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The UARS Microwave Limb Sounder version 5 data set: Theory, characterization, and validation

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[1] This paper describes the latest and (for most products) definitive data set from the Microwave Limb Sounder (MLS) on the Upper Atmosphere Research Satellite (UARS). MLS data have formed the basis of numerous studies, and the version 5 data, produced using more advanced algorithms than earlier versions, represent a significant improvement in quality and scientific applicability for most of the MLS data products. The version 5 data include midstratospheric to lower mesospheric temperature and geopotential height (the latter is a new product from MLS), water vapor from the upper troposphere to the mesosphere, stratospheric and mesospheric ozone, and stratospheric nitric acid, chlorine monoxide, and methyl cyanide (also a new product). The vertical retrieval grid over the stratosphere and lower mesosphere has been doubled, to six surfaces per decade change in pressure ($\sim$2.5 km), compared to three surfaces per decade in previous versions. The accuracy and precision of lower stratospheric ozone, chlorine monoxide, and nitric acid have been improved. For each product, a description of relevant changes to the algorithms is given, along with an update on its validation, a description of the accuracy, precision, and vertical resolution of the data, and information on what quality control methods to apply when using the data. *INDEX TERMS:* 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 0394 Atmospheric Composition and Structure: Instruments and techniques; *KEYWORDS:* Microwave Limb Sounder (MLS), Upper Atmosphere Research Satellite (UARS), upper tropospheric humidity, stratospheric ozone, stratospheric composition, stratospheric chemistry

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1. Introduction

[2] The Microwave Limb Sounder (MLS) is one of ten instruments on the Upper Atmosphere Research Satellite (UARS) [*Reber et al.*, 1993], which was launched from the space shuttle Discovery on 12 September 1991. The UARS instruments measure important aspects of the chemistry, dynamics and energy budget of the Earth's atmosphere. MLS uses a microwave heterodyne technique to observe thermal emission from the Earth's limb; it was designed to measure stratospheric ozone, water vapor and chlorine monoxide. In addition to these data, MLS has also produced useful observations of stratospheric and mesospheric temperature, stratospheric nitric acid, stratospheric sulfur dioxide during periods of significant enhancement (such as following the eruption of Mount Pinatubo), upper tropospheric humidity, and stratospheric methyl cyanide (also called acetonitrile).

[3] The microwave observations made by MLS are converted into geophysical quantities by ground-based data processing software. This paper describes "Version 5" of this software and the data it produces (known collectively as v5 hereafter). The main change from earlier versions of the MLS data set is that the products are reported on a pressure grid with half the vertical spacing of that used in previous versions (now being 6 surfaces per decade change in pressure, corresponding to about 2.5 km) over the stratosphere and in the lower mesosphere (up to 0.1 hPa), though the true resolution of the information in each profile is typically coarser. In addition, the quality of the observations in the lower stratosphere has generally been improved, because of better limb tangent pressure algorithms and the use of nonlinear iterative retrieval methods for some species. The v5 algorithms have also produced data for species not previously reported by MLS: methyl cyanide (CH_3CN) and water vapor in the upper troposphere (note that the latter was also produced by the "Version 4.9" (v4.9) algorithms). Sulfur dioxide (SO_2) abundances, although part of the version 4 MLS data set (v4), are not produced by the v5

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algorithms because of the similarity of the SO_2 spectral signature to that of methyl cyanide.

[4] In addition to the ''main'' part of this paper, a supplement[1] gives more details on the topics described in the following sections. For ease of reading, the section numbering in the supplement follows that in the main paper. References to supplementary material (sections, equations, figures, etc.) are all prefixed with a capital S.

2. UARS MLS Instrument and Operations

[5] Details of the MLS instrument are given by *Barath et al.* [1993]. It contains three radiometers (R1, R2 and R3) measuring the microwave emission spectrum near 63, 205 and 183 GHz, respectively. These combine the signal from the atmospheric limb with a local oscillator signal in nonlinear mixers employing Shottky diodes. This combination yields an intermediate frequency (IF) signal, corresponding to a combination of the radiances in the lower and upper frequency sidebands of the radiometer (i.e., above and below the local oscillator frequency). These IF signals are divided into six bands, chosen to observe emission lines for molecular oxygen (band 1 from R1), chlorine monoxide (bands 2 and 3 from R2), ozone (band 4 from R2, and band 6 from R3), and water vapor (band 5 from R3). The radiances in each band are measured by one of six nominally identical spectrometer filterbanks, each consisting of 15 contiguous channels, covering up to ±255 MHz away from the line center. The channels vary in width from 2 MHz near the line center to 128 MHz in the wings.

[6] In normal operation, MLS makes a ''step and stare'' scan of the Earth's limb from around 1 km to 90 km tangent point altitude every 65.536 s, one MLS Major Frame (MMAF). The MMAFs consist of 32 MLS Minor Frames (MMIFs). Most of the 2.048 s duration of each MMIF is dedicated to limb observations (the remainder is used to step to the next tangent view). Some MMIFs of each scan are used for views of space or a calibration target and/or antenna retrace activities.

[7] The UARS orbit and MLS viewing geometry are such that MLS observes from 34°N to 80°S for a period of about 36 days (one ''UARS month''), at which point the spacecraft performs a 180° yaw maneuver, changing to an 80°N to 34°S observing range.

[8] The Appendix to the supplementary material gives a summary chronology and calendar of MLS operations and data coverage. The main events of note were the mid-April 1993 failure of the 183-GHz radiometer, resulting in the loss of stratospheric water and 183-GHz ozone observations, and the June 1997 cessation of 63-GHz observations in order to save spacecraft power, resulting in a loss of the temperature information. The frequency of MLS operational days has generally decreased over the mission, from close to 100% from late 1991 though 1993 (the primary mission duration), down to about 50% in 1994, and only a few tens of measurement days per year at most from 1995 onward.

[1] Auxiliary material is available via Web browser or via Anonymous FTP from ftp://ftp.agu.org/apend/jd/2002JD002273/; subdirectories in the ftp site are arranged by paper number. Information on searching and submitting electronic supplements is found at http://www.agu.org/pubs/esupp_about.html.

[9] The MLS data processing is divided into separate ''Levels.'' Level 0 data are raw instrument data. Level 1 data are calibrated instrument radiance observations and engineering data. The radiance data form the input for the Level 2 data processing which produces estimates of geophysical parameters along the tangent point track. These data are stored in Level 2 files, and in the Level 3A files, which are a common storage format for the UARS instruments. The official repository for the v5 UARS MLS data is the NASA Goddard Space Flight Center (GSFC) Distributed Active Archive Center (DAAC).

3. Theoretical Basis

[10] The version 5 Level 2 algorithms are based on the optimal estimation approach [*Rodgers*, 1976, 2000]. A key part of this approach is the use of forward models to estimate MLS radiance observations corresponding to a given estimated state. The v5 algorithm mainly makes use of two different forward models; one is a complete line-by-line radiative transfer model, and the other is based on a Taylor series computation using precomputed output from the full model. In addition to the radiance information, the tangent height data are used in a hydrostatic model to obtain additional information on tangent pressure, temperature and geopotential height.

[11] Full details of these algorithms can be found in the supplementary material, section S3.

4. Implementation of Algorithms

[12] The standard products of v5 are temperature, water vapor, ozone separately from the 205-and 183-GHz radiometers, nitric acid, chlorine monoxide, and methyl cyanide. No v5 183-GHz O_3 data are produced from observations following the failure of the 183-GHz radiometer in April 1993. However, water vapor data are still produced, as the tropospheric H_2O observations were not affected by the 183-GHz failure. The stratospheric H_2O values (pressures of 100 hPa or less) for the post-April 1993 period are set to a priori and should not be used in scientific study. Sulfur dioxide data were produced by the MLS v4 algorithms, but are not retrieved in v5, because of the similarity between the spectra of sulfur dioxide and methyl cyanide.

[13] The main data files produced by the version 5 software are those in the UARS standard Level 3AT and Level 3AL formats, one of each for each species per day of observation. The Level 3AT files contain data taken directly from the retrieval state vector (in some cases interpolated in pressure, see below). The Level 3AL files are a linear interpolation of the Level 3AT data along the tangent track to standard latitudes. Information on the format and use of these files is given by *Burke and Lungu* [1996] (available from the MLS web site at http://mls.jpl.nasa.gov/). Both sets of files contain data on a subset of the standard ''UARS'' pressure surfaces, which are evenly spaced at a resolution of six surfaces per decade change in pressure. For the most part, these are the same surfaces as are represented in the state vector. However, in the lower troposphere and upper mesosphere, the state vector resolution is lower, at three surfaces per decade change in pressure; the output data at the intermediate surfaces represent a linear interpolation

between the adjacent levels. Note that the v4 algorithms only retrieved data at three surfaces per decade. The v4 Level 3A data on the intermediate surfaces were all produced by interpolations from adjacent levels.

[14] In addition, Level 3 "Parameter files" (Level 3TP and Level 3LP [*Burke and Lungu*, 1996]) are produced for each day of MLS observations. These files contain information on the quality of the MLS data in the 3AT and 3AL files, along with integrated column amounts estimated from the 3A data. The use of the quality flags found in these files is discussed in section 5.

[15] The software also produces Level 2 files for each day. These contain all the elements of the state vector used in the retrieval, including the species output at Level 3A, along with additional diagnostic information (χ^2 values, etc.). A Level 2 data file specifically describing the details of the upper tropospheric water vapor retrieval is also produced. This is a text file whose format is described in its header. It is very similar to that produced by the version 4.9 upper tropospheric humidity software (see section 4.3 of *Read et al.* [2001]), and is discussed in section S9.

[16] The supplementary material (section S4) gives more information on the implementation of the retrieval algorithms, including the sources of a priori data, and details of the configuration of the software (vertical retrieval ranges, minor species considered, etc.).

5. Proper Use of MLS Data

[17] Understanding the quality of the MLS data is essential for valid scientific use. Each data point in an MLS Level 3AT and 3AL file has an associated precision. As described in section S3.4, these precisions are flagged with a negative sign when they are no better than 50% of the a priori precision, indicating that the data should not generally be used. In addition, the precision is set negative for the 100 hPa stratospheric water vapor data, as these are tightly coupled to the surfaces above through the a priori smoothing. As in previous versions of MLS data, the retrieved points in the data files should be interpreted as the breakpoints of a piecewise-linear representation of the vertical profile.

[18] The appropriate parameter files (Level 3TP or 3LP) should always be used in conjunction with MLS Level 3A data. These contain information for each Level 3AT/3AL profile. The MMAF_STAT field contains a single-character flag that indicates the status of the instrument, as it impacts each profile, according to Table 1. Only profiles for which MMAF_STAT is set to G, T, or t should be used. In addition, the Level 3 parameter files contain the five fields QUALITY_TEMP, QUALITY_CLO, QUALITY_O3_205, QUALITY_O3_183, and QUALITY_H2O. These describe the "quality" of the corresponding profiles according to the values given in Table 2. Only profiles with QUALITY_... = 4 should be used. The QUALITY_O3_205 flag also describes the quality of the nitric acid data, with QUALITY_CLO applying to methyl cyanide.

Table 1. Values of MMAF_STAT in the MLS Level 3 Parameter Files and Their Associated Meaning

MMAF_STAT	Meaning
G	The profile is based on all "Good" radiance data
t	Temperatures missing from NCEP[a] data at pressures greater than 22 hPa
T	Temperatures missing from NCEP[a] data at pressures greater than 100 hPa
M	Too many tangent points are missing from scan
P	A pointing anomaly occurred during the scan
S	Scan mode anomaly (e.g., not normal full scan range)
B	Bad or insufficient radiance data were taken

[a]National Centers for Environmental Prediction.

Table 2. Values of QUALITY_... in the MLS Level 3 Parameter Files, With Their Associated Meaning

QUALITY_...	Meaning
4	Good fit to good radiances
3	Good fit to poor radiances
2	Poor fit to good radiances
1	Poor fit to poor radiances

[19] In addition to the information available from the data files, the MLS science team has inspected the quality of the v5 data set on a UARS-monthly basis. The study involves examination of time series data and of the location and magnitude of "spikes," and the amount of good data available each UARS month. Each UARS month of MLS data has been assigned a grade. These are summarized, along with general comments on each month, on the MLS science team web site.

[20] To summarize, the general caveats for the use of MLS data are as follows: (1) Only data whose associated uncertainty is positive should be used. (2) Only profiles where the MMAF_STAT field is set to G, T, or t should be used. (3) Only profiles where the appropriate QUALITY_... is equal to 4 should be used. (4) The spike information given on the MLS science team web site should be consulted.

[21] These quality control measures do not always filter out large ($>5\sigma$) "spikes"; such occasional anomalous retrievals can be identified by inspection and removed on an individual basis.

6. Validation and Characterization Issues Common to All Species

6.1. Precision Versus Scatter

[22] The Level 3AT and Level 3AL files contain uncertainty values for all data points. These are the square roots of the corresponding diagonal elements of the solution error covariance matrix from equation (S6). These describe a combination of the projection of the radiance uncertainty into state space and the assumed a priori uncertainty. Generally, these values should be interpreted as a measure of the precision (i.e., random error) of the v5 data (the exception is upper tropospheric humidity where the value reported is more descriptive of the accuracy, as described in section S9).

[23] One measure of true precision is the scatter observed in the data in regions where little atmospheric variability is expected (e.g., the tropical stratosphere for some species). Such a measure indicates that the precision of the data is better than is estimated by the algorithms. This is because the scatter in the data points arises purely from radiance terms. The a priori data are generally constant, as they are zonal mean or single profile data for all the fields except

temperature and geopotential height, for which National Center for Environmental Prediction (NCEP) data are used. The size of the precision "overestimate" is determined by the a priori error covariance matrix described in section S3.2. The diagonal terms in this matrix describe the confidence in the a priori data. The off-diagonal terms lead to a preference for smoother solutions. The latter factor had a significant effect in v5.

[24] For many of the v5 data products, the observed scatter is ∼70% of that estimated by the algorithms and placed in the Level 3AT/Level 3AL files. The ratios between the typical estimated uncertainties and the observed scatters are listed as a function of pressure for each species in later sections of this paper. The precisions quoted in the data files vary very little as a function of latitude or time. However, they do take into account occasional variations in instrument performance and vertical coverage, in a manner that a single profile summary cannot. The "best estimate" of the precision of a single data point is the quoted uncertainty on that point given in the data file, multiplied by the ratios reported for each species in later sections of this paper.

