.*, 2000] and the National Institute of Water and Atmospheric Research Ltd (NIWA) ozonesondes [*Bodeker et al.*, 1998] at Lauder, the Jet Propulsion Laboratory (JPL) lidar [*McDermid et al.*, 1991] at Mauna Loa, and the National Oceanic and Atmospheric Administration (NOAA) ozone-

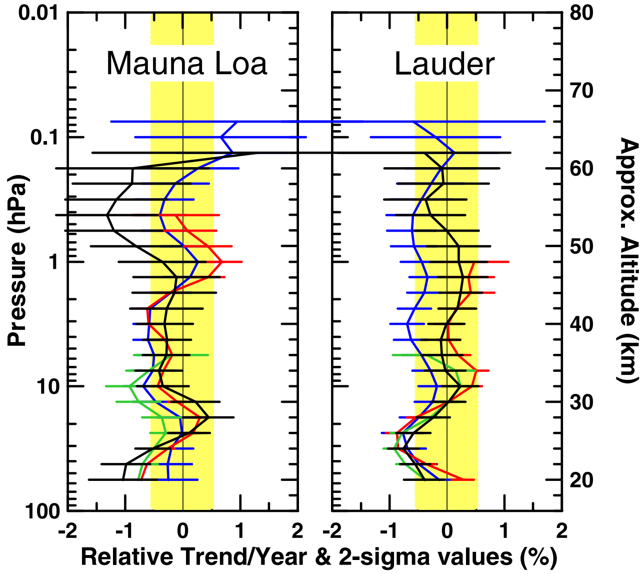

Figure 3. Long-term relative trends, in percent per year, between the MWRs and ozonesondes (green), lidars (red), SAGE-II (black), and HALOE (blue) at (left) Mauna Loa and (right) Lauder. The periods covered for the comparisons at Mauna Loa and Lauder are, in turn: Ozonesondes, 1998–2006, 1992–2006; Lidars, 1995–2006, 1995–2006; SAGE-II, 1995–2005, 1992–2005; HALOE, 1995–2005, 1992–2005.

sondes [*Johnson et al.*, 2002] launched from Hilo, Hawaii, along with SAGE-II and HALOE. The relative trends are obtained by means of linear regression applied to time series of monthly averaged differences between the MWR and comparison measurements at each pressure level. All comparison measurements have been convolved with the averaging kernels of the MWR measurements. Collectively, the trends are less than 0.5% per year and not significant at the 2-sigma level. Except between 42 and 18 hPa at Lauder there is little consistency between the results for the several comparison instruments, suggesting that the scatter among the results is due to the combinations of absolute drifts in the MWR and the comparison measurements. The apparent drift of close to −1%/year seen consistently in all four comparisons in Figure 3 at Lauder between 42 and 18 hPa is a result of a discontinuity in the MWR time series at these levels occurring when the instrument was repaired and brought back online after a receiver failure in 2004. The steps in the time series are on the order of a 3 to 8% increase in ozone VMR after the repair. We are currently working on a technique to include a direct measurement of the systematic effect that caused the discontinuity in the MWR calibration procedure.

[25] For the purposes of this study, therefore, we have scaled the Lauder MWR measurements, from May 2004 onward, between 42 and 18 hPa to account for the discontinuity. This is achieved by comparing measurements made between the MWR and three other instruments before November 2003, with equivalent comparisons made after May 2004. The three data sets that adequately span this period are those from the RIVM lidar, the NIWA ozonesondes, and the collocated GOMOS time series. Table 1 shows the steps in the MWR time series derived from these comparisons, expressed as percentages, with the last column showing the average of the three values. To create a homogenized time series, the MWR measurements made after May 2004 have been scaled downward by the amounts given in the last column. Note that the MWR measurements used in the Lauder MLS-MWR comparisons shown previously in section 3 have not been scaled in any way.

[26] Instrument comparisons with the MWR are made using the same procedures and criteria as those for the MLS-MWR comparisons in the previous section. Apart from the region between 42 and 18 hPa at Lauder, the MLS-MWR difference profiles are the same as those in Figure 1. Except for SAGE-II, daytime and nighttime measurements are compared separately. In the case of HALOE, the twilight measurements are compared with MWR daytime measurements. For SAGE-II, no day/night demarcation is necessary as the comparisons are stopped at 1.0 hPa, before diurnal effects become significant. All the satellite-borne instruments have relatively high vertical resolutions compared to the MWRs and have been convolved using the MWR averaging kernels. Error bars shown in the plots are as described in the previous section.

