

## The UARS Microwave Limb Sounder version 5 dataset: Theory, characterization and validation

N.J. Livesey, W.G. Read, L. Froidevaux, J.W. Waters, H.C. Pumphrey<sup>1</sup>, D.L. Wu, M.L. Santee, Z. Shippony, R.F. Jarnot

Jet Propulsion Laboratory, California Institute of Technology, Pasadena

**Abstract.** This paper describes the latest and most definitive dataset 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. The version 5 data include mid-stratospheric 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). Improvements over the earlier version 4 dataset are seen in most products. 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.

### 1. Introduction

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 September 12, 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, and 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 Mt. Pinatubo), upper tropospheric humidity, and stratospheric methyl cyanide (also called acetonitrile).

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 dataset 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 ( $\text{CH}_3\text{CN}$ ) and water vapor in the upper troposphere (note that the latter was also produced by

---

<sup>1</sup>The Institute for Meteorology, University of Edinburgh

the ‘Version 4.9’ — v4.9 algorithms). Sulfur dioxide (SO<sub>2</sub>) data, while produced for version 4 (v4), are not produced by the v5 algorithms because of the similarity of the SO<sub>2</sub> spectral signature to that of methyl cyanide.

## 2. The UARS MLS instrument and operations

Details of the MLS instrument are given in *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 Schottky 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  $\pm 255$  MHz away from the line center. The channels vary in width from 2 MHz near the line center to 128 MHz in the wings.

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.

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.

The Appendix 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 through 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.

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 UARS MLS data is the NASA Goddard Space Flight Center’s Distributed Active Archive Center (DAAC).

## 3. Theoretical basis

### 3.1. Optimal estimation

The v5 Level 2 data processing algorithms are based on the standard optimal estimation approach [Rodgers, 1976, 2000]. The primary aim of the algorithms is to retrieve geophysical profiles of temperature and composition from the input radiance dataset. A state vector  $\mathbf{x}$  of length  $n$  is constructed that is a representation of aspects of the state of the atmosphere to which the instrument is sensitive. This is typically vertical profiles of temperature and composition, along with some instrumental and other parameters. Section 3.6 describes the MLS state vector for v5. The radiance measurements are represented by a series of vectors  $\mathbf{y}_i$ , each of length  $m_i$ . The retrieval algorithm chooses a value for  $\mathbf{x}$  which minimizes the quantity  $\chi^2$ , defined as

$$\chi^2 = \sum_i [\mathbf{y}_i - \mathbf{f}_i(\mathbf{x})]^T \mathbf{S}_{y_i}^{-1} [\mathbf{y}_i - \mathbf{f}_i(\mathbf{x})], \quad (1)$$

where  $\mathbf{S}_{y_i}$  is the error covariance matrix for the measurement vector  $\mathbf{y}_i$ , and  $\mathbf{f}_i$  is the corresponding forward model. The forward model describes the radiances that one would expect to observe from MLS, were the atmosphere in the state represented by  $\mathbf{x}$ . The Gauss-Newton solution to this minimization can be shown [Rodgers, 2000] to be given iteratively by

$$\mathbf{x}^{p+1} = \mathbf{x}^p + \left[ \sum_i \mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} \mathbf{K}_i \right]^{-1} \sum_i \mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} [\mathbf{y}_i - \mathbf{f}_i(\mathbf{x}^p)]. \quad (2)$$

$\mathbf{K}_i$  is defined as

$$\mathbf{K}_i = \left. \frac{\partial \mathbf{y}_i}{\partial \mathbf{x}} \right|_{\mathbf{x}=\mathbf{x}^p}, \quad (3)$$

and is commonly known as the matrix of weighting functions or Jacobians. The error covariance matrix for the solution is given by

$$\mathbf{S}_{\mathbf{x}^p} = \left[ \sum_i \mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} \mathbf{K}_i \right]^{-1}. \quad (4)$$

As will be seen later, in the MLS case, most retrieval calculations are sufficiently linear that only one iteration is required.

The division of the measurements into several separate vectors is somewhat arbitrary. It would be mathematically equivalent to consider one amalgamated vector. For v5 however, as described in sections 3.3 and 4, the division was maintained in order to simplify the software.