6.2. Vertical Resolution

[25] The definition of vertical resolution chosen here is the full width at half maximum of the rows of the averaging kernel matrix given by equation (S10). These have been scaled from log pressure coordinates into approximate kilometers (using a scale height of 16 km per decade change in pressure) for clarity. The quoted averaging kernel widths are taken from the retrieval of the first profile on 17 September 1992, which is typical of the data set.

6.3. Accuracy of Retrieval Estimates

[26] We use the term accuracy to describe systematic errors in the v5 data. These accuracies vary from species to species, and are described in later sections. Sources of uncertainties in accuracy include the following: (1) uncertainties in spectroscopic parameters, (2) uncertainties in instrument calibration, (3) uncertainties in spacecraft attitude, and (4) biasing toward a priori information.

[27] The magnitude of some systematic uncertainties can be estimated by mapping an estimated uncertainty in spectroscopic and/or calibration parameters into state space. Sometimes the magnitudes can be estimated from comparisons with other data sets, or with a priori information (e.g., knowledge that nighttime lower stratospheric ClO abundances are negligible except in certain situations).

6.4. Further Issues

[28] Section S6 gives more information on the general characterization of the v5 data; in particular, it discusses the impact of the deactivation of the 63-GHz radiometer in June 1997 and the characteristics of the v5 tangent pressure data, which are key to the retrievals of all other parameters.

7. Temperature

7.1. Changes in Algorithms for v5 Temperature

[29] The v5 software produces scientifically useful temperature data over the vertical range 32–0.46 hPa at an interval of six surfaces per decade change in pressure (the UARS standard surfaces). The temperature profile at pressures of 100 hPa and higher is constrained to the a priori values (NCEP or climatology, as described in section S4.1). The data at pressures of 68, 46, and less than 0.46 hPa are not scientifically useful, because of the poor MLS temperature sensitivity in these regions.

Table 3. The v5/v4 and v5/v3 Temperature Differences

	v5 − v4, K			
Pressure, hPa	Global	Tropics	Polar Winter	v5 − v3 Global
0.46	+0.0	−0.6	+2.0	−0.9
1.0	−0.7	−0.4	−2.1	+3.2
2.2	+3.0	+2.2	+3.2	+3.9
4.6	+1.7	+1.7	+0.6	+4.9
10	+2.8	+2.3	+2.8	+4.2
22	+2.8	+1.3	+3.1	+3.1

[30] The v4 algorithms attempted to obtain useful information at 46 hPa, by using a looser a priori error (20 K throughout the vertical profile). Results contained a disappointingly-large number of spikes. V5 adopts a somewhat conservative approach by reducing the a priori uncertainty of temperature to 10 K from 68–3.2 hPa and gradually increasing it to 46 K between 3.2 and 0.0001 hPa (linearly changing with log pressure). Since the v4 algorithms retrieved temperature on coarser pressure grids (three surfaces per decade change in pressure), but reported the retrieval on every UARS surface, the output data on the intermediate surfaces represent the results of an interpolation. Differences will thus be observed between v4 and v5 temperatures at these intermediate surfaces, even at pressures larger than 68 hPa where the only source of temperature information is a priori.

[31] Section S7.1 discusses the mesospheric temperature data produced by v5. These data are only a research product and not considered useful for scientific study.

7.2. Comparison of v5, v4, and v3 Temperatures

[32] In the stratosphere, the v5 temperatures are generally warmer (by 1–3 K) than v4, but v5 is cooler than v4 (by ∼1 K) near the stratopause. These differences reduce the "sharpness" of the retrieved stratopause, which was often too sharp in v4 by comparison to climatology. Table 3 shows v5/v4 and v5/v3 differences, based on the first year of observations. The largest v5/v4 differences are seen in polar winter conditions, where planetary wave activity is strong.

7.3. Estimated Precision and Accuracy of v5 Temperatures

[33] The estimated precision, accuracy and resolution (as defined in section 6.2) of v5 temperatures are given in Table 4. Precisions (1σ) are estimates obtained by computing the observed variability for profiles in the 20°S to 20°N latitude band (from October 1991 to September 1992). Uncertainties in the Level 3 files should be used in conjunction with the ratio column in this table as described in section 6.1 to obtain the best estimate of the precision of each measurement.

[34] Accuracy is estimated from the error analysis described by *Fishbein et al.* [1996]. One observed artifact is a systematic error of ∼0.5 K between ascending and descending measurements that is synchronized with the UARS yaw cycle. This error is evident even in the presence of the diurnal and semi-diurnal tides because of its incoherent character. The presence of yaw-cycle synchronized error

Table 4. Estimated Vertical Resolution, Precision, and Accuracy of v5 Temperature

Pressure, hPa	Vertical Resolution,[a] km	Typical Precision, K	Precision Ratio[b]	Estimated Accuracy,[c] K	v5−NCEP, K
0.46[d]	5	3.3	0.7	5	−1.5
0.68	7	2.1	0.5	5	+5.2
1.0	7	1.8	0.5	5	+8.6
1.5	7	1.7	0.5	5	+6.6
2.2	7	1.5	0.5	4	+4.3
3.2	7	1.5	0.5	4	+1.4
4.6	6	1.4	0.5	5	+0.9
6.8	7	1.4	0.5	4	+0.1
10	7	1.3	0.4	4	+1.1
15	6	1.2	0.4	4	+1.1
22	7	0.8	0.3	4	+1.6
32	7	0.9	0.3	6	+2.0

[a]As defined in section 6.2.

[b]Data file uncertainties should be multiplied by these numbers to obtain a better value for the "1σ" single profile precision (see text).

[c]Accuracies quoted here roughly represent a 95% confidence level ("2σ" values).

[d]The temperature at 0.46 hPa mainly derives from optically thin radiances. These yield information of poorer precision but slightly better resolution than the optically thick radiances that influence the temperature data lower in the atmosphere.

may cause serious problems for studies of short-period atmospheric waves. In the v5 temperature this artifact is reduced by about half from ~1 K seen in v4, but users should be cautious about temperature variations near or below 0.5 K.

[35] Comparisons of the first year's data to NCEP show a global warm bias in the v5 temperature. This bias is less than 2 K at 32–3.2 hPa but 4–9 K near the stratopause (2.2–0.68 hPa). In addition, v5 shows a 1.5 K cold bias at 0.46 hPa. The warm bias of v5 compared to NCEP is greater than that seen in v4 and NCEP by 0.5–1 K.

7.4. Caveats in Use of v5 Temperature

[36] Caveats in the use of v5 temperature are as follows: (1) See the general caveats given in section 5. (2) Only temperature data for pressures between 36 and 0.46 hPa should be used in scientific study. (3) Temperature data following the deactivation of the 63-GHz radiometer (June 1997) should not be used.

8. Geopotential Height

[37] Version 5 is the first MLS algorithm to give geopotential height (GPH) as a standard product. GPH is retrieved in a somewhat different manner from the other products. The state vector contains the GPH of the 100-hPa reference surface, which is retrieved collectively from the 63-GHz radiances and the tangent height information. The linear radiance model and scan model, described in section S3.8, provide the forward models in this retrieval. The GPH values above and below 100 hPa are computed from this reference GPH using a standard hydrostatic integrator (including the gas constant model described by equation (S37)) and the retrieved temperature profile.

8.1. Accuracy and Precision of GPH Data

[38] The GPH accuracy and precision behave in a very different manner from that of other retrieved products. The GPH error comes from two distinct sources. The first is associated with the accuracy and precision of the retrieval of the 100-hPa GPH that is used to "anchor" the GPH profile. The second source is the accuracy and precision of the retrieved temperatures used in the hydrostatic integration to compute the whole profile from the 100-hPa value.

[39] The 100 hPa GPH precision depends mostly on knowledge of the MLS pointing. Random pointing errors are thought to be about 100 m (based on studies of the attitude data provided by the UARS orbit/attitude services) in each tangent point altitude. Since the 100-hPa GPH retrieval is based on the measurements of ~26 tangent points, the precision is expected to be better than the single-pointing precision.

[40] Accuracy is harder to assess, as it is dependent on knowledge of UARS attitude, the uncertainty of which is hard to characterize. However, comparisons with correlative data sets can yield some insight into the accuracy of MLS GPH. Figure 1 compares daily-averaged MLS and NCEP GPH near the equator, where wave activity is relatively low in the lower stratosphere. At 100 hPa the NCEP GPH typically shows variations of less than 100 m around ~16.5 km, while the MLS values vary over 1 km and occasionally 2–3 km. This suggests that an upper limit of MLS GPH accuracy would be about 1.5 km over the measurement period. During some spacecraft/instrument testing periods (such as UARS Days 275–300 and 1605–1639), the MLS GPH accuracy can be as poor as 3 km. The GPH accuracy also degrades slightly with height because of the accumulated uncertainty in the temperatures used in the hydrostatic integration.

[41] The estimated single-profile GPH precisions (1σ) vary from 70 m at 100 hPa to 220 m at 0.01 hPa, based on the variability of MLS GPH measurements between 20°S and 20°N from October 1991 to September 1992. The MLS GPH precision is much better than its accuracy, as is shown by the good tracking between the NCEP and MLS layer thicknesses in Figure 1. MLS GPH difference between pressure surfaces (layer thickness) is less prone to the bias imposed on the 100-hPa GPH. The offsets in the thickness are due to temperature differences between the two data sets.

8.2. Caveats for Using GPH Data

[42] Given the rather poor GPH accuracy, users need first to remove the potential bias in each profile. One may use the 100-hPa value to quantify such a bias as shown in Figure 1 with the NCEP data. The disadvantage of this approach is that some atmospheric variability will be lost by subtracting out the 100-hPa value. Users should disregard the uncertainty values quoted in the Level 3 files and use the estimated uncertainty values given in the previous paragraph. GPH data after June 1997 should not be used as 63-GHz observations were not made during this period.

9. Upper Tropospheric Humidity

[43] A full description of the MLS observations of upper tropospheric humidity (UTH) is given by *Read et al.* [2001]. Here we concentrate on v5 UTH. It is recommended that v4.9 data be used in preference to v5 because the v4.9 water vapor continuum function in air is believed to be superior. This function is essential for the UTH measurement and had

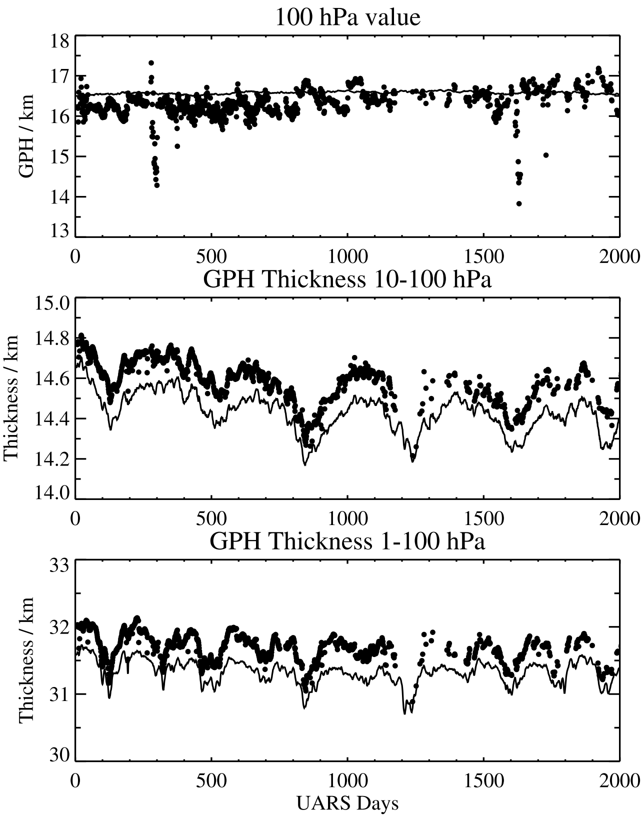

Figure 1. Time series of daily mean MLS (dot) and NCEP (line) GPH at 20°S–20°N. The MLS data are averages of all the ascending orbits each day, whereas the NCEP values are a zonal mean at 1200Z. The ability of MLS to track GPH thicknesses indicates the potential use of these data for scientific study.

to be inferred from MLS data, because no known laboratory measurements existed as of 1998. A derivation of the water vapor continuum function requires knowledge of humidity. For a given tangent height, the majority of MLS measured radiances fall between two distinct brightnesses. The v4.9 H_2O continuum function was derived by assuming that the upper brightness boundary was in an atmosphere having a relative humidity of 100% with respect to ice (%RHi) and with no significant emissions from cirrus ice. The v5 water vapor continuum function used humidity measurements from Vaisala radiosonde measurements that were coincident with MLS observations. Following the production of v5 data, the accuracy of Vaisala radiosonde observations of the uppermost troposphere was significantly called into question by *Miloshevich et al.* [2001], though this claim is not supported by comparisons between Vaisala sonde and MLS v4.9 observations [*Read et al.*, 2001]. As MLS cannot observe thin cirrus, the method for establishing the v4.9 water vapor continuum appears more robust. These issues are discussed more fully by *Read et al.* [2001]. However, no v4.9 data are available after June 1997, when 63-GHz observations were discontinued. V5 data are usable up to June 1998, after which severe instrument scanning problems led to a significant reduction in the amount of UTH data. Also noteworthy in this period is the observation of a significantly lower retrieved UTH (in %RHi) over the poles during winter than had been seen in previous years; this could be an artifact of the data processing.

[44] More details of the v5 UTH data set are given in the supplementary material (section S9).

10. Ozone From 205-GHz Radiometer Data

[45] UARS MLS ozone data from the 205-GHz radiometer (O3_205) have been obtained over the lifetime of the instrument (with very limited data from 1998 to 2001), whereas the 183-GHz radiometer ozone (O3_183) data ended at the mid-April 1993 failure of that radiometer. We have therefore never combined these two retrievals, and discuss them separately. This is also convenient because we recommend O3_205 for studies of stratospheric ozone but O3_183 for studies of mesospheric ozone, given the better sensitivity (stronger line) for the O3_183.

[46] Information on v3 O3_205 data is given in the MLS ozone validation paper [*Froidevaux et al.*, 1996] and by *Cunnold et al.* [1996a, 1996b]. MLS v4 data quality and related studies have been presented by *Harris et al.* [1998] and *Cunnold et al.* [2000]. The various data versions have also been described in the MLS "Data Quality Documents" available on the MLS web site and distributed by the GSFC DAAC.

[47] Here, we summarize the changes that occurred for the v5 O3_205 data and give estimates of v5 precision and accuracy, using comparisons with Stratospheric Aerosol and Gas Experiment II (SAGE II) version 6.1 data and other reliable ozone data sets.

