

## Results from the 1995 Stratospheric Ozone Profile Intercomparison at Mauna Loa

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**Abstract.** In August 1995, multiple instruments that measure the stratospheric ozone vertical distribution were intercompared at the Mauna Loa Observatory, Hawaii, under the auspices of the Network for the Detection of Stratospheric Change. The instruments included two UV lidar systems, one from the Jet Propulsion Laboratory and the other from Goddard Space Flight Center, electrochemical concentration cell balloon sondes, a ground-based microwave instrument, Dobson-based Umkehr measurements, and a new ground-based Fourier transform infrared instrument. The Microwave Limb Sounder instrument on the Upper Atmosphere Research Satellite provided correlative profiles of ozone, and there was one close overpass of the Stratospheric Aerosol and Gas Experiment II (SAGE II) instrument. The results show that much better consistency among instruments is being achieved than even a few years ago, usually to within the instrument uncertainties. The different measurement techniques in this comparison agree to within  $\pm 10\%$  at almost all altitudes, and in the 20–45 km region most agreed within  $\pm 5\%$ . The results show that the current generation of lidars is capable of accurate measurement of the ozone profile to a maximum altitude of 50 km. SAGE II agreed well with both lidar and balloon sonde down to at least 17 km. The ground-based microwave measurement agreed with other measurements from 22 km to above 50 km. One minor source of disagreement continues to be the pressure-altitude conversion needed to compare a measurement of ozone density versus altitude with a measurement of ozone mixing ratio versus pressure.

### 1. Introduction

Significant changes in total column ozone have been documented [Stolarski *et al.*, 1991; World Meteorological Organization (WMO), 1995], but there is an open question as to the altitude at which the changes are occurring. Through a program of systematic comparison and intercalibration, the ground-based and satellite-based measurements of total col-

umn ozone have been brought into basic agreement, usually to within 2–3%. The measurement of the ozone vertical distribution is much more uncertain, with disagreements of 10–30% or more [Harris *et al.*, 1998]. In order to clearly establish the altitude dependence of ozone change, the profile measurement techniques need to be brought into agreement through a series of intercomparisons. Such intercomparisons are being supported by the Network for the Detection of Stratospheric Change (NDSC), which is charged with monitoring long-term changes in stratospheric ozone and in the species that control ozone.

In the 1995 NDSC Stratospheric Ozone Profile Intercomparison at Mauna Loa (MLO3), a number of different instruments were compared at the Mauna Loa Observatory, hereinafter referred to as MLO, on Hawaii (19.5°N latitude, 155.6°W longitude, 3.4 km above mean sea level). The purpose of MLO3 is to provide data to assess the capabilities and to check the consistency of the participating instruments in determining ozone profiles. The comparison was done as a blind intercomparison following the protocol established by the NDSC. The campaign was under the control of an impartial referee (the lead author of this paper), who was responsible for handling all the data so that, as far as possible, the participants did not see each other's results during the campaign. The measurement period began on August 15, 1995, and ended on September 1, 1995. The final processed data for every instrument were submitted to the referee within 1 month of the end of the campaign. MLO3 was a follow-up to the Ozone Profiler at Lauder (OPAL) intercomparison [McDermid *et al.*, 1998], which was

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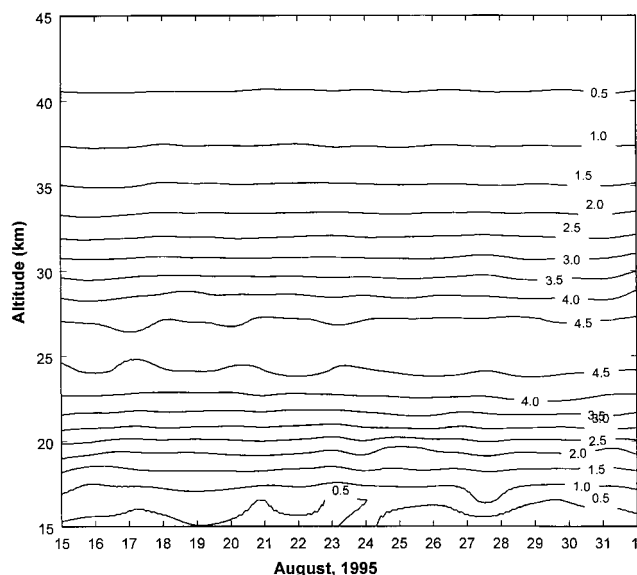
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**Figure 1.** Ozone variability during the Stratospheric Ozone Profile Intercomparison at Mauna Loa (MLO3) comparison as observed by lidar. Ozone number density ( $\times 10^{12}$  molecules  $\text{cm}^{-3}$ ) is plotted as a function of altitude and time.

done at Lauder, New Zealand, in April of 1995, and to the Stratospheric Ozone Intercomparison Campaign (STOIC) comparison held at Table Mountain in 1989 [Margitan *et al.*, 1995]. Lauder is the primary NDSC site for monitoring the stratosphere at southern midlatitudes, while Mauna Loa is the primary NDSC site for the tropics and subtropics. Mauna Loa was chosen as the intercomparison site because it is a very clean, low-aerosol marine environment and because ozone variability is very low in the subtropics (see Figure 1). Low variability minimizes the uncertainty caused by the fact that not exactly the same air volume is measured by every instrument.

## 2. Measurement Systems

Information on the participants and the measurements is given in Table 1. Two UV lidar systems, one from the Jet Propulsion Laboratory (JPL) and the other from Goddard Space Flight Center (GSFC), measure ozone number density as a function of altitude from 15 km to above 50 km altitude. Since lidar promises to be an important technique for long-

term monitoring of ozone in the future, the performance of the lidar systems was of particular interest. Electrochemical concentration cell (ECC) balloon sondes, including several “triples,” were flown daily to obtain profiles of ozone partial pressure along with pressure and temperature from the ground to above 35 km. The Millitech/Langley Research Center microwave radiometer measures ozone mixing ratio as a function of pressure from 56 to 0.1 hPa. Dobson instruments provided daily measurements of total column ozone and were used to make Umkehr measurements of the ozone profile. The Microwave Limb Sounder (MLS) instrument on the Upper Atmosphere Research Satellite (UARS) provided correlative profiles of ozone mixing ratio versus pressure between 100 and 0.2 hPa. One close overpass of Stratospheric Aerosol and Gas Experiment II (SAGE II) on August 30 provided an ozone number density profile from 15 to 55 km. A few measurements were obtained from a prototype Fourier transform infrared (FTIR) instrument being developed at the University of Denver.