### 3.2. Use of virtual measurements

In most cases, the matrix to be inverted by (2) is singular. This indicates that there are aspects of the state vector about which the measurement system has been unable to provide any information. This singularity can be avoided by the use of additional measurement vectors containing ‘virtual measurements’ (as opposed to ‘real’ measurements such as radiances). In v5, as is typical, these take the form of a priori estimates of some or all elements of the state vector, usually formed from some climatology or model data. We choose to represent these by the vector  $\mathbf{a}$  of the same length and physical meaning as  $\mathbf{x}$ , with associated covariance  $\mathbf{S}_a$ . The weighting function matrix associated with  $\mathbf{a}$  is simply the  $n \times n$  identity matrix. Equation (2) then becomes:

$$\mathbf{x}^{p+1} = \mathbf{x}^p + \left[ \mathbf{S}_a^{-1} + \sum_i \mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} \mathbf{K}_i \right]^{-1} \left( \mathbf{S}_a^{-1} [\mathbf{a} - \mathbf{x}^p] + \sum_i \mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} [\mathbf{y}_i - \mathbf{f}_i(\mathbf{x}^p)] \right), \quad (5)$$

with solution error covariance

$$\mathbf{S}_{x^p} = \left[ \mathbf{S}_a^{-1} + \sum_i \mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} \mathbf{K}_i \right]^{-1}. \quad (6)$$

There is a subtlety in that there can be some elements of  $\mathbf{x}$  for which an a priori estimate is not required. For these, the corresponding rows and columns of  $\mathbf{S}_a^{-1}$  are set to zero (note that this makes  $\mathbf{S}_a^{-1}$  singular and therefore  $\mathbf{S}_a$  undefined, though this is not a problem as  $\mathbf{S}_a$  is never explicitly required). An example in v5 of such an exception is tangent pressure, as described in section 3.8.

Care must be taken when using data that have been retrieved in conjunction with a priori information. The error covariance of the solution must be compared with that of the a priori. If the solution covariance is little changed from the a priori covariance, then this indicates that the radiance measurements have added little information to the state vector, and the result is strongly biased toward the a priori.

The addition of the a priori term changes the  $\chi^2$  hypersurface to favor solutions close to the a priori. In addition, it is possible to favor solutions whose deviations from the a priori are correlated (i.e., smoother) by using non-zero values for the off-diagonal terms in the a priori covariance matrix. In v5, sections of the a priori covariance matrix describing particular parameters (e.g., temperature and composition profiles) are filled according to

$$[\mathbf{S}_a]_{ij} = \sqrt{[\mathbf{S}_a]_{ii} [\mathbf{S}_a]_{jj}} \exp \left[ -\frac{|z_i - z_j|}{l} \right], \quad (7)$$

where  $z_i, z_j$  are the height of surfaces  $i$  and  $j$  and  $l$  is some characteristic correlation length scale, all in log pressure coordinates (see section 3.6 for a discussion of the MLS vertical coordinate system). An exponential form is used, corresponding to the covariance that would be obtained from a Markov process (i.e., random walk). Forms such as Gaussian that are ‘broader’ are numerically unstable, as the resulting covariance matrix is close to being positive indefinite, except for small (i.e., not particularly effective) values of  $l$ .

### 3.3. Constrained quantity error propagation

For efficiency and simplicity, retrieval algorithms are often implemented in a series of phases. For example, in v5, for the data taken up to April 1993, the first phase is a retrieval of temperature and tangent pressure taken from the observations of molecular oxygen emission (band 1). This is followed by a retrieval of stratospheric ozone and water vapor (bands 5 and 6). The previously-obtained temperature and pressure values are used as constrained parameters in the forward model for these later retrievals. However, the knowledge of these ‘constrained quantities’ is imperfect; their error covariance has been estimated in the earlier phase by (6). This uncertainty is accounted for in the later retrievals by adding an additional term to the error covariance of the radiances used in these later phases, according to:

$$\mathbf{S}_{y_i} \rightarrow \mathbf{S}_{y_i} + \mathbf{K}_c \mathbf{S}_c \mathbf{K}_c^T, \quad (8)$$

where  $\mathbf{K}_c$  is the matrix of weighting functions describing the sensitivity of the radiances to be used in the later phase (e.g., emission from ozone and water vapor) to the constrained quantities (e.g., temperature and pressure), and  $\mathbf{S}_c$  is the error covariance of the constrained quantities estimated by (6) in the earlier phase.

The original  $\mathbf{S}_{y_i}$  matrices for MLS, are diagonal, as there is no significant correlation in radiance noise between channels or integration periods. This makes the inversion of  $\mathbf{S}_{y_i}$  in (5) computationally simple. However, including the uncertainty in the constrained quantities in the error budget

makes the updated  $\mathbf{S}_{y_i}$  matrices non-diagonal. Their inversion becomes the most computationally intensive aspect of the calculation, due to the large number of measurements compared to state vector elements.