10.1. Changes in Algorithms for v5 205-GHz Ozone

[48] The main change in v5 O3_205 is the use of a finer retrieval grid (see Introduction) below 0.1 hPa. While this can lead to better vertical discrimination, it also generally leads to somewhat poorer precision. Except at 100 hPa, where the precision is better than v4, v5 stratospheric ozone data are generally noisier than v4 data (typical precision is 0.3 ppmv rather than 0.2 ppmv). Better mesospheric precision is obtained in v5, largely because of more precise tangent pressure estimates. The recommended vertical range for use of v5 O3_205 extends from 100 to 0.2 hPa.

[49] The v5 retrievals use radiances with tangent pressures as great as 150 hPa, which can lead to more contamination by clouds, especially in the tropics. Indeed, we find that the spatial distribution of profiles flagged as having poor quality (based on the "QUALITY_O3_205" parameter) appears to correlate with regions of upper tropospheric convection and with cloud ice; these profiles generally show oscillatory behavior in the lower stratosphere, with negative values at 68 hPa and excessively large values at 100 hPa. Compared wih v4 data, about twice the amount (or ~2%) of profiles are flagged as poor overall in v5 data.

10.2. Comparison of Different Data Versions for 205-GHz Ozone

[50] Table 5 provides average differences between O3_205 data versions. Separate comparisons are made for different latitudinal conditions, as noted in the table, for the first 10 full UARS months (essentially for October 1991 through September 1992). Because v4 (and v3) retrievals were performed only on the even UARS surfaces, only the

Table 5. Average Differences Between O3_205 Data Versions

	v5/v4 Differences						v5/v3 Differences Global	
	Global[a]		Tropical[b]		Midlatitude[c]			
Pressure, hPa	ppmv	%	ppmv	%	ppmv	%	ppmv	%
0.46	0.0	0	−0.05	−3	0.0	0	−0.03	−2
1.0	+0.2	+7	+0.3	+10	+0.2	+6	+0.1	+3
2.2	−0.1	−2	−0.1	−2	−0.1	−2	−0.2	−3
4.6	−0.1	−1	0.0	0	−0.1	−2	−0.2	−2
10	−0.1	−1	−0.3	−3	0.0	0	−0.3	−3
22	−0.1	−1	−0.2	−3	−0.1	−1	−0.4	−6
46	+0.6	+37	+1.1	+300	+0.5	+20	+0.2	+9
100	−0.5	−53	−0.9	−82	−0.4	−41	−0.1	−15

[a]Based on ~400,000 profiles from all latitudes for the first full year of data (October 91 through September 92).
[b]Based on ~60,000 profiles from 10°S to 10°N for the first full year of data.
[c]Based on ~25,000 profiles from 35° to 4°N and 35° to 45°S for the first full year of data.

differences on these surfaces are tabulated. V5 data exhibit an overall decrease from v4 of 1 to 3% between 10 and 2 hPa, with a 5 to 10% increase at 1 hPa. The lower stratosphere shows the largest differences, particularly in the tropics; v5 values are systematically larger than v4 at 46 hPa (by about 0.5 to 1 ppmv) and smaller at 100 hPa (by about 0.5 to 1 ppmv). In the polar regions, lower stratospheric differences (not shown in the table) are generally smaller than the midlatitude differences (the decrease from v4 to v5 at 100 hPa is often only 10 to 20%). Polar v5 values are about 2% larger than the v4 values at 10 hPa, and typically 5 to 10% larger at 0.46 hPa; at other pressures, differences in the polar regions are similar to those listed for midlatitudes.

[51] The above changes have a strong systematic component, remaining fairly constant through the years. Linear trends of the differences between the two data versions (for late 1991 to mid-1997) give slopes generally well within 0.2%/yr (with little statistical significance). V5 "trends" are slightly larger than v4 between 22 and 2 hPa; somewhat larger differences (up to a few %/yr) exist at 46 and 100 hPa.

[52] The vertical profile of the v5 ozone rate of decrease during Antarctic ozone hole conditions is different from v4. Figure 2 shows the zonal mean ozone changes for 80°S to 70°S at 46 and 100 hPa for the time period from 14 August to 20 September 1992. While the sum of the mixing ratios at these two levels does not change much between the two data versions, the v5 retrievals yield more of a decrease at 100 hPa and less at 46 hPa. The v5 changes shown in this figure are very well reproduced by the independent O3_183 v5 retrievals, although those values (not shown) are larger by about 0.2 ppmv. Ozone changes are small at lower pressures (and zonal mean values are roughly constant at 22 and 10 hPa during this time period). *Wu and Dessler* [2001] have found that the ozone rates of decrease based on MLS data in the Antarctic polar winter (for 1992, 1993, and 1994) agree well with calculations based on the MLS ClO measurements [see also *MacKenzie et al.*, 1996]. The results of *Wu and Dessler* [2001] applied to v4 MLS data interpolated to 465 K potential temperature. Their main conclusion regarding agreement between measured and modeled rates of ozone decrease would remain valid if MLS v5 data were used, because although v5 data yield a 25% smaller ozone decrease at 465 K, reductions in MLS ClO lead to a similar change in the modeled ozone decrease (J. Wu and A. Dessler, private communication, 2001). MLS ozone comparisons with McMurdo ozonesonde data for August–September 1992 (not shown) confirm that the slower decrease in v5 data at 46 hPa is very similar to the observed decrease for the ozonesonde data and agrees better than does v4 data; also, ozonesonde values at 100 hPa show a small decrease that is consistent with v5 values, but not with v4 abundances, which are too large and actually increase during this time period.

[53] Changes from v4 to v5 MLS ozone data for the Arctic winter are typically not as large as those shown above for Antarctica (and the two data versions tend to track better).

10.3. Validation of v5 205-GHz Ozone

10.3.1. Comparison of 205-GHz Ozone Data With Other Data Sets

[54] We now discuss how the v5 O3_205 data compare with a few other ozone data sets, mainly the SAGE II version 6.1 results. The SAGE II data have been used extensively in the past and compare quite well with accurate ozonesonde profiles (see *Harris et al.* [1998], for example,

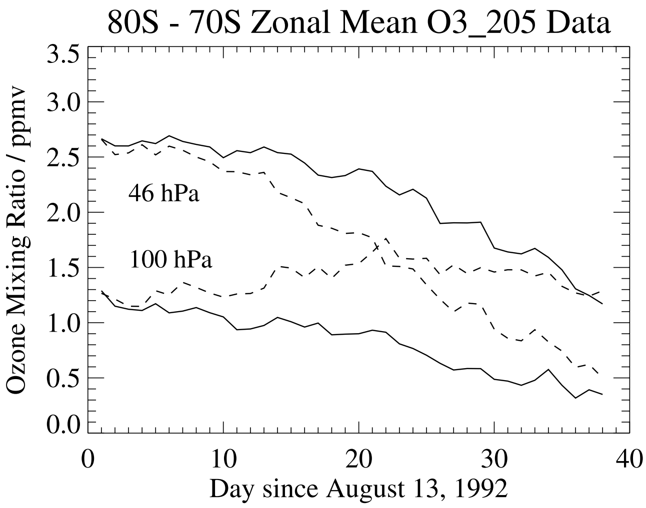

Figure 2. Zonal mean (80°S to 70°S) ozone changes during the 14 August to 20 September 1992 time period, based on MLS O3_205 retrievals for v5 (solid lines) and v4 (dashed lines). MLS retrievals for 100 and 46 hPa are shown. Standard errors in these mean values (averages of about 100 profiles) are roughly 0.04 to 0.08 ppmv.

for comparisons based on version 5.96 SAGE II data). We have analyzed average differences between these versions of MLS and SAGE II data by combining coincident profiles (profiles within 2° latitude and 12° longitude, and for the same day) for various latitude bins and time periods. Average results from the time period 1995 through 1996 for various latitude bins are shown in Figure 3. These years have much smaller potential impact from the Mount Pinatubo volcanic aerosols on SAGE II retrievals than earlier years and still contain a significant amount of MLS ozone data. Other years are discussed below and do not change the first-order results regarding systematic differences. Our comparisons include SAGE II profiles from both sunset and sunrise occultations. We have screened SAGE II data discussed in this paper for ("transient") poor quality profiles by omitting all profiles with error bar larger than 10% of the ozone abundance in the midstratosphere to upper stratosphere (per a recommendation by R. Wang (private communication, 2001)).

[55] The average agreement between SAGE II and MLS profiles is generally within 0.15 ppmv for pressures larger than 10 hPa and within 0.3 ppmv elsewhere, or typically within 5% overall. The largest percentage differences are observed at low latitudes for the lowest MLS retrieval point (100 hPa), with MLS abundances there larger than SAGE II by over 30% (see the 30° S–30°N latitude bin results in Figure 3 for 1995 through 1996). It is difficult to collect enough independent data in the tropics to ascertain the relative merits of SAGE II and MLS lower stratospheric profiles in that region. MLS v4 differences with SAGE II coincident profiles are compared to the differences for v5 (during 1995–1996) in section S10. V5 shows a significant reduction in the average difference with SAGE II, particularly at low latitudes in the lower stratosphere. In general, v5 midstratospheric to upper stratospheric ozone retrievals are slightly larger (by only a few percent) than the SAGE II (version 6.1) values. This small offset has decreased slightly from v4 MLS and SAGE II (version 5.96) comparisons [*Harris et al.*, 1998]. More details for the lower mesosphere are provided in section S10.

[56] Figure 4 shows monthly mean differences between MLS v5 and SAGE II version 6.1 coincidences from October 1991 to June 1998 for different latitudes and pressures. Larger differences occur in the lower stratosphere (68 hPa), primarily before 1993; the largest mean differences are in the tropics, as high as 22 hPa (see middle panel). While the Mount, Pinatubo aerosol had an impact on the SAGE II retrievals [see *Cunnold et al.*, 1996b], and a number of SAGE II measurements are flagged (or not retrieved) because of these effects, it seems that there are still aerosol-related effects at most latitudes (for pressures larger than about 15 hPa) on some of the remaining (unflagged) SAGE II version 6.1 profiles; we do not see such a time-dependent effect in MLS versus ozonesonde comparisons. Also, it is likely that the increase in scatter after mid-1997 in Figure 4 comes from the changeover to a different MLS operational and retrieval mode (and to the lack of MLS profiles). Apart from these effects, the MLS and SAGE II ozone retrievals track quite consistently through most of this nearly 7-year time period. There are significantly fewer coincidences at latitudes higher than 60° (north or south), but nothing abnormal appears in those differences (not shown here).

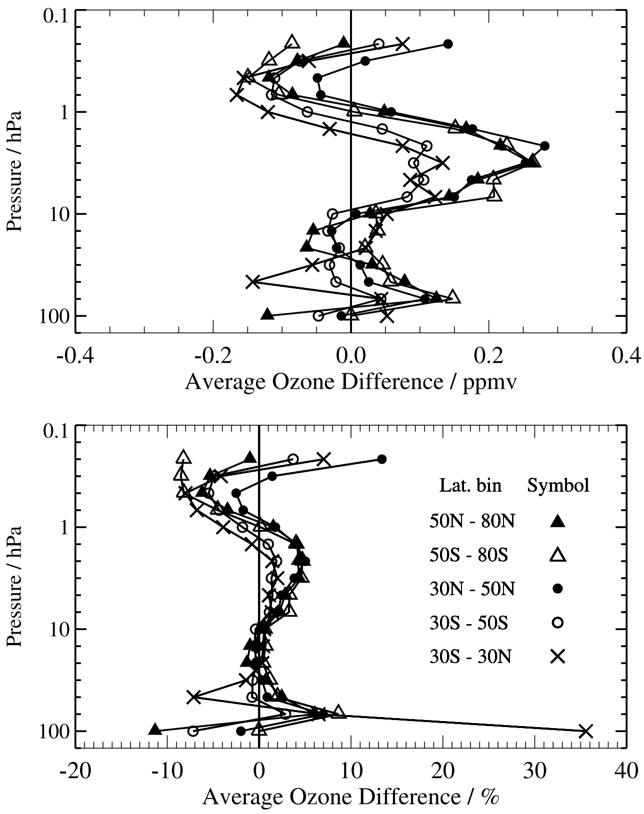

Figure 3. Average ozone differences between MLS O3_205 and coincident SAGE II profiles for the time period 1995–1996 (top panel for ppmv, bottom panel for percent differences) over different latitude ranges given in legend of bottom panel. Differences are MLS (v5) minus SAGE II (version 6.1) values (and percent differences are relative to SAGE II values).

[57] There are also significant improvements (over v4) in the agreement between v5 MLS profiles and tropical ozonesonde data from Ascension Island and Brazzaville. Section S10 discusses data that show much smaller differences between MLS v5 and these ozonesonde averages (typically less than 0.1 ppmv for the average of about 25 total available coincidences for late 1991 through 1992) than for v4. V3 data, not shown here, are also in poorer overall agreement with the sondes than v5. The v3 data also tend to have lower values at 46 hPa than the ozonesonde data for the time period prior to June 1992 [*Froidevaux et al.*, 1996]. This is not the case in v5 for Ascension Island, as shown in section S10, nor for Brazzaville (not shown). The MLS v5 ozone values between 46 and 10 hPa are larger than tropical ozonesonde values by 2 ± 2%, well within the expected combined accuracies (of order 5%).