While measurements were taken by the solar backscattered ultraviolet (SBUV/2) instrument on NOAA 14 during MLO3, the results have not been used in this comparison. The failure of the cloud cover radiometer on SBUV/2 just after launch in the spring of 1995 led to a mode change during MLO3. Questions about the initial calibration plus uncertainty about the mode change led to the decision to not use these data in this comparison. Data from the Halogen Occultation Experiment (HALOE), also on UARS, would have been a valuable addition to the comparison, but unfortunately the instrument was not operating during this 2 week period.

### 2.1. JPL Differential Absorption Lidar

The JPL differential absorption lidar (DIAL) system [McDermid *et al.*, 1995] consists of a 100 W, narrow bandwidth, tunable, XeCl excimer laser providing a main beam at 307.9 nm. The reference wavelength at 353.2 nm is generated by stimulated Raman shifting of a portion of the main beam in a 400 pounds per square inch gage (psig) hydrogen cell. The two beams are transmitted simultaneously, and the backscattered radiation is collected with a 90 cm telescope and measured using photon-counting techniques. To extend the dynamic range of the system (and the altitude range of the retrieved profile), the signal is further divided in the ratio 100:1 and directed through separate detection chains. The high-intensity data are used to obtain the high-altitude part of the profile, while the low-intensity data are used for the lower altitudes. A

**Table 1.** Participants in MLO3

Instrument	Participants	Measurement
Goddard lidar	T. McGee and M. Gross	O <sub>3</sub> ND versus altitude; 14–50 km at 0.15 km
JPL lidar	S. McDermid	O <sub>3</sub> ND versus altitude; 14–50 km at 0.3 km
Ozonesondes	D. Hofmann and B. Johnson	O <sub>3</sub> MR versus pressure; temperature versus pressure; 0–35 km at 0.15 km
Microwave radiometer	J. J. Tsou, B. Connor, and A. Parrish	O <sub>3</sub> MR versus pressure; 20–65 km at ~2 km
Umkehr	G. Koenig, S. Oltmans, and M. Newchurch	O <sub>3</sub> MR versus pressure; 15–43 km at ~5 km
FTIR	F. Murcray	O <sub>3</sub> MR versus pressure; 5–32 km at ~4 km
MLS	L. Froidevaux	O <sub>3</sub> MR versus pressure; 18–60 km at ~2.5 km
SAGE II	J. M. Zawodny	O <sub>3</sub> ND versus altitude; 11–56 km at 1 km
Dobson	M. Clark and S. Oltmans	total column O <sub>3</sub>

Measurements are ozone number density (ND) versus altitude or ozone mixing ratio (MR) versus pressure. Altitude range and reporting interval are given. MLO3, Stratospheric Ozone Profile Intercomparison at Mauna Loa; JPL, Jet Propulsion Laboratory; FTIR, Fourier transform infrared; MLS, Microwave Limb Sounder; SAGE II, Stratospheric Aerosol and Gas Experiment II.

composite ozone profile is created by combining the high-altitude and low-altitude profiles.

The JPL lidar data were collected on 17 nights between sunset and midnight with integration times of 1–2 hours. A typical measurement is integrated for  $10^6$  shots. Because of the possibility of interference between the two very similar lidar systems (which were located within 10 m of each other), the GSFC and JPL lidar systems were operated in sequence each night, alternating early and late shifts. The intrinsic measurement is of ozone number density as a function of altitude. Data were provided for each 0.3 km, usually from 14 to 52 km (see Figure 3). The error estimate associated with each profile is obtained from counting statistics of the  $10^6$  shots on a given evening.

An ozone profile of mixing ratio versus pressure was also computed from the lidar data by using National Center for Environmental Prediction (NCEP) data and model climatological data. The conversions provided by the experimenter used pressure and temperature data versus geopotential height instead of geometric height. The difference is quite small, but in order to obtain consistent conversions for this comparison, we have converted the height versus number density profiles in the original data files by using the NCEP temperature and pressure data versus geometric height. These profiles are used in this paper when JPL lidar profile data are given versus pressure.

## 2.2. Goddard Differential Absorption Lidar

The GSFC lidar [McGee *et al.*, 1991, 1995] is very similar to the JPL lidar. It also uses a XeCl excimer laser to produce a main beam at 307.9 nm, but the reference beam at 355 nm is produced using the third harmonic of a Nd:yttrium/aluminum/garnet (YAG) laser. Both lasers operate at 66 Hz. Backscattered light is collected by using a 76 cm telescope, separated by dichroic optics, and measured by photomultiplier tubes in photon-counting mode. Data are recorded for six channels in 1 ms bins. The backscattered beams at 307.9 and 355 nm are each split into high-intensity/low-intensity channels, with a 96%/4% split for the 308 channel and a 90%/10% split for the 355 channel. The two weaker beams are used to derive the lower-altitude profile, and the two stronger beams are used to derive the upper profile. The two remaining channels measure the  $N_2$  Raman shifted backscatter at 332 and 382 nm (shifted from 307.9 and 355 nm, respectively). These last two channels' measurements can be used to correct for the effects of Mie scattering by aerosols [McGee *et al.*, 1993]. Details of the ozone retrieval are presented by McGee *et al.* [1991]. A typical measurement is integrated for  $10^6$  shots and takes less than 2 hours.

The GSFC lidar data were collected on 16 of the 18 possible nights. The native form of the measurement is number density versus height. Data were provided for each 0.15 km, usually from 15 to 50 km. The actual range resolution varies with altitude, from 1.2 km near 20 km to 6.75 km near 45 km. The GSFC lidar has a less powerful laser and a smaller telescope than the JPL system, and consequently the data become “noisier” near the upper altitude limit. The conversion to mixing ratio versus pressure was obtained by using NCEP temperature and pressure data. The standard deviation estimates in the data files are obtained from counting statistics.