[27] The quality of the nighttime GOMOS retrieval can be affected by the equivalent blackbody temperature of the star being used, its visual magnitude, and the line of sight azimuth between the instrument and the star being pointed at [*Meijer et al.*, 2004; *Kyrölä et al.*, 2006]. The above authors point out that poor quality retrievals, especially above 45 km, are seen with measurements on stars with weak brightness, which adversely affect signal-to-noise ratios, as well as on cool stars, which emit mainly at visible wavelengths, to which the upper atmosphere is mostly transparent. To exclude such profiles from the comparisons, measurements selected here are those made on stars with temperatures greater than 4500 K and, if the temperature is less than 8000 K, a visual magnitude of 1.9 or better. For the other data sets it was not necessary to include any further data screening when selecting profiles.

[28] Table 2 summarizes information regarding the data sets used in this study, including the processing version number, the period during which comparisons were made, and the number of collocated pairs used in determining the average daytime and nighttime difference profiles presented here. Three processing versions are in use in the MIPAS operational data set provided through the European Space Agency (ESA). Versions 4.61 and 4.62 are considered equivalent for validation purposes [*Cortesi et al.*, 2006] and cover the period from launch to March 2004, when the instrument made measurements in full resolution mode.

Table 1. Percentage Steps in the Lauder Microwave Radiometers (MWR) Time Series in 2004, Based on Comparisons With RIVM Lidar, NIWA Ozonesonde, and Envisat GOMOS

Pressure, hPa	Lidar-MWR Step, %	Sonde-MWR Step, %	GOMOS-MWR Step, %	Average Step, %
42	3.1	3.5	2.8	3.1
32	7.6	8.4	7.6	7.9
24	7.7	9.2	7.6	8.2
18	3.7	5.5	3.2	4.1

Table 2. Summary of Instrument Comparisons With the MWRs

Instrument	Version	Day/Night	Period		Number of Pairs[a]	
			Mauna Loa	Lauder	Mauna Loa	Lauder
MLS	2.2	D	2004.7–2006.8	2004.7–2006.6	279–305/203	146–163/92
MLS	2.2	N	2004.7–2006.9	2004.7–2006.6	374/161	202–209/71
SAGE-II	6.2	D/N	1995.6–2005.6	1992.8–2005.5	66	181
HALOE	19	D	1996.0–2005.6	1992.8–2005.7	68–70/16	128–129/24
GOMOS	6.0f	N	2002.6–2006.9	2002.6–2007.0	79–92/42	52–54/21
MIPAS	4.61/2/5	D	2002.5–2004.8	2002.5–2004.7	271–300/87	139–153/47
MIPAS	4.61/2/5	N	2002.5–2004.7	2002.5–2004.7	273–278/152	108–130/56

[a]First value, or range of values, are the number of pairs up to 0.1 hPa, the last value (where present) is the number of pairs at the top of the profile.

Version 4.65 covers the period August to September 2004 and takes into account the reduced spectral resolution.

[29] Results from the comparisons between the various instruments and the MWRs are presented in Figure 4, where Figure 4a shows results for Mauna Loa daytime comparisons, Figure 4b shows results for Mauna Loa nighttime comparisons, Figure 4c shows results for Lauder daytime comparisons, and Figure 4d shows results for Lauder nighttime comparisons. On the left side of each figure difference profiles between each of the instruments and the MWRs are shown. On the right are the resulting MLS-consensus differences, determined by averaging the percentage difference profiles on the left (except for MLS-MWR), at each pressure level, to obtain a single difference profile, which is then subtracted from the MLS-MWR difference profile. The error bars on the right denote the standard error of the mean and are used here to give an indication of where the consensus diverges and agrees. Note that the error bars and the following descriptions of the results ignore some systematic effects, discussed in section 5, which may affect the comparison data in the mesosphere.