Approximations such as ignoring the off-diagonal terms in the new  $\mathbf{S}_i$  matrices are undesirable, as they correspond to a loss of information, tying the retrieval closer to the a priori information. The approach taken in v5 was to partition the problem to achieve an acceptable compromise between speed and accuracy. For selected bands, the full non-diagonal error propagation calculation was performed on all the radiances whose tangent points were at pressures greater than a given threshold (22 hPa), with simple diagonal propagation used at smaller pressures. Radiance observations above and below this threshold tangent pressure were divided into separate measurement vectors (Equation (2)), and the corresponding  $\mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} \mathbf{K}_i$  calculation was performed using either the full  $\mathbf{S}_{y_i}$  matrix or its diagonal. Section 4 gives details of the specific implementation of this calculation in v5.

### 3.4. Diagnosing retrieval performance

A very important diagnostic of retrieval performance is the estimated precision. As explained in section 3.2, this should be compared with the precision placed on the a priori estimates of the state vector components. As an aid to users of MLS data, the sign of the precision in the data files is set negative if it is no better than 50% of the a priori precision. This indicates that at least 25% of the information in the result has come from the a priori, and therefore the data should not be used.

Another diagnostic of retrieval performance is the  $\chi^2$  statistic given in (1). Large values of  $\chi^2$  may indicate instrument anomalies, or deficient instrument calibration, spectroscopy information, or inaccurate forward models. In v5, the  $\chi^2$  information is summarized by a set of flags that indicate to users of the data which profiles can be considered reliable. These flags are discussed in section 5.

Another useful measure of retrieval performance is the matrix of averaging kernels ( $\mathbf{A}$ ) [Rodgers, 1990], given by

$$\mathbf{A} = \frac{\partial \hat{\mathbf{x}}}{\partial \mathbf{x}_t} = \left[ \mathbf{S}_a^{-1} + \sum_i \mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} \mathbf{K}_i \right]^{-1} \sum_i \mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} \mathbf{K}_i, \quad (9)$$

where  $\mathbf{x}$  is the state vector obtained from the retrieval and  $\mathbf{x}_t$  describes the unknown true state of the atmosphere. The columns of  $\mathbf{A}$  describe the response of the retrieval system to delta function perturbations in the atmosphere. The rows of  $\mathbf{A}$  indicate which parts of the atmosphere have contributed to each retrieval level. The widths of the peaks seen in these rows are a useful measure of the vertical resolution of the

retrieved dataset. In the ideal case  $\mathbf{A}$  would be the identity matrix.

### 3.5. The Marquardt-Levenberg approach

Some of the retrieval calculations are sufficiently nonlinear that the Marquardt-Levenberg algorithm is more appropriate than the standard Gauss-Newton method. This formulates the retrieval as:

$$\mathbf{x}^{p+1} = \mathbf{x}^p + \left[ \gamma \mathbf{I}_n + \mathbf{S}_a^{-1} + \sum_i \mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} \mathbf{K}_i \right]^{-1} \left( \mathbf{S}_a^{-1} [\mathbf{x} - \mathbf{a}] + \sum_i \mathbf{K}_i^T \mathbf{S}_{y_i}^{-1} [\mathbf{y}_i - \mathbf{f}_i(\mathbf{x}^p)] \right), \quad (10)$$

where  $\mathbf{I}_n$  is the  $n \times n$  identity matrix, and  $\gamma$  is a scalar. As  $\gamma$  is reduced, this iteration tends toward the Gauss-Newton iteration of (5). Conversely, as  $\gamma$  is increased, this iteration tends toward the more cautious steepest descent algorithm with a step size proportional to  $\gamma^{-1}$ . In v5 the Marquardt-Levenberg algorithm is implemented by first setting  $\gamma$  to some fairly large value that is then increased or decreased by some factor each iteration, according to the change seen in  $\chi^2$ . If  $\chi^2$  decreases, then the value of  $\gamma$  is decreased, making the next iteration more aggressive. If, conversely,  $\chi^2$  increases, then the result of the iteration is rejected, and a new iteration is attempted using a larger value of  $\gamma$ .

### 3.6. Construction of the MLS state vector

The most important components of the state vector for v5 are vertical profiles of temperature and species abundances fit to a set of fixed pressure levels. The geopotential height of the 100 hPa pressure surface is also a state vector element (storing a vertical profile of geopotential height is inappropriate, as the information it would convey is already present in the temperature profile when hydrostatic balance, implicit in all the forward model calculations, is imposed).