## 2.3. ECC Ozonesondes

Balloon sondes were launched each evening from the Hilo Airport, which is  $\sim 60$  km east of the Mauna Loa Observatory. The prevailing winds are from the NE, so the flight paths of the

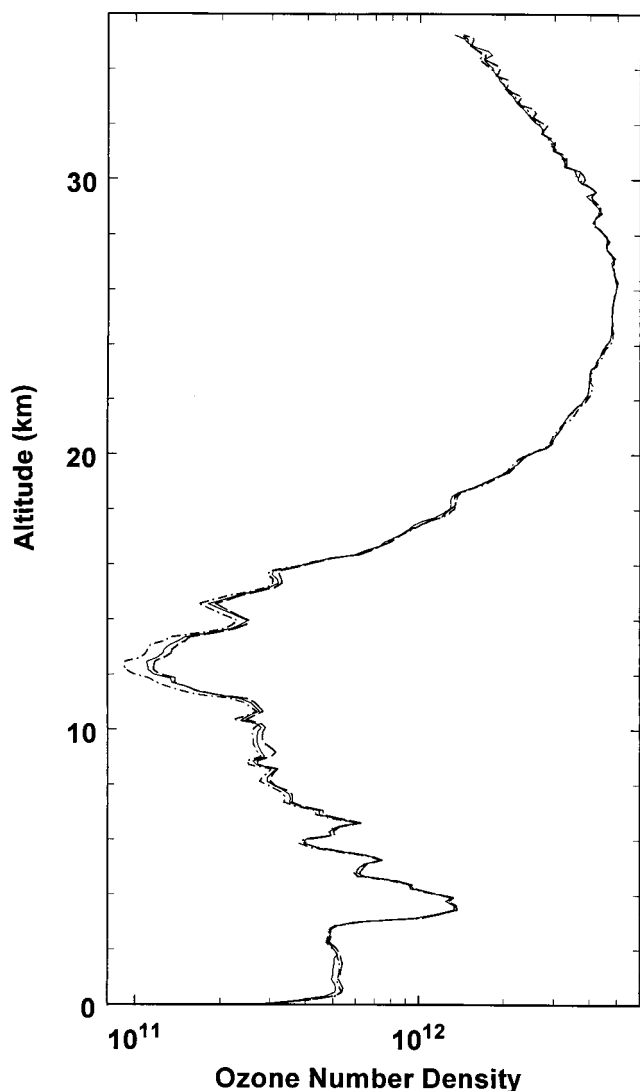
balloons tended to be toward MLO. The balloon sondes were launched just after sunset in order to be nearly coincident with the laser measurements. The flight times were  $\sim 2$  hours with ascent rates of  $5 \text{ m s}^{-1}$ .

The balloon sondes were standard electrochemical concentration cell (ECC) ozonesondes manufactured by EN-SCI Corporation. The ozonesondes were coupled to Vaisala meteorological radiosondes that measure temperature, pressure, and humidity as the balloon ascends. The ECC devices are described in detail by Komhyr *et al.* [1995]. During operation, sampled air is pumped through a 1% buffered, potassium iodide solution. Ozone reacts to form iodine ( $I_2$ ), which changes the electromotive force across the cell, resulting in a flow of current through the external circuit board. The zero-ozone background current averaged  $0.043 \pm 0.019$  for all of the MLO3 flights. The background current was treated as a constant offset throughout the balloon flight. The current due to ozone, the pump efficiency, the pump temperature, and the external temperature are combined to derive estimates of ozone number densities. The pump efficiency correction factor, which is critical to accuracy above 25 km, was determined empirically for each ECC sonde. Ozone mixing ratio as a function of pressure can be derived directly from measured quantities. The integrated column ozone is compared to Dobson as a quality check, but no normalization is done. The ozonesonde data were averaged and reported for each 0.15 km. The maximum altitude for the ozone profile was usually around 35 km, except for the August 20 flight, which only reached 24 km. The altitudes provided with the balloon data were geopotential heights. The measured temperature and pressure data were used to determine geometric height versus pressure before intercomparison with lidar and other data.

Five of the 16 flights were triple ECC flights, on August 15, 19, 22, and 30 and on September 1. On these flights, three complete ECC packages were flown on a single balloon in order to check the consistency of the sensors. It was found that the consistency averaged better than 2% as is shown in Figure 2. Occasionally, one channel would deviate from the other two by several percent for a few minutes (percent errors can exceed 20% near the ozone minimum at 12 km) but then would return to agreement. A single set of ozone values was used from the average for each triple flight.

## 2.4. Microwave Radiometer

The Millitech/Langley Research Center (LaRC) microwave instrument consists of an automated microwave receiver and a 120-channel spectrometer tuned to the ozone transition at 110.836 GHz [Parrish *et al.*, 1992]. The raw data consist of ratios of the power incident from two viewing directions, one from near zenith and one from an elevation of  $10^\circ$ – $25^\circ$ . The ozone profile is retrieved from details of the pressure-broadened line shapes. The retrieval algorithm is discussed by Parrish *et al.* [1992], and an error analysis is presented by Connor *et al.* [1995] and Tsou *et al.* [1995]. The results of the microwave instrument measurements were provided in two data files per day for the 18 days of the intercomparison, one as an average of the daytime measurements and one as an average of the nighttime measurements. Comparisons made here use the nighttime measurements as a slightly better match in time of observation for the lidar and balloon profiles. Profiles of ozone mixing ratio versus pressure are derived from 56 to 0.1 hPa at 23 pressure levels. The measurements were integrated for 9 hours for the nighttime measurements except on



**Figure 2.** The result of flying three electrochemical concentration cell (ECC) packages on a single balloon on September 1.

August 19, 23, and 24, when only 3 hours of measurements were available. NCEP data are used to convert the mixing ratio versus pressure profiles to number density versus height.

### 2.5. Dobson and Umkehr Measurements

Measurements from two Dobson instruments, the Mauna Loa station instrument 76 and the World Standard Dobson instrument 83, were used to compute total column ozone and Umkehr profiles during MLO3. A Dobson spectrometer normally derives total column ozone from the AD wavelengths, wavelength pairs A (305.0/325.0 nm) and D (317.5/339.9 nm). For the traditional Umkehr retrieval of an ozone profile, the C pair (311.5/332.4 nm) is used, and zenith sky measurements are made for a series of solar zenith angles (60°, 65°, 70°, 74°, 77°, 80°, 83°, 85°, 86.5°, 88°, 89°, and 90°). The measurements may be made during either sunrise or sunset. The scattering contribution function peaks at an altitude that depends on the product of the ozone cross section times the optical path. For an Umkehr retrieval, varying optical path (solar zenith angle) provides the altitude scan. The measurements are corrected for minor (<1.5%) aerosol interference [Newchurch and Cun-

nold, 1994] using SAGE II aerosol measurements and inverted in a maximum likelihood retrieval algorithm [Mateer and DeLuisi, 1992] to estimate ozone mixing ratio versus pressure. The Umkehr retrieval produces layer ozone amounts as a function of pressure for layers that increase by exactly a factor of 2 in pressure. The Umkehr retrieval is considered to provide good information in Umkehr layers 4 through 8 (from 64 hPa up to 2 hPa). A spline interpolation is used to obtain mixing ratio profiles on a finer pressure scale for comparison with other profile data.

Instrument 76 operated in a semiautomatic mode, while instrument 83 required an operator. Umkehr measurements were obtained by instrument 76 for 16 days (15 morning and 10 afternoon measurements). Instrument 83 made measurements of total column ozone on 13 days (13 mornings and 4 afternoons).