[58] One possible source of differences between SAGE and MLS profiles at low latitudes is the positive bias introduced in MLS ozone data at 100 hPa by the presence of dense ice cloud. Not all the profiles affected by clouds have been flagged as "bad" by the v5 software, and the bias introduced by cloud has not been quantified. Another possible source of SAGE/MLS differences are the (small) inaccuracies in the SAGE II profiles. Average differences between MLS ozone v5 values and those obtained by the Jet

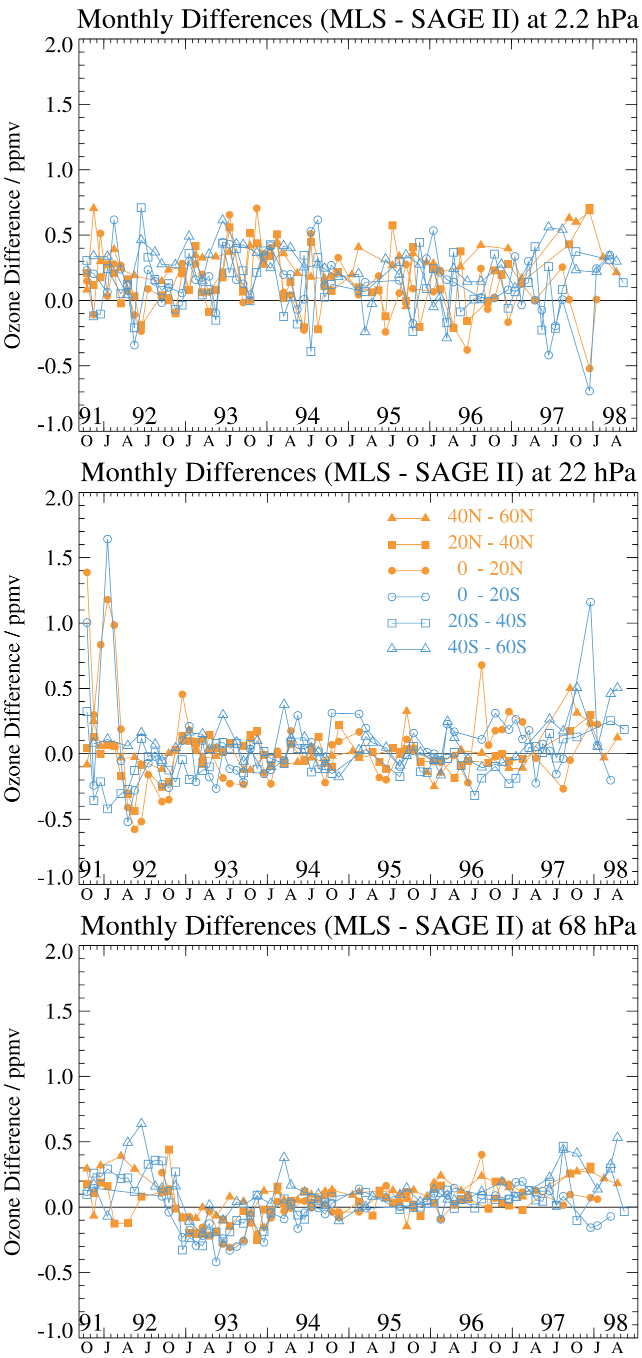

Figure 4. Time series of monthly mean differences between MLS v5 and SAGE II version 6.1 coincident ozone profiles; differences are calculated as MLS minus SAGE mean values. Six 20° wide latitude bins are shown, see legend in center panel; pressure levels are 2.2 hPa (top panel), 22 hPa (center panel) and 68 hPa (bottom panel). Capital letters below the abscissa indicate months (October, January, April, July).

Propulsion Laboratory's UV photometer instrument, during a series of 8 midlatitude balloon flights [*Froidevaux et al.*, 1996], are within 2% for pressures between 68 and 22 hPa, well within the combined accuracies. MLS values are about 4% larger than the photometer data at 15 to 5 hPa, consistent with the offset between MLS and SAGE II profiles in this region. Other comparisons for several different ozonesonde sites and for a larger number of ozonesonde coincidences (about five years of data) indicate average differences of about 5% or less for pressures less than or equal to 46 hPa, as shown in Figure 5. In the lower stratosphere, where mixing ratios can be small, the typical average differences between MLS and sondes are ~0.25 ppmv or less for 68 hPa, and less than 0.15 ppmv for 100 hPa.

[59] Based on the totality of the above MLS ozone comparisons with SAGE II and ozonesondes, we find a small positive bias (2 to 4%) in the v5 O3_205 data for midstratospheric to upper stratospheric regions. This offset is within the combined absolute errors and is therefore of

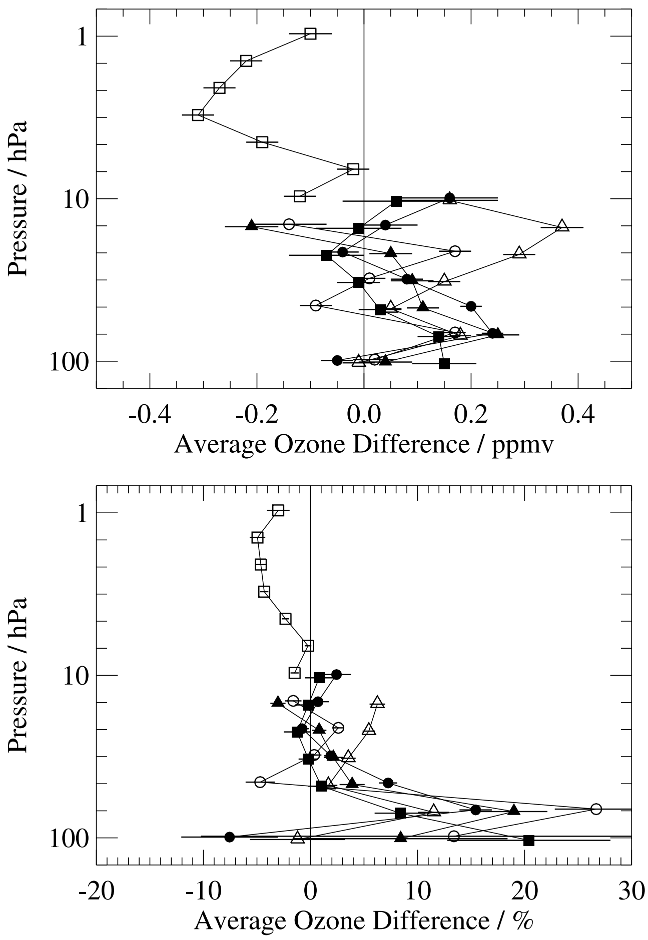

Figure 5. Average differences (top panel: ppmv, bottom panel: percent) between MLS v5 O3_205 and various sets of coincident correlative profiles covering about 5 years or more: ozonesonde data from October 1991 through 1996 for Hilo (20°N, 132 matched profiles, open circles), Boulder (40°N, 70 matches, closed triangles), Uccle (50°N, 126 matches, dots), Payerne (47°N, 201 matches, open triangles), and Lauder (45°S, 103 matches, closed squares), and lidar data from October 1991 through April 1996 for Table Mountain (34°N, 289 matches, open squares). Error bars give the standard errors for these average differences. Coincidences were defined as having latitude differences less than 2.5° longitude differences less than 12° and being on the same day.

marginal significance. The Table Mountain lidar data do not support such a bias in MLS data (or even the sign of this bias). Time series comparisons between MLS and correlative data (not shown here for brevity) give excellent agreement (typically within 5–10%) over seasonal ozone variations of up to a factor of two. The remaining average offset for the midstratosphere to upper stratosphere is essentially as good a result as one can expect. However, larger percentage uncertainties (random and absolute) exist for the MLS data at 68 and 100 hPa, with "2σ" accuracies estimated conservatively at about 0.25 ppmv or 15% (whichever is larger) for 68 hPa and 0.1 ppmv or 15% (whichever is larger) for 100 hPa. The MLS v5 ozone values at 68 and 100 hPa appear to be systematically larger than ozonesonde values by about 10–15%. The MLS/SAGE II comparisons give smaller average differences, which would imply that the SAGE II version 6.1 values are slightly larger than the ozonesonde data in at least parts of the lower stratosphere. MLS v5 precision and accuracy estimates are summarized below in section 10.4.

[60] *Danilin et al.* [2003] used trajectory calculations to increase the number of matches between MLS and other measurements during the northern winter of 1999/2000 (poleward of 50°N). Their results agree with those presented here, and yield v5 MLS ozone average values a few percent larger than those from SAGE II, although in most cases the differences are statistically consistent with zero. Based on *Danilin et al.* [2003], the MLS values are up to ~12% larger than those from the Polar Ozone and Aerosol Measurement (POAM) III; they are also larger than those from POAM II [*Manney et al.*, 2001]. *Manney et al.* [2001] compared MLS v5 ozone fields to a variety of other satellite data sets (mean values as a function of equivalent latitude as well as averaged coincidences) for November 1994; good agreement (often within ~5% in the upper stratosphere, and 0.25 ppmv in the lower stratosphere) was typically found in the morphology and absolute values. MLS values tend to be slightly on the high side of these average comparisons, although, based on the larger number of intercomparisons discussed here, we believe that a bias of no more than a few percent exists in the MLS results, except at 68 and 100 hPa.

10.3.2. The 205-GHz Ozone After 15 June 1997

[61] A major change in MLS operations was the cessation of 63-GHz observations after mid-1997, as described in section S6.1. MLS ozone data have been scrutinized for any degradation or discontinuities that may be tied to this deactivation or subsequent antenna scan slip problems. Regarding the changeover to operations using radiometer 2 only (after mid-1997), we have performed software tests with retrievals not using 63-GHz radiances, for days of normal operation. These tests indicate that, for pressures less than 46 hPa, retrieved ozone values are within a few percent of the standard retrievals. For 46 to 68 hPa, values are typically a few to 10% larger than in the standard case, and for 100 hPa, the test values are smaller than in the standard case by about 0.1 to 0.2 ppmv. There are indications that such small (and artificial) shifts do indeed exist after the actual transition from normal operations to single-radiometer mode, based on time series plots not shown here. Nevertheless, the MLS data from mid-1997 through mid-1998 agree with previous years' data to within a few to 10%; the same seems to hold for the late July 1999 Antarctic data, the February/March 2000 data (obtained at high northern latitudes only), and the 18–25 August 2001 data.

Table 6. Estimated Vertical Resolution, Precision, and Accuracy of v5 O3_205

Pressure, hPa	Vertical Resolution,[a] km	Estimated Precision ppmv	Estimated Precision %	Precision Ratio[b]	Estimated Accuracy[c]
0.22	6	0.4	35	0.6	6%
0.32	8	0.35	25	0.6	6%
0.46	5	0.35	20	0.6	6%
0.68	4	0.3	12	0.6	6%
1.0	5	0.3	10	0.6	6%
1.5	5	0.3	7	0.7	6%
2.2	4	0.3	5	0.7	6%
3.2	4	0.3	4	0.8	6%
4.6	4	0.3	4	0.8	6%
6.8	4	0.3	4	0.8	6%
10	3.5	0.3	4	0.8	6%
15	3.5	0.3	4	0.9	6%
22	3.5	0.3	5	0.9	6%
32	3.5	0.3	8	0.9	6%
46	3.5	0.25	10	0.7	6%
68	4	0.25	20	0.6	max. of 0.25 ppmv or 15%
100	4	0.4	>50	0.7	max. of 0.1 ppmv or 15%

[a]As defined in section 6.2.
[b]Data file uncertainties should be multiplied by these numbers to obtain a better value for the "1σ" single profile precision (see text).
[c]Accuracies quoted here represent roughly a 95% confidence level ("2σ" values).

[62] Based on the loss and degradation of MLS data after mid-1998, we recommend not using this time period as part of trend analyses, even if the ozone abundances appear reasonable to first-order (see the supplementary section S10). The time period from mid-1997 to mid-1998 yields seemingly much better results, but some caution should apply for this period as well.

10.4. Vertical Resolution, Precision, and Accuracy of v5 205-GHz Ozone

[63] Our estimates of O3_205 accuracy are based on the discussion in section 10.3. For most of the stratosphere (from 0.46 hPa down to 46 hPa), this accuracy is estimated at 6% or better, at the 95% confidence ("2σ") level. Despite improvements in the lower stratosphere, there are remaining limitations that do not allow for such good accuracy there, especially in the tropics where the abundances are low; the MLS 68-hPa data have an accuracy of 15% or 0.25 ppmv, whichever is larger, and 15% or 0.1 ppmv for 100 hPa. Table 6 gives these v5 accuracies for O3_205, along with the estimated vertical resolution and typical single-profile precision. These precisions are 1σ values, based on the minimum monthly individual profile variability for 5°S to 5°N during the first 10 full UARS months of the MLS mission (October 1991 through September 1992). As discussed previously, the estimated uncertainties in the O3_205 data files should be multiplied by the values given in the fifth column of Table 6 to obtain the best estimate of precision.

10.5. Known Artifacts and Systematic Effects in v5 205-GHz Ozone

[64] Known artifacts and systematic effects in v5 205-GHz ozone are as follows:

Table 7. Average Differences Between O3_183 Data Versions

	v5/v4 Differences						v5/v3 Differences Global	
	Global[a]		Tropical[b]		Midlatitude[c]			
Pressure, hPa	ppmv	%	ppmv	%	ppmv	%	ppmv	%
0.046	+0.01	+2	−0.03	−5	−0.05	−7	−0.05	−7
0.1	+0.04	+4	+0.03	+3	+0.05	+5	+0.05	+6
0.22	+0.2	+18	+0.2	+18	+0.2	+20	+0.1	+9
0.46	+0.2	+9	+0.1	+8	+0.15	+8	+0.04	+2
1.0	+0.4	+12	+0.4	+14	+0.4	+12	+0.2	+6
2.2	+0.5	+9	+0.5	+9	+0.5	+9	−0.07	−1
4.6	0.0	0	0.0	0	−0.1	−1	−0.3	−4
10	+0.5	+6	+0.5	+5	+0.5	+7	+0.5	+6
22	−0.5	−8	−0.4	−6	−0.6	−10	−0.4	−7
46	+0.5	+23	+0.8	+97	+0.4	+15	+0.6	+31

[a]Based on ∼400,000 profiles from all latitudes for the first full year of data (Oct. 91 through Sep. 92).
[b]Based on ∼60,000 profiles from 10°S to 10°N for the first full year of data.
[c]Based on ∼25,000 profiles from 35° to 45°N and 35° to 45°S for the first full year of data.

[65] 1. A small positive MLS offset, of order 2 to 4% on average, is observed in average comparisons of v5 O3_205 MLS data versus ozonesonde profiles in the midstratosphere and SAGE II values in the midstratosphere to upper stratosphere. This is within the accuracies we expect from the data sets, although not in accord with a similarly small, but negative, offset between MLS and Table Mountain Facility lidar data in the midstratosphere to upper stratosphere. At pressures near 68 hPa, the MLS values are ∼10 to 15% larger than ozonesonde data, although the magnitude of the offset in this region is less than 5 to 10% if one compares MLS v5 with SAGE II V6.1 data (during 1995–1996).

[66] 2. The uncertainties in the O3_205 Level 3A files overestimate the actual precision of the measurements. Uncertainties in the MLS data files should be multiplied by a factor of 0.6 to 0.9, depending on altitude (see Table 6 and section 6.1).

10.6. Caveats in Use of v5 O3_205

[67] Caveats in the use of v5 O3_205 are as follows: (1) See the general caveats detailed in section 5. (2) See the known artifacts described in the previous subsection. (3) The profiles in the Level 3A files extend from 464 hPa to 0.00046 hPa; however, only values from 100 hPa to 0.22 hPa are considered sufficiently reliable for general use in scientific studies using individual profiles. Averaging (e.g., zonal mean) can be used to obtain information for pressures lower than 0.22 hPa.