[30] At Mauna Loa the MLS consensus agreement is remarkably good, generally better than 3%, from 56 to 0.4 hPa (about 20 to 54 km). Above 0.4 hPa, MLS is biased low by between 3 and 13% compared to the consensus. In Figure 4b, and to a lesser extent in Figure 4a, the negative bias is reinforced by the positive MIPAS-MWR differences. MLS measures lower than all the other instruments between 0.5 and 0.1 hPa (about 52 to 64 km), however the agreement between the MLS-MWR and GOMOS-MWR difference profiles up to 0.07 hPa is within 5%. The agreement between the MLS-MWR and HALOE-MWR profiles is within 5% to 0.2 hPa (about 60 km), and 10% above this, and agreement between MLS and MWR is within 6% above 0.2 hPa.

[31] A similar picture is evident with the Lauder comparisons. For the daytime comparisons MLS-consensus agreement is better than 5% below 0.1 hPa (about 64 km). For the nighttime comparisons, MLS agreement with the consensus is within 3% below 0.4 hPa (about 54 km) and has a negative bias of between 3 and 8% above this. The positive differences between MIPAS and the MWR again contribute to this result. In Figure 4d, MLS agreement with MWR is within 5% above 40 hPa (about 22 km), and the MLS-MWR and GOMOS-MWR profiles are within 5% of each other through the entire profile.

[32] The agreement between MLS and historical SAGE-II data is remarkably good, 2 to 3%, from 56 to 0.1 hPa (about 20 to 48 km) at Mauna Loa. It is not quite as good at Lauder but still mostly better than 5% over this range.

5. Discussion on the Use of the Satellite Data Sets

[33] Diurnal ozone variations must be considered when comparing the solar occultation HALOE measurements with those of the other instruments used in this work, as they are made at sunrise or sunset while the comparison measurements are nearly all made fully in daylight or in darkness. MWR measurements at 34°N [*Connor et al.*, 1994] and photochemical models [e.g., *Natarajan et al.*, 2005] show that the most prominent features of the diurnal variations consist of a rapid decrease in ozone just before sunrise and a rapid increase just after sunset (i.e., at solar zenith angles of 90° at the tangent point locations of the occultation measurements). The occultation measurements thus correspond most closely to daytime measurements made with the other instruments. The amplitude of the diurnal cycle relative to the nighttime amount becomes significant at about 50 km and increases with altitude up to at least 70 km. For example, the ratio of nighttime to daytime ozone values is about 1.2 at 55 km, 1.5 at 60 km, and 2.5 at 65 km [*Siskind et al.*, 1995]. At the resolution of the MWR measurements (about 13 km at these levels), the behavior of the diurnal cycle during daytime generally consists of a slight rise for about 2 hours after sunrise followed by a slight, slow decline through nearly the rest of the day. We have not selected the times of the daytime measurements, so they are best represented by the average of the diurnal variation during daytime. Compared to this, ozone amounts at sunset are usually larger than the immediately proceeding daytime amounts (about 15% at 62 km), as the diurnal sunset increase in ozone is just beginning. So the daytime measurements may slightly underestimate the ozone amounts at the times of sunset occultation measurements. The sunrise values may be either slightly greater or slightly less than the daytime average depending on altitude.

[34] Diurnal ozone variations must also be taken into account when calculating tangent point ozone values from the physical quantities measured by an occultation instrument. In the case of HALOE, a photochemical model is incorporated into the retrieval algorithm for this purpose. However, *Natarajan et al.* [2005] point out that this model only extends up to 61 km and that errors are introduced in the HALOE results because the stronger diurnal variations at the higher altitudes that the line of sight passes through on the near and far sides of the tangent point are not taken