### 2.6. UARS MLS

The Microwave Limb Sounder (MLS) instrument on the Upper Atmosphere Research Satellite (UARS) measures thermal emissions in 6 mm wavelength bands with double-sideband heterodyne radiometers centered near 63, 183, and 205 GHz by scanning through the atmospheric limb [Waters, 1989; Froidevaux *et al.*, 1996]. The measurements in the 183 and 205 GHz spectral bands may be used to retrieve ozone profiles. The ozone data used in this study are retrieved from the 15 channels spaced contiguously about the ozone line centered at 206.13205 GHz. Details of the retrieval algorithm are given by Froidevaux *et al.* [1996].

The UARS MLS made measurements on 11 days during MLO3, and a data file for the MLS profile closest to Mauna Loa each day was provided. These data are from a preliminary version 4 data set (software version 4.15). Comments about the more definitive MLS data set (version 5) are provided in section 4. The matched profiles were always coincident within 2° of latitude and 5° of longitude. The MLS profiles are in the form of ozone mixing ratio at pressures from 100 to 0.2 hPa at 17 levels. The error bars provided make it clear that the lowest two layers should not be used, and the profiles should be cut off at 46 hPa. This is consistent with the work of Froidevaux *et al.* [1996], who note that the ozone values for the 205 GHz retrievals are most reliable in the 22–0.5 hPa region.

### 2.7. SAGE II

The Stratospheric Aerosol and Gas Experiment II (SAGE II) on the ERBS satellite is designed to measure atmospheric aerosols and ozone using the occultation technique [Mauldin *et al.*, 1985; Cunnold *et al.*, 1989]. Measurements are made at 1020, 940, 600, 525, 453, 448, and 385 nm during spacecraft sunrise or sunset events, about 15 of each per day. The locations of the measurements are well distributed in longitude but vary slowly in latitude, sweeping between the high-latitude extremes in about a month. The 600 nm channel in the center of the Chappuis absorption band is used to retrieve ozone profiles from near the surface (if there are no clouds) to near 60 km. Details of the SAGE II ozone inversion algorithm are presented by Chu *et al.* [1989]. Results used here are from the version 5.96 algorithm, which has an improved aerosol correction.

There were no SAGE II measurements near the latitude of MLO until near the end of the campaign. A close matchup occurred on August 30 when a measurement was made for which the tangent point was ~270 km west of MLO. Ozone was retrieved between 10 and 56 km at 1 km resolution, with



**Table 2.** Observations Made Each Day During MLO3

	Days in August–September 1995																		Total
	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	1	
Balloon	x	x	x	x	x	x	x	x		x	x	x		x	x	x	x	x	16
Goddard lidar	x	x	x	x	x	x	x	x	x	x		x		x	x	x	x	x	16
JPL lidar	x	x	x	x	x	x	x	x	x	x	x			x	x	x	x	x	17
Microwave	x	x	x	x	x	x	x	x	x	x	x		x	x	x	x	x	x	18
MLS		x	x		x	x		x	x		x	x		x	x	x			11
SAGE II																	x		1
Umkehr	x	x	x	x	x	x	x	x	x		x	x	x	x	x		x	x	16
FTIR																	x	x	2
I83 total	x	x	x	x	x	x		x	x				x	x	x	x	x		13

I83 is the World Standard Dobson instrument 83.

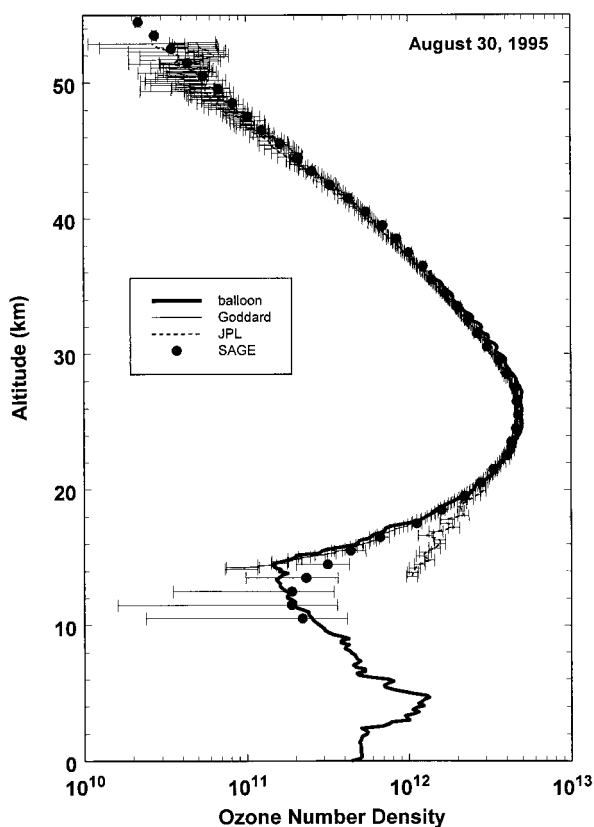
some aerosol contamination being indicated between 18 and 21 km. Two sigma error bars are also provided. The SAGE team normally prefers to provide only their primary data product, number density versus altitude. For comparison with instruments that measure ozone mixing ratio versus pressure, we converted the SAGE profile using NCEP data, consistent with the conversion done for the lidar instruments.

## 2.8. FTIR

The Fourier transform infrared (FTIR) instrument used for the retrievals included here was being installed at MLO during the ozone campaign. For that reason, data are available for the last few days only. The instrument is a 284 cm path difference

interferometer (nominal  $0.003\text{ cm}^{-1}$  spectral resolution), manufactured by Bruker Instruments, Germany. It was operated with a Mercury-Cadmium-Telluride detector and a band-pass filter covering  $750\text{--}1300\text{ cm}^{-1}$ . Solar radiation is maintained on the interferometer entrance by a two-axis, servo-controlled tracking system. For these studies, two interferograms were coadded, with a total collection time of  $\sim 5$  min.

Information about the altitude distribution of a particular species is contained in the line shape due to pressure broadening. In the midinfrared, typical broadening coefficients are  $\sim 0.1\text{ cm}^{-1}$  per atmosphere, and the transition between pressure broadening and Doppler broadening occurs around 30 km altitude. An iterative technique for determining the profile was developed, and it is described in detail by *Liu et al.* [1996]. For ozone an isolated absorption line near  $1163\text{ cm}^{-1}$  was used. It has an appropriate strength and low temperature dependence. The retrieval technique starts from an initial guess profile and iteratively adjusts the shape of the profile to improve the detailed spectral fit. In altitude regimes where the spectra provide no information, the profile stays at the initial guess. For the ozone line used here no information came from the spectrum above  $\sim 32$  km. A complete error analysis for ozone has been done by *Nakajima et al.* [1997] for a series of observations over Japan.