In the case of temperature and composition, the terms in the state vector describe the tie points in a piecewise linear representation of the vertical profile (temperature or volume mixing ratio). An exception to this is water vapor, for which the representation basis below 100 hPa represents a piecewise linear interpolation in log mixing ratio. This is a better model of the vertical structure of water vapor profiles in this region, which display quasi-exponential growth with increasing pressure, and is equivalent to a linear interpolation in relative humidity with respect to ice. All the forward model calculations accurately model these representations (rather than using, for example, a layer mean).

The task of the retrieval algorithm is to deduce an optimum value of the state vector consistent with the observed radiances, tangent point altitudes, and a priori information. The forward model calculations that form part of the algorithm need some additional parameters in order to match these observations. The most important of these are the limb tangent point pressures (or more precisely  $-\log_{10}[\text{Tangent pressure} / \text{hPa}]$ , as this is a less non-linear variable) for each observation. The tangent point pressures for the 63-GHz observations are retrieved from the molecular oxygen signal in the 63-GHz radiances. The tangent pressures for the other radiometers are deduced from the 63-GHz tangent pressures, using estimates of the angular offsets of the radiometer fields of view taken from ground- and space-based calibration. These angular offsets are part of the state vector (although not retrieved).

Some atmospheric constituents (e.g., nitrous oxide, ozone isotopes, etc.) contribute to the MLS radiances in a manner too insignificant to yield useful MLS observations of their abundance, but not so insignificant that their effects can be neglected. The abundances of these constituents are also included in the state vector, and constrained to climatological values.

Our use of the term ‘state vector’ is different from that adopted in, for example, *Rodgers* [1976]. We use the term state vector to describe all the variable (or potentially variable) parameters needed by the forward model, whether they are retrieved or not.

Most of the MLS data products are based upon measurements of spectral contrast (channel to channel variations in the observed radiances). The instrument resolves a spectral line, or a set of lines, and the spectral form of the signal yields information on the state vector. The observed spectral lines are superimposed on some spectrally flat offset, which arises through both instrumental effects such as thermal emission from the antenna, and atmospheric effects such as continuum emission.

The offset is modeled in the retrieval as a scan independent absolute radiance offset, plus a vertical profile of extinction coefficient on the same pressure surfaces as used for the atmospheric constituents. The former is intended to capture the instrumental effects, which are typically independent of scan angle; the latter is a good model of unexplained atmospheric absorption (e.g., due to forward model approximations or spectroscopic uncertainties). In order to partition these appropriately, the extinction coefficient is not retrieved for pressures less than 0.068 hPa. There are two exceptions to this scheme. The first is the case of the retrieval of temperature, tangent pressure and geopotential height from the 63-GHz radiances. Here, an independent radiance baseline offset is retrieved for each tangent height, as this was found

to better model the residual radiances seen in this band. The second exception is the UTH retrieval, which is based on observations of absolute radiance as opposed to spectral contrast, so baseline or extinction retrievals are not appropriate.

### 3.7. The MLS Suite of Forward Models

For the majority of the MLS channels, the forward model used in v5 is based on a linear approximation (see section 3.7.6). The coefficients used in this approximation are taken from the results of a ‘full’ forward model. The ‘full’ model is also used directly in v5 for appreciably nonlinear radiances used for the upper tropospheric humidity (UTH) retrieval and for some channels in the retrieval of the 183-GHz H<sub>2</sub>O and O<sub>3</sub>.

The full forward model is given by

$$\hat{I}_{\text{ch}}^L = r_u \frac{\int_{\Omega_A} \int_{\nu_{l_0}}^{\infty} I(\nu, \Omega, \mathbf{x}) \Phi(\nu) G(\Omega, \Omega_t, \nu) d\Omega d\nu}{\int_{\Omega_A} \int_{\nu_{l_0}}^{\infty} \Phi(\nu) G(\Omega, \Omega_t, \nu) d\Omega d\nu} + r_l \frac{\int_{\Omega_A} \int_{-\infty}^{\nu_{l_0}} I(\nu, \Omega, \mathbf{x}) \Phi(\nu) G(\Omega, \Omega_t, \nu) d\Omega d\nu}{\int_{\Omega_A} \int_{-\infty}^{\nu_{l_0}} \Phi(\nu) G(\Omega, \Omega_t, \nu) d\Omega d\nu}, \quad (11)$$