11. Ozone From 183-GHz Radiometer Data

[68] Information on data quality and characteristics of previous versions of O3_183 is given by *Froidevaux et al.* [1996] and *Ricaud et al.* [1996]. The data versions are described in the MLS "Data Quality Documents" available on the MLS web site. Here, we briefly summarize the changes that occurred for the v5 O3_183 data, and give our estimates of v5 precision and accuracy. O3_183 remains the recommended MLS data set for mesospheric ozone, but O3_205 is still recommended for the stratosphere. *Pumphrey and Harwood* [1997] have shown that the raw 183-GHz ozone radiances contain useful information up to about 90 km (roughly 0.002 hPa), but mixing ratio retrievals are limited by uncertainties in tangent pressure, temperature, and vertical resolution (which is of order 10 km at upper mesospheric heights). Retrievals of ozone are discussed here for the vertical range up to 0.01 hPa, with no attempt at special studies for higher altitudes, where the v5 retrievals show increasing (and larger than 50%) a priori contribution.

11.1. Changes in Algorithms for v5 183-GHz Ozone

[69] The main changes in v5 O3_183 are the use of a finer retrieval grid (see Introduction) below 0.1 hPa and the use of an iterative retrieval for this band, as discussed in section S4. The retrieval grid change also leads to somewhat poorer estimated precision, except in the lower stratosphere, where the use of more radiances than in v4 and the improvements in tangent pressure precision outweigh this effect. Also, new values were deduced for the sideband ratios and ozone spectral parameters for this band [*Pumphrey and Bühler*, 2000].

11.2. Comparison of Different Data Versions for 183-GHz Ozone

[70] Table 7 shows average differences between the three data versions for MLS O3_183. V5 O3_183 data exhibit an overall increase from v4 of about 5 to 10% (and occasionally 20%) in the upper stratosphere and lower mesosphere. V5 values also show a small (5 to 10%) decrease from v4 values at 22 hPa, but a more significant increase at 46 hPa (especially in the tropics); v5 values in the polar stratosphere below 10 hPa are generally slightly smaller than the v4 values (these differences are not shown in the table).

[71] Section S11 shows the O3_183 northern midlatitude mesospheric ozone diurnal cycle discussed for MLS v3 data by *Ricaud et al.* [1996]. Changes from v4 to v5 are larger for pressures greater than 0.1 hPa, mainly because of the finer v5 retrieval grid. The conclusions of the *Ricaud et al.* [1996] study have not been affected. The amplitude of the diurnal cycle has not changed significantly, and the pressures at which the models shown by *Ricaud et al.* [1996] were in poorer agreement with the MLS data are the same (namely, 0.22 and 0.1 hPa, where the models predict a significantly larger day-to-night increase than is measured).

11.3. Validation of v5 183-GHz Ozone

[72] Our comparisons of zonal mean differences show that the v5 O3_183 values between 0.46 and 46 hPa are larger than the O3_205 values by about 2 to 5%, within the combined estimated accuracies. Comparisons with SAGE II data and MLS O3_205 profiles are shown in Figure 6,

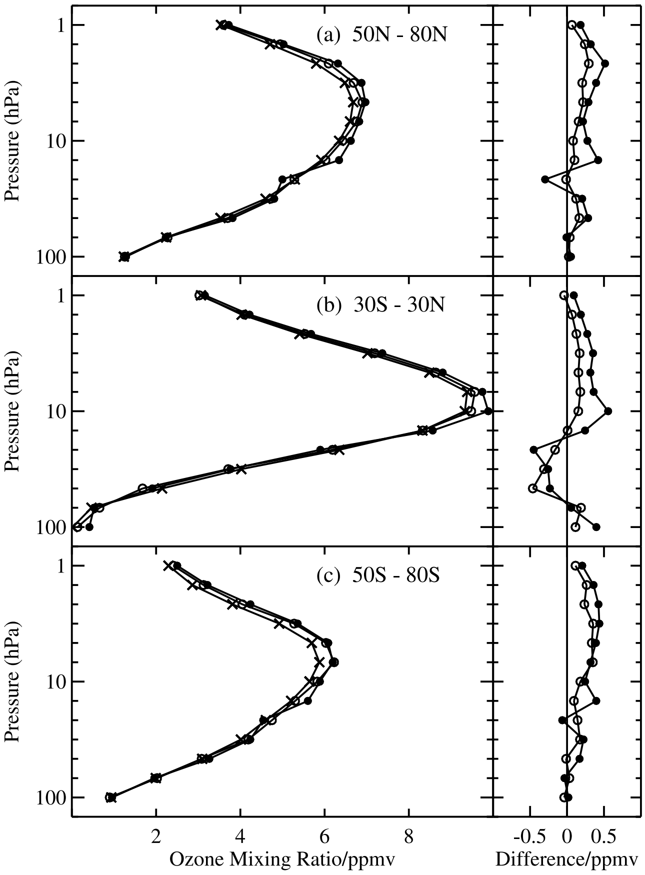

Figure 6. Left panels show, for different latitudes, a comparison of zonally-averaged SAGE II ozone (crosses) with MLS O3_183 (dots) and O3_205 (open circles), for all available coincident MLS and SAGE II profiles from January through March 1993. Right panels give differences (MLS − SAGE II). The averages are based on roughly 200 to 275 profiles at high latitudes and over 550 profiles at low latitudes. Some artifacts in v5 O3_183 profiles, namely the systematically large tropical values at 100 hPa, and the notch at 22 hPa at higher latitudes, are seen here.

where the MLS O3_183 zonal average profiles (coincident with SAGE II profiles, using the same criteria as for the O3_205 validation) in three broad latitude bins exhibit higher values than both the MLS O3_205 and the SAGE II profiles. Since we have shown in section 10.3 that v5 O3_205 values are on average a few percent larger than several other accurate data sets, O3_183 values are therefore a few percent larger yet. While the two MLS ozone retrievals exhibit similar difference patterns from SAGE II, there are some suspicious O3_183 features: abundances at 100 hPa are overestimated at low latitudes (see Figure 6b), and notches in the profiles appear at higher latitudes (see Figures 6a and 6c) at 22 hPa. Similar artifacts are observed in comparisons (not shown here) of average O3_183 profiles with coincident ozonesonde profiles. In addition, zonal mean O3_183 values are sometimes negative at 68 hPa.

[73] For reasons discussed above, we do not recommend the use of O3_183 data in the lower stratosphere (100 or 68 hPa). The quality of O3_183 data is generally somewhat poorer than that of O3_205 in the stratosphere. Likely reasons for the poorer O3_183 data quality include our inability to use one excessively noisy wing channel in this band and poorer calibration of sideband ratios for the 183-GHz radiometer. Based on upper stratospheric comparisons of O3_183 with O3_205 and SAGE II profiles, we believe that the main issue for MLS mesospheric O3_183 is a ~5% positive bias.

11.4. Vertical Resolution, Precision, and Accuracy of v5 183-GHz Ozone

[74] Table 8 gives the v5 O3_183 estimated vertical resolution, precision and accuracy, obtained in the same manner as for O3_205. Based on our midstratospheric to upper stratospheric comparisons for O3_183, and assumptions of continuity into the mesosphere, we estimate a conservative absolute accuracy of 10% for O3_183 in most of the stratosphere and mesosphere. Averaged values of O3_183 can be used at pressures lower than 0.05 hPa (the top pressure in the table shown here), probably up to 0.01 hPa or somewhat higher, but we have not evaluated the data quality at those heights. We recommend not using the O3_183 data for pressures larger than 46 hPa, since its artifacts and accuracy are worse in this region, particularly in the tropics.

11.5. Known Artifacts and Systematic Effects in v5 183-GHz Ozone

[75] Known artifacts and systematic effects in v5 183-GHz ozone are as follows: (1) MLS O3_183 has a positive bias, averaging about 5 to 10%, based on comparisons with the v5 O3_205 data, as well as ozonesonde profiles and SAGE II values in the midstratosphere to upper strato-

Table 8. Estimated Vertical Resolution, Precision, and Accuracy of v5 O3_183

Pressure hPa	Vertical Resolution[a] km	Estimated Precision ppmv	Estimated Precision %	Precision Ratio[b]	Estimated Accuracy[c]
0.046	6	0.2	[d]	0.5	max. of 0.1 ppmv or 10%
0.068	6	0.15	[d]	0.4	max. of 0.1 ppmv or 10%
0.1	6	0.15	[d]	0.4	max. of 0.1 ppmv or 10%
0.15	8	0.15	[d]	0.3	max. of 0.1 ppmv or 10%
0.22	5	0.15	10	0.3	10%
0.32	7	0.15	10	0.3	10%
0.46	3.5	0.15	8	0.4	10%
0.68	3.5	0.2	8	0.7	10%
1.0	4	0.2	6	0.7	10%
1.5	3.5	0.2	5	0.7	10%
2.2	3.5	0.25	4	0.9	10%
3.2	3.5	0.25	4	0.8	10%
4.6	3	0.3	4	1.0	10%
6.8	3	0.3	3	1.0	10%
10	3	0.3	3	0.9	10%
15	3	0.3	4	1.0	10%
22	3	0.3	5	1.0	10%
32	3	0.25	6	1.0	15%
46	3.5	0.2	8	0.9	20%

[a]As defined in section 6.2.

[b]Data file uncertainties should be multiplied by these numbers to obtain a better value for the "1σ" single profile precision (see text).

[c]Accuracies quoted here represent roughly a 95% confidence level ("2σ" values).

[d]At pressures lower than about 0.2 hPa, day/night differences in ozone become significant enough that absolute (ppmv) precision becomes the most convenient quantity to use.

Table 9. Vertical Resolution, Precision, Known Bias, and Accuracy for V5 ClO[a]

Pressure, hPa	Vertical Resolution,[b] km	Typical Precision, ppbv	Precision Ratio,[c]	Known Bias, ppbv	Estimated Accuracy After Subtracting Known Bias[d]
1.0	8	0.5	0.7		0.1 ppbv + 15%
1.5	7	0.5	0.7		0.1 ppbv + 15%
2.2	6	0.5	0.7		0.1 ppbv + 15%
3.2	5	0.4	0.7		0.1 ppbv + 15%
4.6	5	0.4	0.8		0.1 (0.15) ppbv + 15%
6.8	5	0.4	0.8	−0.02	0.05 (0.15) ppbv + 15%
10	4	0.4	0.8	0.01	0.05 (0.15) ppbv + 15%
15	4	0.4	0.8	0.03	0.05 (0.15) ppbv + 15%
22	4	0.3	0.8	0.05	0.05 (0.15) ppbv + 15%
32	4	0.3	0.7	0.08	0.05 (0.15) ppbv + 15%
46	4	0.3	0.7	0.08	0.05 (0.15) ppbv + 15%
68	5	0.3	0.6	0.07	0.05 (0.15) ppbv + 15%
100	5	0.6	0.8	0.01	0.2 ppbv + 15%

[a]See text for further explanation.
[b]As defined in section 6.2.
[c]Data file uncertainties should be multiplied by these numbers to obtain a better value for the "1σ" single profile precision (see text).
[d]Accuracies quoted here represent roughly a 95% confidence level ("2σ" values). Values in parentheses apply to polar winter vortex data.

sphere. (2) Values are too large at 100 hPa at low latitudes and negative averages occur at 68 hPa. (3) There are some pervasive notches in the profiles at 22 hPa, at high latitudes in particular. (4) The uncertainties in the O3_183 Level 3A files overestimate the actual precision of the measurements. For best estimates of precision, uncertainties in the data files should be multiplied by 0.3 to 1.0, depending on altitude (see Table 8 and section 6.1).

11.6. Caveats in Use of v5 183-GHz Ozone

[76] Caveats in the use of v5 183-GHz ozone are as follows: (1) See the general caveats listed in section 5. (2) See the known artifacts described in the previous subsection. (3) The profiles contained in the Level 3A files extend from 464 hPa to 0.00046 hPa; however, only values from 46 hPa to 0.046 hPa are considered sufficiently reliable for general use in scientific studies (although some information exists in average values at lower pressures).

12. Stratospheric and Mesospheric Water Vapor

[77] Version 3 of the MLS stratospheric H_2O product is described and validated by *Lahoz et al.* [1996]. Since that time, version 4 has been released, followed by a development prototype known as version 104 (v104). The latter was a retrieval of stratospheric water vapor only and was produced to demonstrate the possibility of retrieving MLS data on a grid with 6 levels per pressure decade. It rapidly became clear that v104 was a much better data set than v4, and it has gone on to be used in a number of scientific studies. The validation of v4 and v104 water vapor is described by *Pumphrey* [1999]. Generally, we recommend that the v104 H_2O data set be used in preference to v5. The v104 data are available from the GSFC DAAC, archived as "version 6" MLS H_2O data. Details on this recommendation and the v5 H_2O data set in general are given in section S12.

13. Chlorine Monoxide (ClO)

[78] *Waters et al.* [1996], describing validation of MLS v3 ClO data, provides background for the material in this section and a general reference for the MLS ClO measurements. Major changes from v3 to v4 ClO were: (1) correction of the "old" line strength that was inadvertently used in v3 processing [*Waters et al.*, 1996], with the expected 8% lowering of ClO values from v3, and (2) retrieval of HNO_3, which can reduce the retrieved values of enhanced lower stratospheric ClO (in the polar winter vortices) by ~0.2 ppbv. More information on the v4 ClO data is in the MLS v4 Data "Quality Document" available on the MLS web site. Changes between v3, v4 and v5 ClO are within the uncertainties of comparisons with other measurements, and the emphasis here is on describing changes between these versions. V5 is the ClO data version recommended for scientific studies.

13.1. Changes in Algorithms for v5 ClO

[79] The major changes for ClO in v5 are because of (1) retrievals on each UARS surface, and (2) retrieving CH_3CN instead of SO_2. Although v5 retrievals are done on each UARS surface (between 100 and 0.46 hPa), the vertical resolution of v5 ClO (see Table 9 later in this section) is approximately the same as for v4 and v3. The additional free parameters in v5 allow better definition of the profile, and the v5 profiles are generally smoother due to off-diagonal terms in the a priori ClO covariance matrix that favor smoother profiles (see section S3.2). The CH_3CN retrievals in v5 allow a better fit of the measured radiances in MLS bands 2 and 3 when there is negligible volcanically-injected SO_2 in the stratosphere, including a fit of some residual curvature in the spectra that previously led to unrealistic negative values in averaged nighttime ClO between ~22 and ~4.6 hPa.