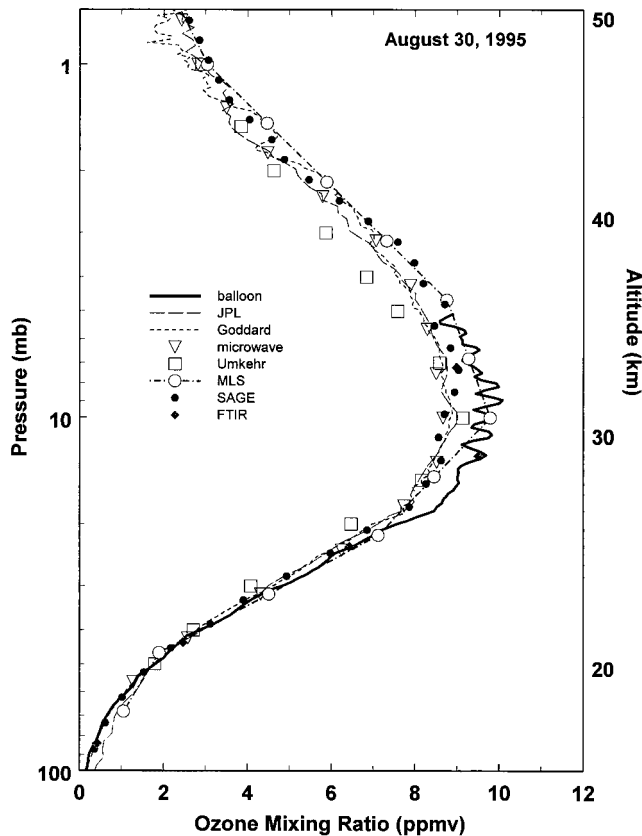


**Figure 3.** Plot of observations of ozone number density versus altitude on August 30, 1995. Error bars are shown for the lidars and for Stratospheric Aerosol and Gas Experiment (SAGE).

## 3. Comparison Methodology

Observations were made for the intercomparison from August 15 through September 1, 1995, so there are 18 possible days on which comparisons can be made. The schedule of observations actually made is shown in Table 2. There was only one SAGE overpass during the mission, on August 30, when a measurement was made close to Mauna Loa. Since there were also measurements from all the other instruments that day, it is instructive to examine the comparisons for that single day before looking at average comparisons. Figure 3 is a plot of ozone number density versus altitude for the two lidar instruments, SAGE, and the balloon sonde. The ECC sonde that day was one of the triples in which three independent ECC packages were flown on the same balloon, adding to the credibility of that balloon measurement. The error estimate plotted with each lidar profile is obtained from counting statistics of the  $10^6$  shots on that evening. For the lidars and for SAGE, altitude is the natural variable. Because balloon sensors measure both temperature and pressure, altitude can be determined directly.

The comparisons in Figure 3 show that the lidar measurements have an error based on counting statistics that varies



**Figure 4.** Comparison of ozone mixing ratio as a function of pressure for measurements made on August 30, 1995. Lidar and SAGE have been converted to pressure scale for this comparison. MLS, Microwave Limb Sounder; FTIR, Fourier transform infrared.

from  $\sim 4\%$  near 15 km, to 0.3–0.8% near 30 km where the more sensitive range begins to be used, to  $\sim 10\%$  at 45 km. Near 50 km the errors become much larger, near 50% for the Goddard lidar. There are algorithmic differences between the two lidars, particularly in the upper stratosphere, that have to do with the amount of vertical averaging that must be done to obtain a good profile. Neither lidar is able to measure ozone below  $\sim 15$  km, because the returned signal from lower altitudes is too large in the clean atmosphere over MLO and exceeds the dynamic range that the systems can accept.

The Goddard profile matches the balloon profile down to the tropopause, but the JPL profile deviates significantly at altitudes below 20 km. The deviation of the JPL lidar results at low altitudes is now understood. The JPL lidar was designed to measure ozone in the upper stratosphere and has a high power-aperture product in order to accomplish this. This can lead to signal saturation from intense returns in the lower atmosphere. This problem was anticipated and expected to show as pulse pileup in the photon-counting detection system. This intercomparison revealed an unexpected saturation in the hardware of the detection system that did not result in pulse pileup and went undetected. This problem has now been remedied, but the raw data obtained under these saturated conditions cannot be corrected using the normal procedures for pulse pileup.

Between 20 and 42 km the agreement between the two lidars is excellent, to within  $\pm 3.3\%$ . Between 45 and 50 km the

Goddard number density is, on average, 6% lower than that for JPL, varying  $\pm 16\%$ . This difference is due to the fact that the Goddard system is less powerful than the JPL system and does not have the signal strength to maintain accuracy above  $\sim 45$  km.

The SAGE profile agrees well with the balloon profile, within 4% between 18 and 27 km. Below 18 km this version of the SAGE II algorithm has known problems arising from an incomplete oblate Earth model and a deficiency in the atmospheric refraction calculation. SAGE agrees with the lidar profiles within 3% between 20 and 42 km. SAGE is 6% lower than the JPL lidar result in the 45–50 km region.

Figure 4 is a comparison for the same day, August 30, but of mixing ratio as a function of pressure. Mixing ratio comparisons are better for revealing the behavior of ozone in the middle stratosphere, while number density comparisons are better for examining the lower stratosphere and troposphere. The balloon ECC sonde, Umkehr, MLS, microwave, and FTIR measurements are all intrinsically a function of pressure. The lidar and SAGE measurements were converted to mixing ratio versus pressure using NCEP data for that day. (The conversion introduces some uncertainty into the comparison as will be discussed in section 6.) Near the mixing ratio maximum, the 6–15 hPa region, the Goddard and JPL lidars, the microwave, and SAGE all agree, on average, to within 2%. MLS is  $\sim 6\%$  higher than the lidars, while the Umkehr mixing ratios are  $\sim 4\%$  lower. In the upper stratosphere, the 2–6 hPa region, the lidars, SAGE, and the microwave continue to agree to within 2%, MLS is  $\sim 8\%$  high, and Umkehr drops to 13% lower. The FTIR profile begins to disagree with the other measurements at altitudes above  $\sim 27$  km and, for the ozone line used here, has no information above 32 km. Below 27 km the FTIR ozone is 2–5% higher than that from balloon or lidar.