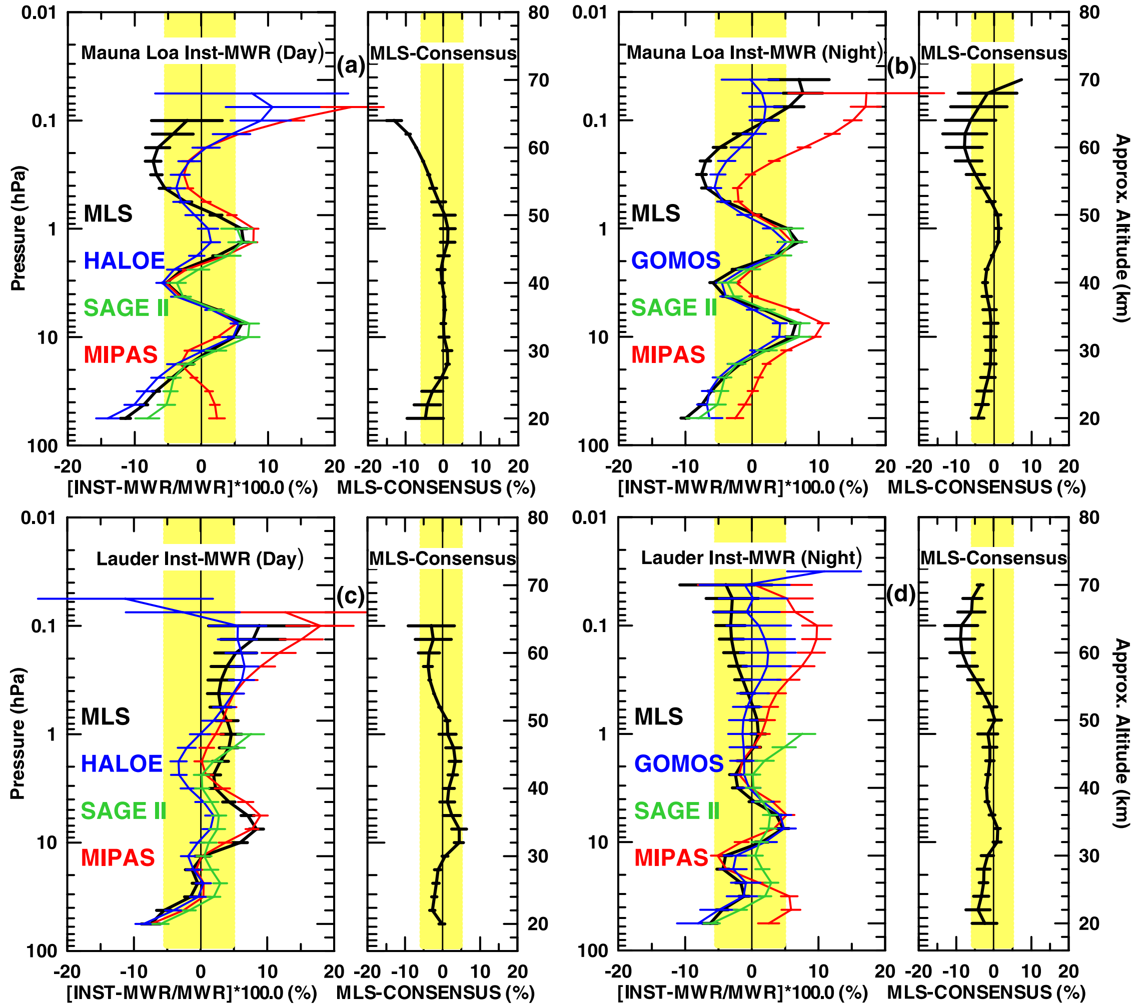

Figure 4. (a) Instrument-MWR ozone VMR daytime difference profiles at Mauna Loa, given as percentages plotted against atmospheric pressure, with MWR as the reference. Black line shows MLS-MWR, blue line shows HALOE-MWR, red line shows MIPAS-MWR, and green line shows SAGE II-MWR. (b) Instrument-MWR ozone VMR nighttime difference profiles at Mauna Loa, given as percentages plotted against atmospheric pressure, with MWR as the reference. Black line shows MLS-MWR, blue line shows GOMOS-MWR, red line shows MIPAS-MWR, and green line shows SAGE II-MWR. (c) Same as Figure 4a except for Lauder. (d) Same as Figure 4b except for Lauder. On the right-hand sides of Figures 4a–4d are shown resulting MLS-consensus difference profiles, when the non-MLS profiles from the corresponding left-hand sides are averaged, and subtracted from the MLS-MWR difference profile.

into account. When they do that, they find that their retrieved HALOE values decrease somewhat (20 to 25% at 65 to 70 km in one example) compared to the standard v19 product for sunrise but only slightly for sunset observations.

[35] Comparison of HALOE with MWR profiles should be affected both by the difference between sunset and daytime ozone values and by the diurnal effects on HALOE sunrise observations that are not accounted for in the standard product. These effects should both produce negative values in HALOE-MWR difference profiles (one at sunrise and the other at sunset) at altitudes where the effects are significant. However, the only indication that such effects may be occurring is the sharp negative shift in the profile at Lauder above about 64 km. According to the work of *Natarajan et al.* [2005], the sunrise systematic error should approach its maximum value by about 61 km. The measurements reported by *Connor et al.* [1994] also show sunset ozone levels about 15% higher than daytime levels at about 61 km. However, these results from the literature may not be directly applicable to the work reported herein as both were done at different latitudes. Unfortunately, measurements of diurnal variations are not routinely derived from the MWR observations at the present instrument locations. Given the limited measured and model data available, we can presently only conclude that there may be errors that affect the top of the HALOE-MWR profile.