[80] The v5 data at 100 hPa are more stable and have more realistic values than in previous versions. We believe the v5 ClO data at 100 hPa are acceptable for use in scientific studies but, as with all MLS data, their uncertainties must be appreciated. ClO data in the files at pressures greater than 100 hPa should never be used. Data at pressures less than 1 hPa are not necessarily reliable (because of small residual artifacts in the measured radiance

(see Figure S20 of *Waters et al.* [1996]) although averages of these data exhibit the expected diurnal behavior (more ClO at night).

13.2. Comparison of v5, v4, and v3 ClO

[81] We compare v5, v4 and v3 ClO for three categories of observations: (1) low latitudes and midlatitudes, and high latitude summer, where there is no "enhanced" lower stratospheric ClO that could be caused by winter polar processes, (2) Antarctic and (3) Arctic vortex regions with enhanced lower stratospheric ClO. Data used in all comparisons were selected by QUALITY_ClO = "4," MMAF_STAT = "G," "T" or "t," and positive uncertainties in the data files (see section 5). All v3 data values have been multiplied by 0.92 to correct the known line-strength error in v3 data.

13.2.1. Low-Latitude to Midlatitude Annual, and High-Latitude Summer

[82] Figure 7 compares averages of measurements made between 45°S and 45°N over an annual cycle, and "summer" measurements made poleward of 45°. The major change in v5 is a 0.1–0.2 ppbv increase over v4 and v3 values between ~46 and ~4.6 hPa, due to retrieval of CH_3CN. This change removes the negative values that are present in v3 and v4 average nighttime data at these altitudes. However, the v5 night values of ~0.1 ppbv at 68 to 22 hPa are unrealistically large, and, as for v3 and v4, day/night differences are needed for confidence of better than ~0.2 ppbv in absolute values. Day/night differences for all the versions agree to within 0.03 ppbv for the 45°S–45°N average at all altitudes, and to within the approximate precision of the averages for high latitude "summer." More ClO is present during night than day above 1 hPa in all versions, as theoretically predicted [e.g., *Ricaud et al.*, 2000] because of decreased nighttime ClO loss by ClO + O.

13.2.2. Antarctic Vortex

[83] Figure 8 compares averages of measurements made in the Antarctic 1992 winter vortex where lower stratospheric ClO was enhanced. The mid-August 1992 v5 Antarctic daytime peak value of 2.3 ppbv at 22 hPa agrees to within 0.04 ppbv with that of v4, both of which are 0.3 ppbv less than v3. V5 has 0.4 ppbv more ClO at 100 hPa than v4 or v3. Other mid-August 1992 daytime changes are generally <0.2 ppbv and within the noise of the averages; v5 and v4 night values in the lower stratosphere are ~0.2 ppbv less than v3. The altitude of the daytime profile minimum, separating upper and lower stratospheric ClO, is lower in v5 (at 6.8 hPa) than in v4 (at 4.6 hPa) but higher than in v3 (at 10 hPa). The night values in v5 are unrealistically negative by ~0.15 ppbv at 4.6 and 6.8 hPa, above the expected noise of ~0.04 ppbv in the average. V5 is, however, an improvement in this regard over v4, which is negative by 0.33 ppbv at 4.6 hPa, and v3, which is negative by 0.21 ppbv.

[84] The mid-September 1992 Antarctic v5 profile has significantly more daytime ClO at 100 hPa (0.84 ppbv) than does v4 (−0.04 ppbv) or v3 (−0.11 ppbv). V5 has, correspondingly, less daytime ClO at 46 hPa (1.47 ppbv) than v4 (2.00 ppbv) or v3 (2.18 ppbv). The altitude of the profile minimum is lower in v5 (at 15 hPa) than in v4 or v3 (at 10 hPa), and has lowered since mid-August in all versions. The altitude of the enhanced lower stratospheric ClO peak moves downward with time after mid-August in all versions, as has been reported earlier for MLS v3 data [*Waters et al.*, 1996] and seen in ground-based microwave observations [*de Zafra et al.*, 1995; *Solomon et al.*, 2000]. Night average values from MLS are negative at 10 and 15 hPa, but only by their noise level of 0.04 ppbv. As can be seen by comparing the two columns in Figure 8, the enhanced ClO changes from previous versions can depend upon the specific ensemble of data being examined: there is substantially less change in the 15–18 August 1992 data than in the 17–19 September 1992 data.

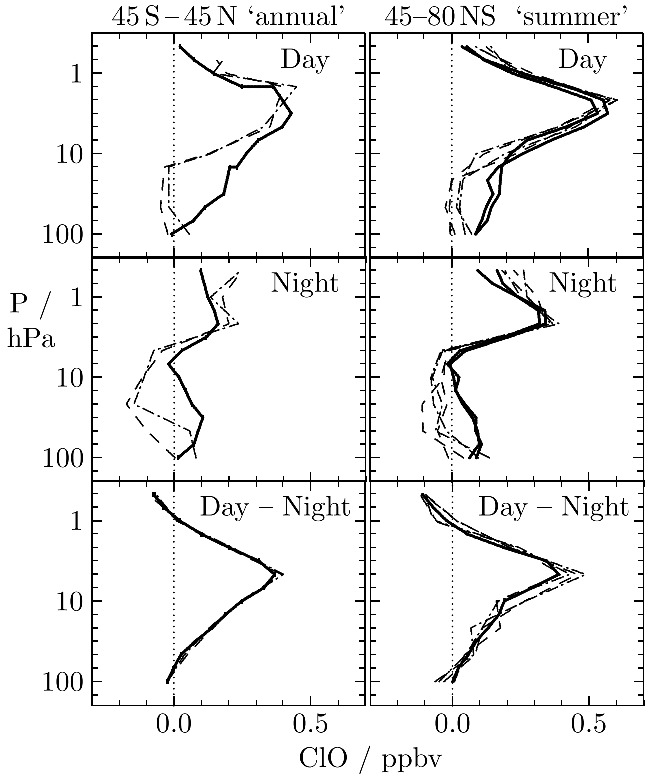

Figure 7. Averages of measurements made between 45°S and 45°N (left panels) and poleward of 45° in "summer" (right panels). Solid thick lines are v5 data, dash-dot-dash are v4 and dashed v3. "Day" averages are for local solar zenith angles (sza) <90° "Night" are for sza >90° and local solar times between midnight and 6 a.m. The 45°S–45°N measurements were made between 21 September 1991 and 20 September 1992, and are averages of ~80,000 individual profiles for day and ~90,000 for night; predicted 1σ precisions for these averages are better than 0.003 ppbv at all altitudes. The 45°–90° measurements were made between 2 May and 28 October 1992 in the north and between 4 November 1991 and 30 April 1992 in the south (averages of ~25,000 day profiles and ~4000 night profiles each for north and south); predicted precisions for these averages are better than ~0.02 ppbv at all altitudes. Two curves for each linestyle in the right panels show separate averages for north and south. Ticks on the vertical axes are at breakpoints of the piecewise-linear representation of the profile.

13.2.3. Arctic Vortex

[85] Figure 9 compares averages of measurements made in the Arctic winter vortex in January 1992 and 1996 where

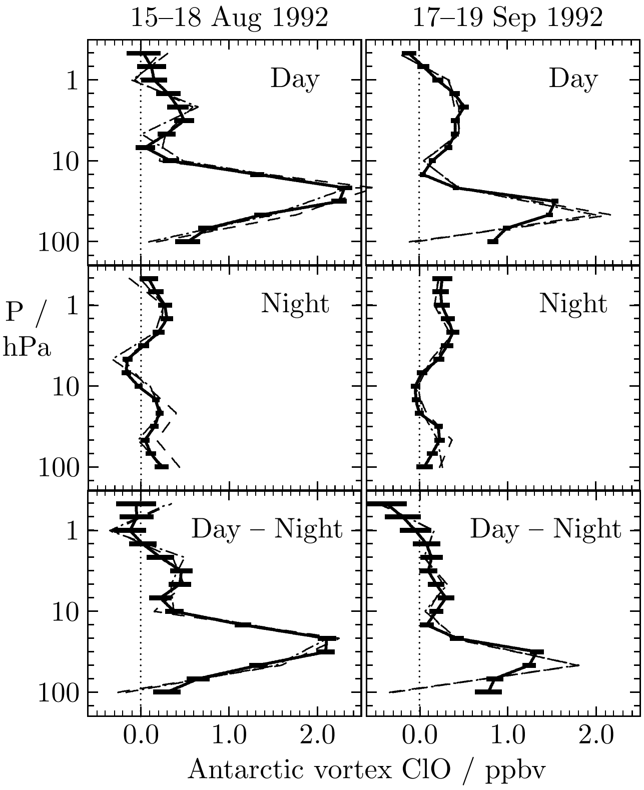

Figure 8. Average of ClO retrievals from measurements made in the 1992 Antarctic winter vortex at locations of greatest ClO enhancement in the lower stratosphere. Solid thick lines are v5 data with horizontal bars indicating the ±1σ predicted precision of the averages; dash-dot-dash are v4 and dashed are v3 (in places these merge). The 15–18 August measurements (left panels) were made at 70°–80°S and 120°W–90°E: "Day" is for sza <87° and the average of 25–26 (depending upon data version) individual profiles; "Night" is for sza >100° and the average of 95–96 profiles. The 17–19 September 1992, measurements (right panels) were made at 75°–80°S and all longitudes: "Day" is for sza <90° and the average of 151–155 profiles; "Night" is for sza >95° and the average of 75 profiles. (The reason for the different solar zenith angles here, and in Figure 9, for distinguishing "night" and "day" is the number of measurements that were available at different zenith angles.)

lower stratospheric ClO was enhanced [*Waters et al.*, 1993; *Santee et al.*, 1996]. Day ClO mixing ratios at the profile peak agree to better than 0.1 ppbv for all versions, but the altitude of the peak is higher in v5 (at 32 hPa) than in v4 and v3 (at 46 hPa). The January 1996 v4 and v3 100 hPa unrealistic large negative values (∼−1 ppbv, representative of the individual profiles that went into the average and not due to a single very bad profile) are not present in v5, which has ∼0.5 ppbv daytime ClO at 100 hPa for both years. Average nighttime ClO values for both years agree among all versions to within the noise, except in January 1996 at 10 hPa, where v4 and v3 are more unrealistically negative (−0.2 ppbv) than v5 (−0.05 ppbv). Although the changes in enhanced ClO from previous versions are more similar for the two Arctic examples shown in Figure 9 than for the Antarctic examples shown in Figure 8, these may not be representative of all situations for the Arctic. The specific data ensemble under consideration must be examined to determine the changes for that ensemble.

13.3. ClO Data After 15 June 1997

[86] Daily zonal means (from data taken before the 63-GHz radiometer was turned off) show that the "205-GHz only" values differ from the standard v5 values by ∼0.05 ppbv or less at all vertical levels except 100 hPa, and except in situations of enhanced ClO in the polar winter vortices. At 100 hPa, and for polar enhanced ClO (at all levels), the difference can be up to ∼0.2 ppbv. This offset is not necessarily removed by day/night differences. Thus, the ClO data after 15 June 1997 (when only the 205-GHz radiometer was operated) are expected to have biases (positive and negative) relative to earlier data of up to ∼0.2 pbbv at 100 hPa and in polar enhanced situations, and up to ∼0.05 ppbv elsewhere.

13.4. Estimated Vertical Resolution, Precision, and Accuracy of v5 ClO

[87] Table 9 gives the v5 ClO vertical resolution, typical single profile precision, known bias, and estimated accuracy. The typical single profile precisions in Table 9 are 1σ values, based on the minimum monthly rms variability in

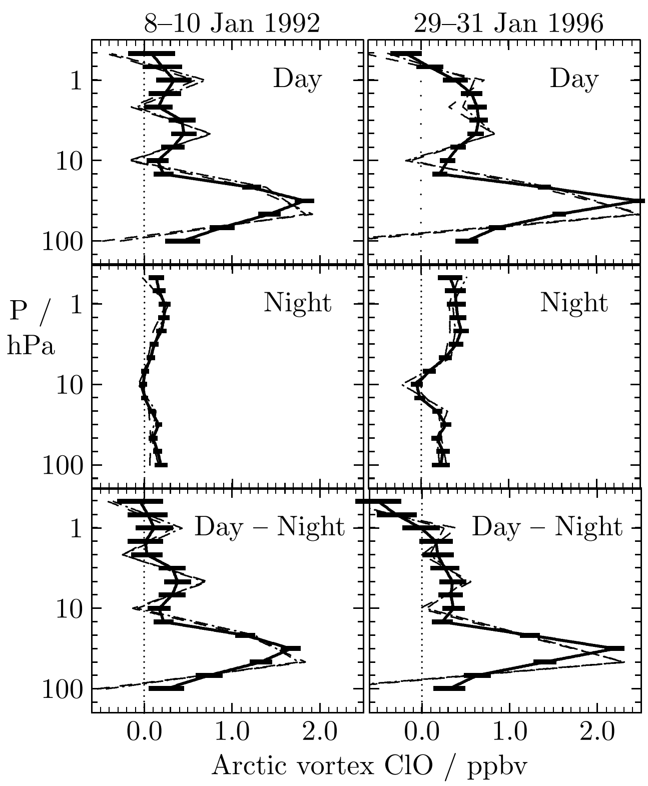

Figure 9. As in Figure 8 but for Arctic January measurements. The 8–10 January 1992 measurements (left panels) are from 60°–80°N and 30°W–60°E: "Day" is for sza <87° and the average of 12–14 individual profiles; "Night" is for sza >100°, and the average of 116–118 profiles. The 29–31 January 1996 measurements (right panels) are from 60°–80°N and 45°–105°E: "Day" is for sza <90°, and the average of 28–30 profiles; "Night" is for sza >110°, and the average of 51–52 profiles.

individual night retrievals from measurements equatorward of 45° for the first full year of measurements. The observed ClO variability under these conditions is dominated by instrument noise and is a good indicator of the precision for individual profiles. The uncertainties given in the v5 ClO data files overestimate the actual precision (i.e., are conservative), as mentioned in section 6.1, and should be multiplied by the "ratio" values in the fourth column of Table 9 to obtain a better value for the precision. ClO precision can be improved by averaging individual profiles: the precision for an average of N profiles is $\sqrt{N}$ better than the precision for an individual profile. Precision of the retrieved v5 ClO values has been improved over previous versions, especially at the highest and lowest altitudes, as seen both in the observed standard deviation of the values and in the estimated precision given in the data files. This is mainly due to the improved estimates of tangent pressure obtained by v5.