The balloon mixing ratio at altitudes above 27 km is clearly higher than all the other measurements except MLS, by  $\sim 9\%$ . (The structure seen in the balloon profile near 30 km on this day is unusual only in its apparent regularity.) The higher ozone measured by the balloon sonde can be explained primarily from the pump correction factors (PCFs) used in the ozone algorithm and the cathode solution recipe used in the sensor cell. The MLO3 ozonesondes used a 1% KI buffered solution recipe, as recommended by the ECC manufacturers (1994 EN-SCI manual and Science Pump manual). However, the PCFs were determined empirically for the MLO3 ozonesondes, using a new method developed at National Oceanic and Atmospheric Administration (NOAA)/Climate Monitoring and Diagnostics Laboratory (CMDL) [Johnson *et al.*, 1998]. These PCFs ranged from an average of 1.024 ( $\pm 0.011$ ) at 100 hPa to 1.244 ( $\pm 0.041$ ) at 5 hPa, 2 and 11% higher than the respective EN-SCI manual values which were measured by Komhyr *et al.* [1995]. The EN-SCI manual states that their recommended PCF values may be too low, but their use in data processing will compensate roughly for increased sensitivity (higher ozone) of the ECC sensor as the cathode electrolyte concentration increases owing to evaporation. The Jülich Ozonesonde Sonde Intercomparison Experiment (JOSIE) also showed that using the larger PCFs with the 1% buffered solution gives too high ozone amounts above  $\sim 50$  hPa if account is not taken of the increasing sensitivity of the solution [WMO, 1998]. NOAA/CMDL performed a laboratory test to estimate the increased sensitivity, due to evaporation, by running ozonesondes at a constant ozone mixing ratio. The response slowly increased by  $\sim 6\%$  after 2 hours. Tarasick *et al.* [1998] did

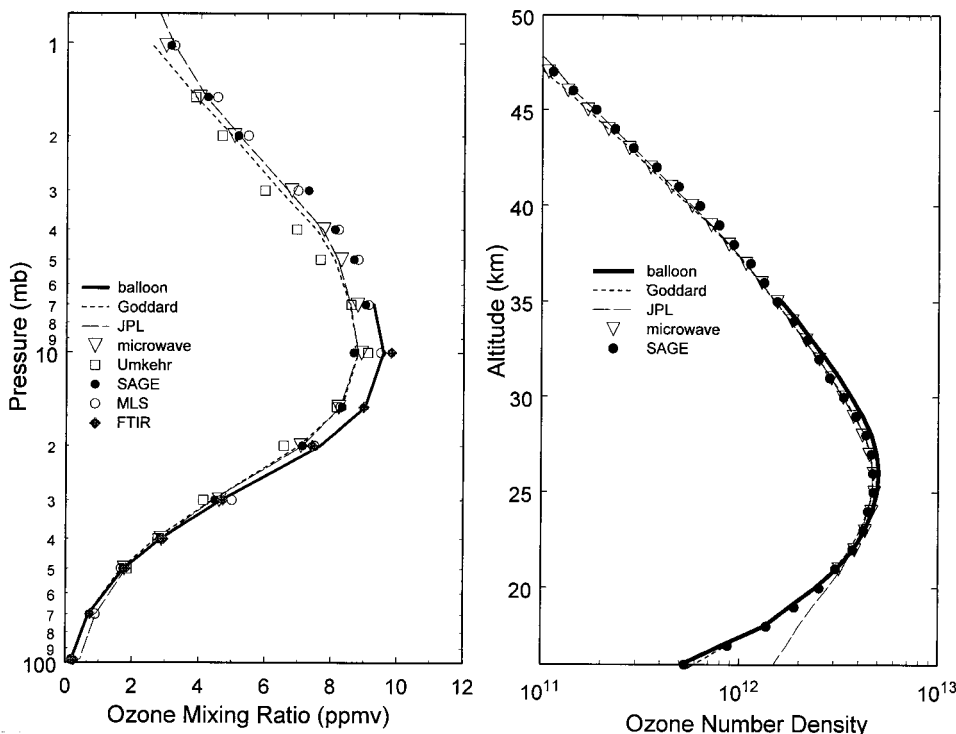


Figure 5. Ozone profiles averaged over the entire 18 day comparison.

similar laboratory tests with an ECC ozonesonde and reported up to 7% higher measured ozone after 90 min.

Evaporation is greater at decreasing pressures in an actual balloon flight, so the 6% correction would be considered a conservative estimate. More recent laboratory and field (dual ozonesondes) tests at NOAA/CMDL have shown that the buffer is primarily responsible for the increased sensitivity to ozone, by as much as 10–15% higher ozone at burst altitude (~5 hPa) (B. Johnson et al., manuscript in preparation, 1999). In summary, the increased ozone sensitivity of the 1% KI cathode solution, which occurs with solution evaporation, is roughly compensated by the EN-SCI recommended pump efficiency correction factor, even though these are independent factors. The 0–6% correction, beginning at 50 hPa, was apparently too conservative when using the PCF values measured by NOAA/CMDL for the MLO3 experiment.

#### 4. Results of Comparing Averaged Profiles

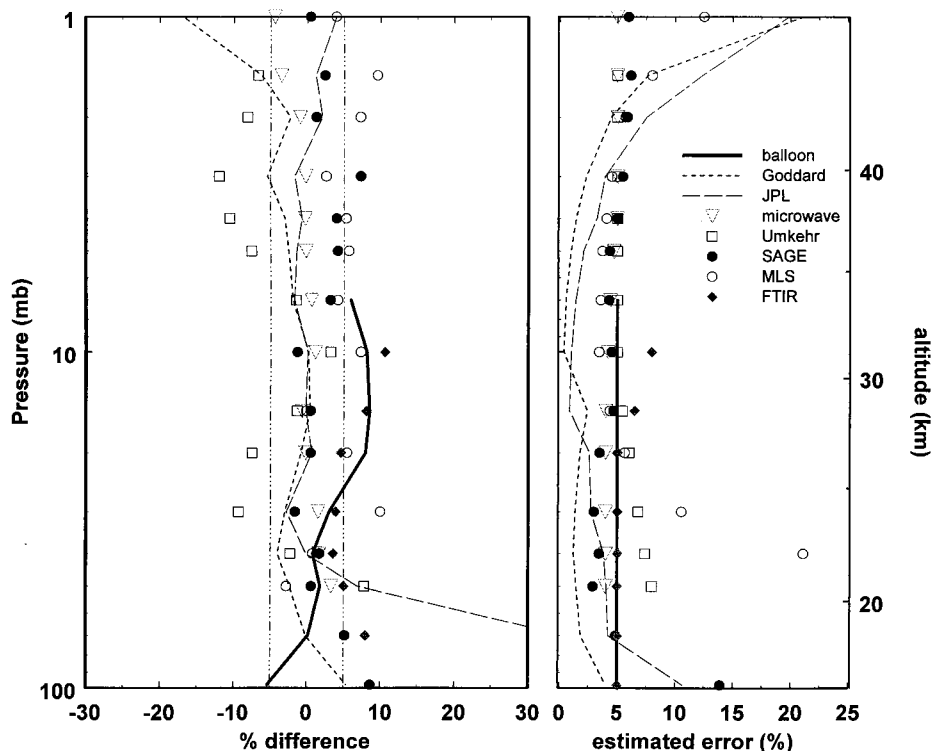
It is of course more reliable to examine the average behavior of each instrument over the 18 day period of measurements than to base conclusions on only 1 day. The average ozone profiles (mixing ratios on the left and number densities on the right) for each instrument are shown in Figure 5. (Note that the SAGE data shown are based on only 1 day and the FTIR data shown are based on 2 days, which will increase the uncertainty of these comparisons.) The averages confirm that the profile differences seen in the plots for August 30 were typical and not unique to that 1 day.