[36] MIPAS derives ozone profiles from spectral measurements of atmospheric emission in the midinfrared from a limb view of the atmosphere. The Oxford MIPAS group has

posted an error analysis applicable to the ESA operational level 2A ozone product at http://www.atm.ox.ac.uk/group/mipas/err/, which is based on work described by *Dudhia et al.* [2002]. An error budget is given on the website and it indicates that several error components may be systematic in character, that is, not likely to change in sign over an averaging period of a number of months. These are related to the assumption of local thermodynamic equilibrium (LTE) in the operational retrieval, the errors in the calculated spectral data, and the calibration of the MIPAS radiometric gain, instrumental line shape, and assigned wavelengths. Generally, these components tend to increase above about 50 km, and the non-LTE and instrumental line shape (ILS) components dominate above 60 km, each reaching 10% at 65 and 68 km, respectively. Of the above, only the non-LTE error has presently been given a sign (C. Piccolo, Oxford University, private communication, 2007). It would generally cause ozone amounts to be overestimated in the absence of other errors. The ILS error could have either sign. The combination of these two components would thus make it more likely that MIPAS overestimates than underestimates ozone above about 60 km.

[37] The MLS-consensus results are undoubtedly more uncertain above about 56 km than below. While all the instruments are affected by increasing errors above this height, the MLS-MWR results, bolstered by the continuing GOMOS-MWR agreement, and the probability that the MIPAS amounts are overestimated, suggest that MLS may be in better agreement with the consensus than shown in Figure 4.

6. Conclusions

[38] This study presents an assessment of MLS v2.2 ozone retrievals from the midstratosphere to the mesosphere by comparing collocated MLS measurements with those from ground-based MWRs located at Lauder, New Zealand, and Mauna Loa, Hawaii. We then use the MWRs as references to determine differences between MLS and a consensus derived from HALOE, SAGE-II, GOMOS, and MIPAS measurements over the NDACC stations at Mauna Loa and Lauder.

[39] The MLS measurements agree with those made with the NDACC MWR at Mauna Loa to generally within 5% from 24 to 0.04 hPa (about 26 to 70 km) and within about 10% down to 56 hPa. Agreement between MLS and NDACC MWR measurements at Lauder is similar, except that 7 to 10% differences extend from about 56 to 18 hPa (about 20 to 28 km) because of an additional systematic error component that appears in the measurements following a repair of the Lauder instrument carried out in 2004. The oscillatory nature of the MLS-MWR differences is a characteristic of the MWR measurements at both NDACC sites.

[40] The MLS measurements agree remarkably well between 56 and 0.4 hPa (about 20 to 54 km) with the consensus of HALOE, SAGE-II, GOMOS, and MIPAS measurements at both the Mauna Loa and Lauder locations, generally within 3%. Above this, the agreement between MLS and the consensus of GOMOS, MIPAS, and HALOE degrades and also becomes less certain, with negative differences reaching 10%. Measurements of diurnal ozone variations at the MWR sites, improved modeling of diurnal effects on the HALOE data, and assessment of residual MIPAS instrument characterization uncertainties for possible systematic components are all needed to better understand comparison results at these altitudes. Nonetheless, MLS, MWR, and GOMOS measurements remain consistent within about 5% up to at least 0.1 hPa (about 64 km) during the day and 0.04 hPa (about 70 km) at night. HALOE also agrees well with MLS and MWR, but this result is weakened in the absence of better knowledge of diurnal effects. MIPAS measurements may also be consistent to the same level after accounting for non-LTE effects.

[41] **Acknowledgments.** We wish to acknowledge and thank the teams at JPL, RIVM, NIWA, and NOAA for making their lidar and ozonesonde data sets available through the Network for the Detection of Atmospheric Composition Change (NDACC). We thank the European Space Agency for services afforded in making available the GOMOS and MIPAS measurements. We also thank Bojan Bojkov of the Aura Validation Data Center (AVDC) and the MLS Data Distribution Team for access to the MLS data set (see http://avdc.gsfc.nasa.gov). Finally, we thank the University of Oxford team for communicating unpublished information on the MIPAS error budget. Work at the Jet Propulsion Laboratory, California Institute of Technology, was carried out under a contract with the National Aeronautics and Space Administration. This paper is based upon work supported by NASA under award NNG05GN46G.