[88] Values in the "known bias" column of Table 9 were determined as described in the supplementary material. They are from the thick line in Figure S23 (in section S13) for the first ~3 years of the mission. These differ negligibly from values for the subsequent period up to 15 June 1997, and thus they apply to the majority of MLS data. Slightly better bias values for data taken after 15 June 1997, when the 63-GHz radiometer was turned off, are given by the thin line in Figure S23.

[89] The "estimated accuracy" column of Table 9 gives the "bias" (i.e., additive) uncertainty in ppbv after subtracting the "known bias" and the "scaling" (i.e., multiplicative) uncertainty in percent. Values given in the table represent 90–95% confidence levels (roughly 2σ). Values for the bias uncertainties at 6.8 hPa and higher pressures are based on the scatter of the clustered points at each level in Figure S22. The bias uncertainty is increased to 0.15 ppbv for winter polar vortex conditions because, as shown in Figure 8, unrealistic negative values of 0.15 ppbv at 4.6 and 6.8 hPa were retrieved in the Antarctic winter vortex for which we do not have an explanation. The winter polar vortex bias uncertainty of 0.15 ppbv may, however, be overly conservative (too large) at lower altitudes, where no negative values above the noise have been observed, and the nighttime positive values appear (at least roughly) consistent with values expected from enhanced ClOOCl thermal decomposition (see section S13). Users of the data should remove biases by taking day/night differences whenever possible. We have ascribed a 0.1 ppbv bias uncertainty to the ClO data at the higher levels because we do not believe that biases at higher altitudes should be larger than at low altitudes; again, this may be conservative because biases are actually expected to be smaller at the higher altitudes.

[90] The overall estimate of accuracy is the root sum square of the bias uncertainty and the scaling uncertainty (the retrieved mixing ratio value times the percentage given in the last column of Table 9). The scaling uncertainty in v5 data, based on the arguments given by *Waters et al.* [1996], is ~15% (at the ~90–95% confidence level) at all surfaces where the data are considered useful. The improved v5 precision causes less contribution of the a priori to the "scaling" uncertainty (see Figure 8 of *Waters et al.* [1996]), which is significant at pressures of 1 hPa and less, and 46 hPa and greater.

[91] The overall uncertainty for a datum is the root sum square of accuracy and precision. Note that precisions given here (and in the Level 3 files) are 1σ values, whereas accuracies are 90–95% confidence (roughly 2σ) values.

13.5. Known Artifacts in v5 ClO

[92] Known artifacts in v5 ClO are as follows:

[93] 1. There are known minor biases in v5 retrieved ClO values in the lower stratosphere. Better estimates of ClO are obtained by subtracting the "known bias" values in Table 9 from the values given in the v5 MLS data files. For data after 15 June 1997, a slightly better correction for the biases is obtained from the thin curve in Figure S23 of the supplementary material.

[94] 2. ClO low-latitude values at ~46–4.6 hPa are artificially high in September and October 1991 (by up to ~0.5 ppbv in September and decaying through October to less than 0.1 ppbv). This is due to residual contamination by Pinatubo SO_2 which is not accounted for in the v5 retrievals. Day/night differences remove this artifact.

[95] 3. A negative bias of ~0.15 ppbv at 6.8 and 4.6 hPa appears in averages of the mid-August 1992 night data for the Antarctic vortex. These negative ClO values do not appear in averages for mid-September 1992 Antarctic data, nor in Arctic vortex data examined to date. The reason for them is not understood.

[96] 4. Nonlinearities with respect to temperature can cause retrieved ClO values to be up to approximately 5–10% too large in the cold winter polar (especially Antarctic) vortex. This effect has not been thoroughly quantified, but we believe that it is covered by the uncertainties in Table 9.

[97] 5. As mentioned earlier, uncertainties given in the ClO v5 data files overestimate the actual precision of the measurements. Uncertainties in the data files should be multiplied by the "ratio" values in the fourth column of Table 9.

13.6. Caveats for Using V5 ClO

[98] Caveats for using v5 ClO are as follows: (1) See the general caveats described in section 5, (2) see the artifacts described in the previous subsection, (3) values in the files for pressures greater than 100 hPa should never be used, and (4) values in the files for pressures smaller than 1 hPa are not necessarily reliable.

14. Nitric Acid

[99] Although measurement of $HNO_3$3 was not initially an MLS objective, a significant HNO_3 feature situated just outside the spectral region used to measure ozone imposes a slope through the 205-GHz band that is used to retrieve profiles of gas-phase HNO_3 in the lower stratosphere. HNO_3 became a standard MLS data product in v4; general information on the v4 HNO_3 quality, resolution, and suitability for various scientific studies (in particular investigations of polar stratospheric clouds) is given by *Santee et al.* [1998] and *Santee et al.* [1999].

[100] After the MLS v5 data set was produced, it was discovered that neglecting emission from HNO_3 ν_9 and ν_7 excited vibrational states caused v5 values to significantly overestimate HNO_3 abundances at some levels in the stratosphere. An empirical correction to the MLS v5

Table 10. Differences Between v6 and v4 HNO_3[a]

Pressure, hPa	Global[b] ppbv	Global[b] %	Tropical[c] ppbv	Tropical[c] %	Midlatitude[d] ppbv	Midlatitude[d] %	Enhanced[e] ppbv	Enhanced[e] %	Polar Depleted[f] ppbv	Polar Depleted[f] %
22	+0.3	+4	+2.7	+406	−1.0	−11	−3.5	−20	−0.5	−10
46	+0.6	+13	+1.6	+347	+0.3	+4	+0.1	+1	+0.8	+75
100	−0.6	−40	−0.8	−185	−0.4	−23	−0.5	−8	−2.1	−83

[a]Differences are v6 − v4; percentages are relative changes from v4.
[b]Based on ~400,000 profiles from all latitudes for the first full year of data.
[c]Based on ~60,000 profiles from 10°S to 10°N for the first full year of data.
[d]Based on ~30,000 profiles from 35°N to 45°N and from 35°S to 45°S for the first full year of data.
[e]Based on ~5,000 profiles from 70°N to 80°N during the period from December 1992 to mid-January 1993.
[f]Based on ~4,500 profiles from 70°S to 80°S during the period from mid-August to mid-September 1992.

HNO_3 data set has been derived and is described in section S14 in sufficient detail to allow its application to the v5 HNO_3 Level 3A files by the user. The empirical correction is a linear, strongly temperature-dependent scaling of the original v5 HNO_3 values. Applying the correction leads to reductions in the reported v5 HNO_3 mixing ratios of about 4–8% at 100 hPa, 10–20% at 32 hPa, and 25–35% at 10 hPa, depending on the latitude and season. For the most part this has mitigated discrepancies with correlative data sets, especially near the profile peak, but it has not eliminated them entirely, and at some levels agreement is markedly poorer. Comparisons with other HNO_3 data sets are discussed in more detail in section 14.3. In the following, the corrected HNO_3 data set is referred to as "v6." These v6 HNO_3 data are available from the GSFC DAAC.

14.1. Changes in Algorithms for v5 HNO_2

[101] More rigorous error propagation as well as improvements in the O3_205 (retrieved in the same band as HNO_3) and tangent point pressure retrievals have led to substantially better (by a factor of 2–3) HNO_3 precision in v5 than in v4, even though the v5 retrievals are performed on every UARS surface. In addition to the strong HNO_3 feature just outside band 4, several weak HNO_3 lines in bands 2 and 3 are now included in the retrievals, providing information at higher altitudes and extending the vertical range for reliable measurements up to 4.6 hPa (from 22 hPa in v4).

[102] Because HNO_3 is retrieved in the 205-GHz ozone band, the relevant quality flag for HNO_3 data is QUALITY_O3_205. In v5 the algorithm for setting this parameter was modified because of changes in the χ^2 statistic describing the fit to the radiances. The χ^2 statistic for this band is now less correlated with anomalies in retrieved HNO_3 than it was in v4, and more profiles are being flagged bad (see section 10.1). Overall, about twice as many profiles (~2%) are discarded in v5 than in v4. Thus in some cases individual profiles that passed the recommended quality control measures in v4 will be screened out using the same procedures with v5 data, even though they do not appear obviously bad. In addition, the HNO_3 data are generally "spikier" in v5 than they were in v4, where "spikes" are identified by comparison of their deviation from monthly zonal means. Some of these spikes pass all of the recommended quality control measures, but they can be identified by inspection and removed on an individual basis.

14.2. Comparison of v6 and v4 HNO_3

[103] Differences between v6 and v4 average profiles are summarized in Table 10. Because v4 retrievals were performed on the even UARS surfaces and were reliable at and below 22 hPa, only the differences on these surfaces are tabulated. Also, because the distribution of HNO_3 in the lower stratosphere exhibits large seasonal and latitudinal variations, separate comparisons are made for different conditions, as noted in the table. In general these differences remain fairly constant through the years of MLS operation, since they have a strong systematic component. The only exceptions are the differences between the v5 and v4 "polar enhanced" average profiles, which display variations at these levels as the peak in the HNO_3 mixing ratio shifts in altitude from year to year.

[104] V6 HNO_3 global average mixing ratios are slightly larger than those in v4, except at 100 hPa, where v6 values are smaller at all latitudes. V6 values are significantly larger in the equatorial regions at 22 and 46 hPa, where strong negative biases (2–3 ppbv over a broad area) in v4 have been eliminated. In contrast, at midlatitudes and high latitudes, especially during early winter when HNO_3 is enhanced inside the vortex, v6 values are substantially smaller at 22 hPa than in v4.

14.3. Comparison of v6 With Other HNO_3 Data Sets

[105] Comparisons of v6 HNO_3 data with both v5 HNO_3 and simultaneous, colocated HNO_3 measurements (version 9) from the UARS Cryogenic Limb Array Etalon Spectrometer (CLAES) [*Kumer et al.*, 1996] are shown in Figure 10 for the 22-hPa level, where the discrepancy between CLAES and MLS v5 is generally largest (as much as ~5 ppbv in the zonal mean when HNO_3 is enhanced at polar latitudes during fall/winter). Similar latitudinal and temporal patterns are seen in both CLAES and MLS HNO_3. In particular, agreement is excellent in the timing and overall morphology of HNO_3 buildup in fall/early winter in both polar vortices and in the development of the collar and denitrified regions in the southern polar vortex. With the correction applied to the MLS v5 data, the disagreement between CLAES and MLS HNO_3 values at 22 hPa is reduced below ~2 ppbv under conditions of wintertime enhancement in the polar vortices. For these conditions, however, MLS v6 zonal-mean values are still larger than those from CLAES by up to ~2.5 ppbv at 32 hPa and ~4.5 ppbv at 68 hPa (not shown). At most other seasons/latitudes, the disagreement between CLAES and MLS v6 HNO_3 is below ~1 ppbv at 22 hPa. The agreement is also within ~1 ppbv (and frequently much better) everywhere at and above 15 hPa, during the summer at all latitudes and altitudes, and throughout the tropics at all altitudes (not shown), except during the first ~100 days of the mission,

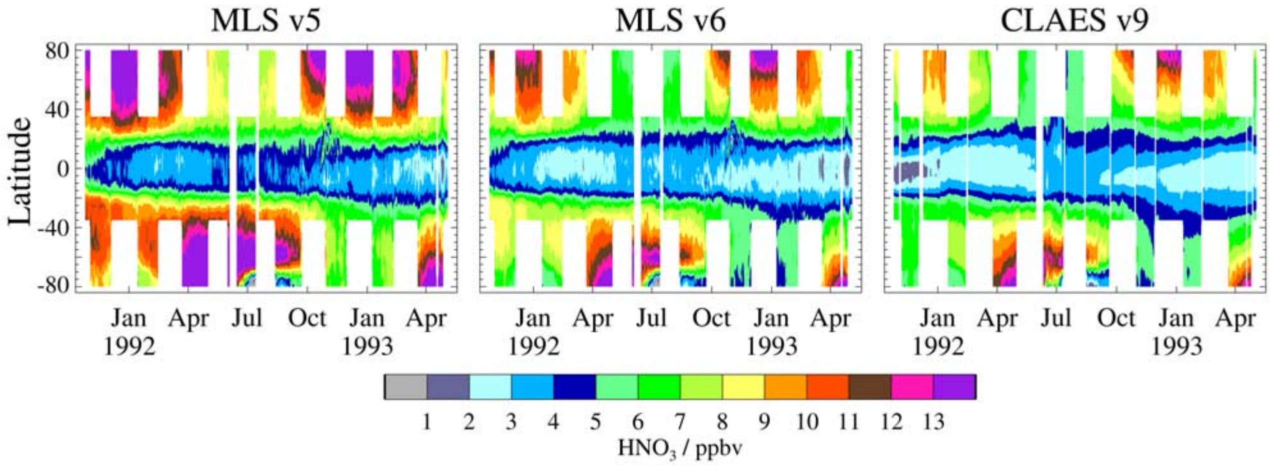

Figure 10. Time series of daily zonal-mean MLS v5, MLS v6, and CLAES version 9 HNO_3 at 22 hPa as a function of latitude for the 18-month lifetime of CLAES. Blank spaces in the plots correspond to periods when data are missing or the instruments were observing the opposite hemisphere.

when enhanced stratospheric SO_2 (which is not retrieved in v5) from the eruption of Mount Pinatubo causes a high bias in MLS HNO_3 of as much as ~2–3 ppbv in the equatorial regions. Although the agreement inside the Antarctic collar region is also within ~1 ppbv at all levels, it is not as good (~1.0–2.5 ppbv) at the lower levels in the region of severe denitrification in the core of the Antarctic winter polar vortex, where CLAES does not record values as low as those from MLS (not shown).

[106] Comparisons with version 3 Atmospheric Trace Molecule Spectroscopy (ATMOS) measurements [*Irion et al.*, 2002] (not shown) also indicate that the overall features of the stratospheric HNO_3 distribution are in good agreement in the two data sets. Grouping the data into broad latitude bins and averaging matched pairs within these bins demonstrates that, in general, the correction has improved (in some cases considerably) the agreement at the lower levels. In the fall northern hemisphere tropics, ATMOS and MLS v6 HNO_3 agree at and below 22 hPa to within 0.5 ppbv. The overall shapes of the profiles are similar in the fall southern hemisphere midlatitudes, but the peak in the MLS profile occurs at a slightly lower altitude, causing differences of about 1–1.5 ppbv at most levels. Vortex and extra-vortex air exhibit different profile shapes during southern hemisphere spring that are captured well in both data sets, although MLS measures ~0.5–1.5 ppbv more HNO_3 inside the vortex between 68 and 32 hPa than does ATMOS. In contrast, however, agreement between the two data sets has worsened above ~22 hPa, where v6 HNO_3 values are systematically smaller than those from ATMOS by 0.5–1.5 ppbv.