In order to quantitatively compare profiles, it is more useful to examine percent difference plots. If the true ozone profile is known, the difference plot is a powerful tool for identifying any weakness in a measurement. However, for a field measurement campaign like this, true ozone is not known. A strategy fol-

lowed in previous intercomparisons has been to compare each instrument's profile to the average of all the measurements. The drawback is that if there are systematic errors in one or a few of the instruments, structure will be introduced into the comparisons of other instruments that can be confusing.

In the absence of a "truth" profile, the different measurement techniques can best be evaluated on the basis of consistency. When profiles are inconsistent, a judgment must be made on the basis of knowledge of instrument limitations. For example, the balloon profile is known to be in error above 27 km (20 hPa), because of incompatible pump correction factors and sensing solution chemistry. The JPL lidar has an identified saturation problem below 20 km, while the Goddard lidar loses sensitivity above 43 km. The MLS positive offset has been identified as algorithmic. A "consensus" reference profile was created on the basis of instruments that have no known errors over various altitude ranges. Balloon data are used from the surface to 25 km. Goddard lidar data are used between 16 and 43 km. JPL lidar data and microwave data are used between 22 and 50 km. SAGE data are used between 20 and 50 km. The measurements in the consensus profile agree to within an average of  $\pm 3\%$  and no worse than  $\pm 5\%$ . We emphasize that the purpose of the consensus profile is to serve as a stable reference. However, we do feel that if three or more instruments using different physical measurement techniques are consistently in very good agreement, this is strong evidence that the results are accurate and technique independent.

Figure 6 is a plot of the deviation of each instrument average profile from the reference profile. In order to compute differences, it was necessary to spline the average profiles to consistent pressure levels, but no smoothing was done. An immediate conclusion is that almost all the instruments agree to within  $\pm 10\%$  (which was the best that could be expected of profile



**Figure 6.** (left) Percent deviation of the comparison average for each instrument from a "consensus" reference profile. (right) The estimated percent error for each measurement.

measurements just a few years ago), and most of the measurements agree within  $\pm 5\%$ . The average balloon data are higher than the reference near 30 km by  $\sim 8\%$  for reasons explained in section 3. The Goddard lidar profile is lower than the reference at 45 km and above, a region in which the signal is marginal and must be heavily averaged. The JPL lidar develops a serious positive bias at 20 km and below as noted in section 3. The MLS profile tends to be consistently high relative to the reference, generally by  $\sim 5\%$ . This will likely be remedied in version 5 MLS data, which, at this latitude, are typically 2–6% lower in the 2–22 hPa range than the version used here. The Umkehr profile is lower than the reference near 25 km by  $\sim 8\%$ , is close to agreement near 32 km, and then is lower by  $\sim 10\%$  near 40 km similar to other Umkehr-SAGE comparisons [Newchurch *et al.*, 1998]. For this comparison campaign the error from the aerosol correction term is nearly negligible, being less than 1.5% in all layers.

## 5. Comparison of Total Column Ozone

The measurement of total column ozone is currently far more accurate than that of the ozone altitude dependence. A well-calibrated Dobson or Brewer can arguably measure total ozone to an accuracy of  $\pm 1\%$  [Komhyr *et al.*, 1989; WMO, 1995], so  $\pm 1.5\%$  would be a conservative estimate. Measurements of total column ozone made by the World Standard Dobson instrument 83 on 11 days during the comparison have been used to evaluate the overall accuracy of the profiling instruments. The results are given in Table 3. Since Dobson measurements are made during the day and the lidars measure at night, there is an assumption that diurnal variation is small and that large changes in ozone are not occurring.

Since no instrument measures the altitude distribution from

the surface to the top of the atmosphere, adjustments must be made. The average total ozone measured by Dobson 83 during the comparison was 260.3 Dobson units (DU). The integrated column measured by the Goddard lidar on the same 11 days was 241.4 DU, but this column was generally down to a minimum altitude of  $\sim 15$  km. The amount of ozone between the MLO station altitude (3.4 km) and 15 km was taken from each day's balloon profile and added to the lidar column. This amounted to an average of 26.7 DU. This gives an adjusted

**Table 3.** Comparisons of Average Total Column Ozone for 11 Days on Which Dobson Measurements Were Made

Measurement	Column Ozone Amount, DU
MLO Dobson measurement	260.3 ( $\pm 1.5\%$ )
Goddard lidar comparison	
Goddard lidar column	241.4
MLO to bottom of lidar ( $\sim 15$ km) from balloon	26.7
Adjusted Goddard total	268.1 (+3.0%)
JPL lidar comparison	
JPL lidar column	265.4
MLO to bottom of lidar ( $\sim 15$ km) from balloon	25.4
Adjusted JPL column	290.9 (+11.8%)
Balloon comparison	
Balloon column*	254.1
Column from sea level to MLO	-7.1
Column above balloon ( $\sim 35$ km) from lidar	27.5
Adjusted balloon column	274.5 (+5.4%)
Lidar high-altitude comparison	
Goddard column above 27 km	113.3
JPL column above 27 km	112.9

Percent differences are shown in parentheses.

\*Value is based on 10 good balloon profiles.



column for the Goddard lidar measurement of 268.1 DU, 3% higher than the Dobson average. Since the ozone from the balloon measurement added to the lidar column amounts to only 10% of the total, an error of as much as 10% in this adjustment term would introduce only a 1% error into the column. As long as the correction terms are small, they will introduce little error into the total column ozone comparison. A similar comparison for the JPL measurement results in a positive 11.8% bias relative to Dobson. This is strong confirmation that the bias below 20 km relative to the balloon measurement is indeed an error in the lidar retrieval. When the two lidar measurements are compared by integrating the column above 27 km, they agree to within 0.3%, demonstrating the high degree of consistency of the two lidar measurements in the middle and upper stratosphere.