References

Bertaux, J. L., E. Kyrölä, and T. Wehr (2000), Stellar occultation technique for atmospheric ozone monitoring: GOMOS on Envisat, *Earth Obs. Q.*, *67*, 17–20.

Bodeker, G. E., I. S. Boyd, and W. A. Matthews (1998), Trends and variability in vertical ozone and temperature profiles measured by ozonesondes at Lauder, New Zealand: 1986–1996, *J. Geophys. Res.*, *103*(D22), 28,661–28,682.

Brühl, C., et al. (1996), HALOE ozone channel validation, *J. Geophys.Res.*, *101*(D6), 10,217–10,240.

Brinksma, E. J., et al. (2000), Validation of 3 years of ozone measurements over Network for the Detection of Stratospheric Change station Lauder, New Zealand, *J. Geophys. Res.*, *105*(D13), 17,291–17,306.

Connor, B. J., D. E. Siskind, J. J. Tsou, A. Parrish, and E. E. Remsberg (1994), Ground-based microwave observations of ozone in the upper stratosphere and mesosphere, *J. Geophys. Res.*, *99*(D8), 16,757–16,770.

Connor, B. J., A. Parrish, J.-J. Tsou, and M. P. McCormick (1995), Error analysis for the ground-based microwave ozone measurements during STOIC, *J. Geophys. Res.*, *100*(D5), 9283–9292.

Cortesi, U., et al. (2006), Co-ordinated validation activity and quality assessment of MIPAS-ENVISAT ozone data, Atmospheric Science Conference, Eur. Space Agency, Frascati, Italy, 8–12 May.

Cunnold, D. M., W. P. Chu, R. A. Barnes, M. P. McCormick, and R. E. Veiga (1989), Validation of SAGE II ozone measurements, *J. Geophys. Res.*, *94*(D6), 8447–8460.

Dudhia, A., V. L. Jay, and C. D. Rodgers (2002), Microwindow selection for High-Spectral-Resolution Sounders, *Appl. Opt.*, *41*, 3665–3673.

European Space Agency (2000), Envisat: MIPAS, An instrument for atmospheric chemistry and climate research, *ESA SP-1229*, Noordwijk, Netherlands.

Fischer, H., and H. Oelhaf (1996), Remote sensing of vertical profiles of atmospheric trace constituents with MIPAS limb emission spectrometers, *Appl. Opt.*, *35*, 2787–2796.

Froidevaux, L., et al. (2006), Early validation analyses of atmospheric profiles from EOS MLS on the Aura satellite, *IEEE Trans. Geosci. Remote Sens.*, *44*(5), 1106–1121.

Froidevaux, L., et al. (2007), Validation of Aura Microwave Limb Sounder stratospheric ozone measurements, *J. Geophys. Res.*, doi:10.1029/2007JD008724, in press.

Gil-López, S., et al. (2005), Retrieval of stratospheric and mesospheric O_3 from high resolution MIPAS spectra at 15 and 10 μm, *Adv. Space Res.*, *36*, 943–951.

Hocke, K., et al. (2007), Comparison and synergy of stratospheric ozone measurements by satellite limb sounders and the ground-based microwave radiometer SOMORA, *Atmos. Chem. Phys. Disc.*, *7*, 4117–4131.

Johnson, B. J., S. J. Oltmans, H. Vömel, H. G. J. Smit, T. Deshler, and C. Kröger (2002), Electrochemical concentration cell (ECC) ozonesonde pump efficiency measurements and tests on the sensitivity to ozone of buffered and unbuffered ECC sensor cathode solutions, *J. Geophys. Res.*, *107*(D19), 4393, doi:10.1029/2001JD000557.

Kyrölä, E., et al. (2004), GOMOS on Envisat: An overview, *Adv. Space Res.*, *33*, 1020–1028.

Kyrölä, E., et al. (2006), Nighttime ozone profiles in the stratosphere and mesosphere by the Global Ozone Monitoring by Occultation of Stars on Envisat, *J. Geophys. Res.*, *111*, D24306, doi:10.1029/2006JD007193.