[107] MLS v6 HNO_3 data have also been compared to Improved Limb Atmospheric Spectrometer (ILAS) version 5.20 and Ground-based Millimeter-wave Spectrometer (GBMS) HNO_3 measurements using trajectory techniques. *Danilin et al.* [2002] show that applying the correction reduces the discrepancy between MLS and ILAS HNO_3 to ~0.5 ppbv over the range from 450 to 750 K in potential temperature (~19–27 km). Above 750 K, however, the offset between the two data sets increases when the correction is applied to ~1 ppbv, with MLS values lower. Similarly, *Muscari et al.* [2002] find that v6 MLS HNO_3 values are consistently smaller than those from GBMS throughout an annual cycle at high southern latitudes at 740 and 960 K, where differences can exceed 3 ppbv. These results, together with those from the ATMOS comparison, indicate that a significant low bias is present in the MLS v6 HNO_3 data at the topmost levels.

[108] During Antarctic fall, when HNO_3 mixing ratios are generally increasing inside the lower stratospheric polar vortex, GBMS HNO_3 abundances at the South Pole agree well with those obtained by MLS in the 70–80°S latitude band at 465 K but are ~1–3 ppbv larger over the range 520–655 K [*Muscari et al.*, 2002]. Note that this difference is in contrast to that found between CLAES and MLS, as discussed above. *Muscari et al.* [2002] attribute this discrepancy to a combination of the sharp latitudinal gradients in HNO_3 at this time of year and the spatial resolution limits of the trajectory matching technique. Comparisons between MLS v6 and GBMS HNO_3 data [*Muscari et al.*, 2002] also reveal significant differences during Antarctic late winter, when GBMS values drop to near zero throughout the lower stratosphere while MLS values reach a lower limit of ~1–2 ppbv at 520 K and ~3–5 ppbv at 585 and 620 K (and CLAES values are even higher, as mentioned previously). Very low HNO_3 abundances are consistent with expected polar stratospheric cloud formation and sedimentation processes at this time of year. We believe that the higher MLS HNO_3 is likely an artifact arising from the departure of the zonal climatological temperatures used as the linearization points in the MLS forward model (see section S3.7.6) from actual stratospheric temperatures, which are extremely low during Antarctic late winter. Similar high biases due to nonlinearities with respect to temperature in the MLS retrieval system are also seen in the ClO abundances in the Antarctic winter polar vortex (section 13).

14.4. Estimated Vertical Resolution and Precision of v6 HNO_3

[109] Best precision in the HNO_3 retrievals is attained at 68 hPa. The general range of useful sensitivity is given in Table 11. While the v5 HNO_3 precision is, to first order,

Table 11. Estimated Vertical Resolution and Precision of v6 HNO_3

Pressure, hPa	Vertical Resolution,[a] km	Typical Precision, ppbv	Precision Ratio[b]
4.6	10.5	1.5	0.7
6.8	10.0	1.4	0.7
10	9.5	1.3	0.7
15	9.0	1.2	0.7
22	7.5	1.2	0.7
31	6.5	1.1	0.7
46	6.0	1.0	0.7
68	6.0	0.8	0.6
100	4.5	1.3	0.9

[a]As defined in section 6.2.
[b]Data file uncertainties should be multiplied by these numbers to obtain a better value for the "1σ" single profile precision (see text).

independent of latitude and season, the scientific utility of the data (i.e., signal to noise) can vary with HNO_3 abundance. For example, at 100 hPa the single-profile precision greatly exceeds the average HNO_3 mixing ratio in the tropics (where averaging of several profiles is thus necessary to obtain useful data) but not in the winter polar regions, where HNO_3 is enhanced. In most cases some averaging will also be necessary at levels above 10 hPa. The reliability of the data above 4.6 hPa has not been established, and at this time they are not recommended for use in scientific studies.

[110] The typical single-profile precisions given in Table 11 are 1σ values. They were obtained by computing (for the first full year of measurements) the minimum monthly rms variability in the corrected HNO_3 profiles retrieved in a 10° latitude band centered around the equator. In this region meteorological variability should be small relative to the estimated retrieval error; thus the observed variability is expected to be dominated by instrument noise, providing a good indicator of the measurement precision. Essentially similar results are obtained for a 30° latitude band centered around the equator and for the polar regions during summer. Because natural atmospheric variation is not completely negligible, the true precisions may be slightly better than these estimates.

[111] The theoretical precision values provided in the HNO_3 Level 3A files, which were estimated by the retrieval algorithm, account for variations in the uncertainty that might occur from profile to profile for various reasons (e.g., missing channels or tangent point scan positions would increase the uncertainties). Although these theoretical estimates are generally consistent with the empirically-determined values in Table 11, the estimated uncertainties tend to be conservative; i.e., they are larger than the empirical precisions by about 10–40%, depending on altitude, because of the influence of the a priori estimate and its vertical smoothing on the retrieved profile. Therefore, as described in section 6.1, the estimated uncertainties in the Level 3A files should be multiplied by the ratios given in the fourth column of Table 11 to obtain the best estimate of precision. In general, precision can be improved by averaging together individual profiles: the precision of an average of N profiles is $1/\sqrt{N}$ times the precision of an individual profile.

14.5. Known Artifacts in v6 HNO_3

[112] Known artifacts in v6 HNO_3 are as follows: (1) In the equatorial regions, enhanced stratospheric SO_2 (which is not retrieved in v5) from the eruption of Mount Pinatubo causes a high bias of as much as ~2–3 ppbv in MLS HNO_3 for the first ~100 days of the mission. (2) A significant (~1–3 ppbv) low bias is present in MLS v6 HNO_3 above ~740 K (~15 hPa). (3) Nonlinearities with respect to temperature in the MLS retrieval system cause a high bias in v6 HNO_3 during Antarctic late winter of as much as 3–5 ppbv at 585 and 620 K (with a smaller effect at 520 K). (4) The uncertainties in the HNO_3 Level 3A files overestimate the actual precision of the measurements. Uncertainties in the MLS data files should be multiplied by a factor of ~0.7, depending on altitude (see Table 11).

14.6. Caveats in Use of v5 (and v6) HNO_3

[113] Caveats in the use of v5 (and v6) HNO_3 are as follows: (1) See the general caveats given in section 5. The QUALITY_O3_205 flag is the appropriate indicator of HNO_3 data quality. (2) See the artifacts described in the previous subsection. (3) Omission of some HNO_3 excited vibrational state lines from the retrieval system caused v5 HNO_3 values to significantly overestimate abundances at some levels in the stratosphere. The linear scaling correction described in section S14 should be applied to the v5 HNO_3 profiles. Corrected HNO_3 data have been referred to here as "v6." (4) The estimated absolute accuracy of the v6 MLS HNO_3 data has not been quantified in detail, but, based on the limited comparisons described here, we expect these data to be accurate to within ~3 ppbv above ~15 hPa and ~2 ppbv below ~15 hPa, except in the lower stratospheric winter polar vortices, where biases as large as 4–5 ppbv may be present. (5) Only data between 100 hPa and 4.6 hPa are sufficiently reliable for general use in scientific studies.

15. Methyl Cyanide

[114] A discussion of the role of CH_3CN in the stratosphere, and of the MLS CH_3CN data, is given by *Livesey et al.* [2001]. The MLS CH_3CN data are scientifically useful between 68 and 1 hPa. Although CH_3CN abundance at 100 hPa is retrieved, it is believed that spectral features from $H_2{}^{18}O$ contaminate the CH_3CN signal,leading to an unpredictable bias in the CH_3CN at 100 hPa.CH_3CN data at lesser pressures are not significantly affected by $H_2{}^{18}O$ signals. The data are not reliable for pressures less than 1 hPa, because the spectral contrast in the radiance observations is approaching the accuracy limit of the instrument.

[115] Individual profiles of MLS CH_3CN data have a precision of 40–60 pptv, which is comparable to the typical stratospheric CH_3CN abundances. For scientific study, therefore, some form of averaging is generally required. For example, a monthly zonal mean data set with a 10° latitudinal resolution will have a precision of 1 pptv at 10 hPa.

[116] Occasionally, strong enhancements are seen in the MLS CH_3CN data set in the lower stratosphere. The most notable of these is an enhancement in August 1992 off the coast of Florida, with mixing ratios as high as 10^3 pptv observed. A detailed study has concluded that they represent true enhancements in lower stratospheric CH_3CN, not instrumental artifacts. The August 1992 event has been linked to a forest fire in Idaho (north of the 34°N limit of MLS observations at that time) some days earlier (N. J. Livesey et al., manuscript in preparation, 2003); the causes

Table 12. Estimated Precision, Vertical Resolution, and Accuracy for v5 MLS CH_3CN Data[a]

Pressure, hPa	Vertical Resolution,[b] km	Typical Precision, pptv	Precision Ratio[c]	Estimated Accuracy[d]
1.0	8	90	0.9	10 pptv and 20%
1.5	8	60	0.7	10 pptv and 20%
2.2	7	60	0.8	10 pptv and 20%
3.2	4	50	0.7	10 pptv and 20%
4.6	6	50	0.7	10 pptv and 20%
6.8	5	40	0.7	10 pptv and 20%
10	4	40	0.7	10 pptv and 20%
15	4	30	0.7	10 pptv and 20%
22	4	30	0.7	10 pptv and 20%
32	4	30	0.7	10 pptv and 20%
46	4	30	0.7	10 pptv and 20%
68	4	30	0.7	10 pptv and 20%

[a]See text for details.
[b]As defined in section 6.2.
[c]Data file uncertainties should be multiplied by these numbers to obtain a better value for the "1σ" single profile precision (see text).
[d]Accuracies quoted here represent roughly a 95% confidence level ("2σ" values).

of the few similar events in the data set are under investigation.

15.1. Estimated Vertical Resolution, Precision, and Accuracy in v5 CH_3CN

[117] Table 12 summarizes the precision, vertical resolution and accuracy of the MLS CH_3CN data set. The precision quoted is the minimum rms variability seen in any of the first ten full UARS months of the MLS mission (October 1991 to September 1992), in the latitude band from 5°S to 5°N. As described in section 6.1, the "best estimate" of the true precision for individual profiles can be obtained by scaling the uncertainty quoted in the data files by the ratio column in Table 12.

[118] The accuracy is defined in terms of possible bias and scaling terms. These estimates were obtained by analogy with ClO (see section 13), accounting for the relative line strengths of CH_3CN and ClO and assuming 10% uncertainty in the CH_3CN pressure-broadened linewidth parameter. The overall estimate of accuracy is the root sum square of the bias uncertainty and the scaling uncertainty (the product of the retrieved mixing ratio value with the percentage given here). The lack of correlative CH_3CN data during the MLS mission [*Livesey et al.*, 2001] limits our ability to assess accuracy by comparison with other observations. The overall uncertainty for a datum is the root sum square of the accuracy and the precision.

15.2. Known Artifacts and Systematic Effects in v5 CH_3CN

[119] Known artifacts and systematic effects in v5 CH_3CN are as follows: (1) Data at 100 hPa are contaminated by emission from $H_2{}^{18}O$ and should not be used. (2) Data at pressures less than 1 hPa are unreliable because of instrumental limitations. (3) The spectral signature of CH_3CN in the MLS passband is very similar to that of SO_2. As SO_2 is not retrieved in v5, the retrieval algorithms will interpret any enhancement in SO_2 as an enhancement in CH_3CN. The high SO_2 resulting from the Pinatubo eruption leads to an unquantified high bias in the pre-1992 CH_3CN data, which should not be used. A shorter-lived, localized bias resulting from the eruption of Mount Lascar in Chile is seen in data from 22–24 April 1993.

15.3. Caveats in Use of v5 CH_3CN

[120] Caveats in the use of v5 CH_3CN are as follows: (1) See the general caveats given in section 5. The QUALITY_ClO field is the appropriate indicator of CH_3CN data quality. (2) See the artifacts discussed in the previous subsection. (3) Data should only be used between 68 hPa and 1 hPa, and after January 1992.

16. Summary and Conclusions

[121] We have shown that the MLS v5 algorithms produce data that are generally of higher quality than earlier versions. Halving the spacing of the vertical reporting grid throughout the stratosphere and lower mesosphere, while slightly worsening the precision of the individual data points in many cases, has given better definition of features such as enhancements in ClO. Improvements in the precision of retrieved tangent pressure (particularly in the lower stratosphere) have in some cases ameliorated precision loss due to increased resolution. The accuracy of many species in the lower stratosphere has been improved, through the use of radiances from lower tangent heights than were previously considered. Comparisons with correlative data generally show improvements. New products for v5 are geopotential height and methyl cyanide (CH_3CN).

[122] For some species, there exist other versions of MLS data that are considered preferable to v5. For upper tropospheric humidity, the v490 data [*Read et al.*, 2001] are of better quality than v5, though no v490 data are available after June 1997. For stratospheric and mesospheric water vapor, the prototype v104 data set [*Pumphrey*, 1999] is felt to be of superior overall quality to v5 and is available from the GSFC DAAC as "version 6." The v5 nitric acid data exhibit a bias due to the omission of contributions from excited states. This bias can be corrected as described in section S14. Corrected HNO_3 data are available from the GSFC DAAC as "version 6." SO_2 abundances are not part of the v5 data set, however they are reported in v4.

[123] The v5 algorithms implement comprehensive quality checking, resulting in quality control information for each product. MLS data should only be used in conjunction with this information, and with reference to the other caveats described in this paper.

[124] **Acknowledgments.** We dedicate this paper to the memory of our dear friend and colleague Zvi Shippony, without whose tireless contributions to the MLS forward model algorithms and software this work would not have been possible. The authors gratefully acknowledge contributions to the MLS version 5 algorithms, software, validation and operation from T. A. Lungu, V. S. Perun, E. Fishbein, B. J. Sandor, B. P. Ridenoure, R. P. Thurstans, A. C. Smedley, R. S. Harwood, the staff of the UARS CDHF, and J. Johnson of the GSFC DAAC. We also thank R. R. Lay and D. A. Flower for their support and management of UARS MLS operations, along with the UARS operations staff. We thank the UARS project science office for their consistent support and assistance. We thank R. J. Salawitch for calculations of nighttime ClO abundances as a function of temperature and pressure. We thank E. Cohen for suggesting that we investigate the contribution of the excited vibrational states of nitric acid to the MLS observations. We also acknowledge the assistance and contributions of the various investigators whose correlative data sets were kindly provided for UARS validation studies (e.g., data from sites belonging to the Network for Detection of Stratospheric Change, and the SAGE II data

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