The balloon measurement can be similarly compared with Dobson. Here the amount of ozone between sea level (the balloons are launched from Hilo) and MLO (at 3.4 km altitude) must be subtracted, an average of 7.1 DU. The balloons usually reached  $\sim 35$  km before the ECC sondes failed. (Data for August 20, when the balloon only reached 24 km, were not included in the average.) The column above the balloon maximum altitude was taken from the JPL lidar measurement and amounts to an average of 27.5 DU, again only  $\sim 10\%$  of the total. The adjusted balloon total column amounts to 274.5 DU, 5.4% higher than the Dobson total. This is additional evidence that the ozone measured by the ECC sonde near 30 km was indeed too high.

## 6. Minor Error Sources

The complete intercomparison of ozone profiles obtained during MLO3 requires that all the data sets be converted to a consistent vertical scale, whether in pressure or in height. No matter which sets are converted, additional uncertainties are introduced. Some of the participants can provide estimates of these conversions on their own, while others use information from other sources (often the NCEP analysis). The information needed to calculate pressure versus height can be obtained from temperature versus pressure measurements (e.g., as obtained from balloon sondes), density versus height (e.g., as obtained from Lidar systems), or temperature versus height.

While the physical laws governing the relationships among height, pressure, density, and temperature are well established, there are complications and opportunities for errors in applying them. Two opportunities for computational errors were encountered in working with the data sets in the intercomparison. The first, the simplest to make and to correct, involves the geopotential heights normally provided in the NCEP and balloon sonde data. The conversions to and from geometric height are

$$H = [g(L)/g_0][(ZR)/(R + Z)],$$

or

$$Z = (HR)/[Rg(L)/g_0 - H],$$

where  $R$  is the radius of the Earth,  $Z$  is the geometric height,  $H$  is the geopotential height,  $L$  is latitude,  $g(L)$  is the local value of gravity, and  $g_0$  is a standard value of gravity. The errors in using geopotential height as geometric height grow quadratically with height. If  $g(L)$  equals  $g_0$ , then the geopotential heights are less than the geometric heights by approximately 1/16 km at 20 km, 1/7 km at 30 km, 1/4 km at 40 km,

and 2/5 km at 50 km. These errors lead to ozone number density errors of approximately 1, 2, 5, and 8%, respectively, with a change in sign between 20 and 30 km. The sign of the error depends on how the height versus pressure conversion is applied. As pointed out by a reviewer, the corrections applied in the intercomparisons presented here did not account for the difference between  $g(L)$  and  $g_0$ . The recommended value for  $g_0$  is 9.80665 m s<sup>-2</sup>; however, the value used in practice for  $g_0$  is 9.8 m s<sup>-2</sup>. The estimated value for  $g(L)$  for Mauna Loa is 9.78638 m s<sup>-2</sup>. This leads to a linear error in height that grows from a 0 km error at 0 km to  $\sim 1/14$  km at 50 km and ozone number density errors of  $\sim 1\%$  at 50 km.

A more subtle effect of the decrease in gravity with height involves the associated change in the gradient of neutral atmosphere column amount with pressure. Because of the radius-squared dependence of gravity, the number of molecules in a column with constant cross section in the layer between, for example, 100 and 99 hPa, is less than the number of molecules in the layer between 2 and 1 hPa. This information is used in the computation of height versus pressure from temperature or density information. One must also check to make sure that participants reporting their results as ozone versus pressure have not incorrectly made an implicit change of variables from number of molecules in the path and the relative path length to pressure. This problem also complicates the computation of ozone mixing ratios. From computations with a standard atmosphere, one can find that the incorrect pressure estimate computed from neutral atmosphere density without including the decrease in gravity is related to the true pressure by

$$P_e \approx P[1 + (2Z + 14)/R],$$

where  $P$  is the actual pressure,  $P_e$  is the incorrect pressure,  $Z$  is the geometric height in kilometers, and  $R$  is the Earth's radius in kilometers. The ozone error is a product of the pressure error, which is approximately linear in log pressure, times the ozone gradient, which varies with pressure. Typical ozone errors from using the incorrect pressure estimates are  $-1.5\%$  at 30 hPa, no error at 10 hPa,  $+1.2\%$  at 3 hPa, and  $+0.8\%$  at 1 hPa.

## 7. Conclusions

This intercomparison shows that progress is being made toward bringing the profile measurement techniques into agreement. Almost all the instruments agreed to within  $\pm 10\%$ , which was the best that could be expected of profile measurements just a few years ago, and most agreed within  $\pm 5\%$ . We feel that it is a significant indicator of progress when instruments using different physical measurement techniques are consistently in such good agreement.

Both lidars, the microwave instrument, and SAGE II agree within 5% between 22 and 43 km, providing strong evidence that the lidars and the microwave instrument are making accurate measurements in this range. The JPL lidar, microwave instrument, and SAGE II continue to agree within 5% up to 50 km, providing evidence that the measurements of the JPL lidar and the microwave instrument continue to be accurate to that altitude. The Goddard lidar, sonde, and SAGE II agree within 5% down to 18 km, providing evidence that these three are making accurate measurements down to this level. The SAGE II disagreement with the balloon profile below 18 km is due to known algorithmic problems arising from an incomplete oblate Earth model and a deficiency in the atmospheric refraction

calculation. (Conclusions about the accuracy of SAGE II at the 5% level cannot be drawn from the single measurement.) The balloon data were used from the surface to 25 km and agreed well with the Goddard lidar and with SAGE II in the 18–25 km region. The positive bias of ~8% near 30 km seen in this comparison resulted from using the larger measured pump correction factors along with the 1% KI buffered cathode solution. This is now better understood as a result of this intercomparison and does not indicate an intrinsic problem with the balloon measurement.

The MLS data used in this comparison, version 4.15, tended to be high near the mixing ratio peak, by ~5%. The latest (version 5) MLS data are expected to yield lower ozone mixing ratios, by 2–6%, for the middle to upper stratosphere, in better agreement with other instruments in this comparison. The Umkehr profile was low near 25 km by ~8%, was close to agreement near 32 km, but then was low by ~10% near 40 km. Although the participation of the FTIR instrument was limited to only 2 days, information was provided up to 32 km. The results were 2–5% high up to 24 km, increasing to ~10% high near 32 km.

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