Livesey, N. J., W. V. Snyder, W. G. Read, and P. A. Wagner (2006), Retrieval algorithms for the EOS Microwave Limb Sounder (MLS) instrument, *IEEE Trans. Geosci. Remote Sens.*, *44*(5), 1144–1155.

McCormick, M. P. (1987), SAGE II: An overview, *Adv. Space. Res.*, *7*(3), 219–226.

McDermid, I. S., D. A. Haner, M. M. Kleiman, T. D. Walsh, and M. L. White (1991), Differential absorption Lidar systems at JPL-TMF for tropospheric and stratospheric ozone measurements, *Opt. Eng.*, *30*, 22–30.

Meijer, Y. J., et al. (2004), Pole-to-pole validation of Envisat GOMOS ozone profiles using data from ground-based and balloon sonde measurements, *J. Geophys. Res.*, *109*, D23305, doi:10.1029/2004JD004834.

Natarajan, M., L. E. Deaver, E. Thompson, and B. Magill (2005), Impact of twilight gradients on the retrieval of mesospheric ozone from HALOE, *J. Geophys. Res.*, *110*, D13305, doi:10.1029/2004JD005719.

Parrish, A. (1994), Millimeter-wave remote sensing of ozone and trace constituents in the stratosphere, *Proc. IEEE*, *82*, 1915–1929.

Parrish, A., B. J. Connor, W. P. Chu, J. J. Tsou, and I. S. McDermid (1992), Ground-based microwave monitoring of stratospheric ozone, *J. Geophys. Res.*, *97*(D2), 2541–2546.

Raspollini, P., et al. (2006), MIPAS level 2 operational analysis, *Atmos. Chem. Phys.*, *6*, 5605–5630.

Rodgers, C. D. (1976), Retrieval of atmospheric temperature and composition from remote measurements of thermal radiation, *Rev. Geophys.*, *14*, 609–624.

Russell, J. M., III, L. L. Gordley, J. H. Park, S. R. Drayson, W. D. Hesketh, R. J. Cicerone, A. F. Tuck, J. E. Frederick, J. E. Harries, and P. J. Crutzen (1993), The Halogen Occultation Experiment, *J. Geophys. Res.*, *98*(D6), 10,777–10,797.

Siskind, D. E., B. J. Connor, R. S. Eckman, E. E. Remsberg, J. J. Tsou, and A. Parrish (1995), An intercomparison of model ozone deficits in the upper stratosphere and mesosphere from two data sets, *J. Geophys. Res.*, *100*(D6), 11,191–11,202.

Steinbrecht, W., et al. (2006), Long-term evolution of upper stratospheric ozone at selected stations of the Network for the Detection of Stratospheric Change (NDSC), *J. Geophys. Res.*, *111*, D10308, doi:10.1029/2005JD006454.

Waters, J. W., et al. (2006), The Earth Observing System Microwave Limb Sounder (EOS MLS) on the Aura satellite, *IEEE Trans. Geosci. Remote Sens.*, *44*(5), 1075–1092.

I. S. Boyd, NIWA Environmental Research Institute, University of Massachusetts, 710 North Pleasant Street, Amherst, MA 01003, USA. (boyd@fcrao1.astro.umass.edu)

L. Froidevaux, Jet Propulsion Laboratory, MS183-701, 4800 Oak Grove Drive, Pasadena, CA 91109, USA. (lucien.froidevaux@jpl.nasa.gov)

E. Kyrölä, Earth Observation, Finnish Meteorological Institute, P. O. Box 503, FIN-00101 Helsinki, Finland. (erkki.kyrola@fmi.fi)

A. D. Parrish, Department of Astronomy, University of Massachusetts, 710 North Pleasant Street, Amherst, MA 01003, USA. (parrish@astro.umass.edu)

J. M. Russell III, Center for Atmospheric Sciences, Hampton University, Hampton, VA 23668, USA. (james.russell@hamptonu.edu)

T. von Clarmann, Forschungszentrum Karlsruhe, Institut für Meteorologie und Klimaforschung, P. O. Box 3640, D-76021 Karlsruhe, Germany. (thomas.clarmann@imk.fzk.de)

J. M. Zawodny, NASA Langley Research Center, Mail Stop 420, Hampton, VA 23681, USA. (joseph.m.zawodny@nasa.gov)