where  $\hat{I}_{\text{ch}}^L$  is the measured calibrated radiance for channel ch,  $\nu_{l_0}$  is the local oscillator frequency,  $r_u$  is the higher frequency (relative to  $\nu_{l_0}$ ) sideband relative response,  $r_l$  is the lower frequency sideband relative response ( $r_l + r_u \equiv 1$ ).  $I(\nu, \Omega, \mathbf{x})$  is the atmospheric limb radiance,  $\Phi(\nu)$  is the instrument spectral response for channel ch,  $G(\Omega, \Omega_t, \nu)$  is the antenna field of view (FOV) response function,  $\nu$  is frequency,  $\Omega$  is solid angle,  $\Omega_t$  is the FOV boresight direction,  $\Omega_A$  is the integration limit in solid angle, and  $\mathbf{x}$  is the forward model state vector. The state vector contains coefficients for evaluating a vertical profile of the constituent concentrations ( $f$ ) and temperature ( $T$ ), negative base 10 logarithm of the FOV direction limb tangent pressure ( $\zeta_t$ ), and some geometric and hydrostatic quantities for converting tangent pressure into pointing angles and heights. Embedded in the forward model is hydrostatic equilibrium, which relates height to pressure and temperature.

Note that by taking  $r_u$  and  $r_l$  outside the frequency integral, we have neglected their possible small variations over the width of an individual filter channel. Also note that we have neglected the small variations in field of view response over the filter width by separating  $G$  and  $\Phi$ . This approximation is further extended by using the same  $G(\Omega, \Omega_t, \nu)$  for all channels in a radiometer. The FOV width across a radiometer changes by less than 1% across all its channels which is smaller than the measured FOV beam width uncertainty of 5% [*Jarnot et al.*, 1996]. A 5% beam width error is shown to have negligible impact upon the 205 and 183 GHz

ozone retrievals [Froidevaux et al., 1996]. This result is expected to hold true for all retrieved species.

The next four sub-subsections describe the components of this calculation, starting with the field of view integration and finishing with the computation of  $I(\nu, \Omega, \mathbf{x})$ . The fifth sub-subsection describes the forward model derivative algorithm.

**3.7.1. Field of view integration** The FOV integration assumes there is no frequency variation in the antenna gain function  $G$  inside a band, which makes the spectral and spatial integrations separable. The limb radiance is assumed to have negligible variability across the <24 km azimuth (horizontal) width of the FOV, allowing the FOV shape to be collapsed into a one dimensional function in elevation (vertical) angle,  $\chi$  ( $G[\Omega, \Omega_t, \nu] \rightarrow G[\chi, \chi_t, \nu]$ ). As the FOV function in  $\chi$  is the same at all pointing positions,  $\chi_t$ , the spatial integral can be converted into a convolution integral, and solved with fast Fourier transforms. This gives

$$\hat{I}_{\text{ch}}^L(\chi_t, \mathbf{x}) = \mathcal{F}^{-1} \left\{ \mathcal{F} \left[ \bar{I}_{\text{ch}}^L(\chi, \mathbf{x}) \right] \mathcal{F} [G(\chi_t - \chi)] \right\}, \quad (12)$$

where  $\bar{I}_{\text{ch}}^L$  is the spectrally integrated limb radiance for channel ch,  $\mathcal{F}$ , and  $\mathcal{F}^{-1}$  are the Fourier transform operator and its inverse respectively. Elevation angles  $\chi$  and  $\chi_t$  are computed by

$$\chi = \sin^{-1} \left[ \frac{(1+n) h_c \min(h_c, h_{\oplus})}{h_s h_{\oplus}} \right], \quad (13)$$

where  $h_c$  is the unrefracted tangent geocentric altitude for elevation angle  $\chi$ ,  $1+n$  is the refractive index at  $h_c$ ,  $h_{\oplus}$  is the geocentric Earth radius, and  $h_s$  is the geocentric satellite altitude. The refractive index minus one,  $n$ , is given by

$$n = \frac{7.76 \times 10^{-5} P}{T} \left[ 1 + 4810 \frac{f^{\text{H}_2\text{O}}}{T} \right], \quad (14)$$

where  $P(=10^{-\zeta_r})$  is tangent pressure in hPa,  $T$  is tangent temperature in Kelvins, and  $f^{\text{H}_2\text{O}}$  is the tangent water vapor volume mixing ratio. The sine of the incident line of sight (LOS) angle perpendicular to the tangent surface is  $\min(h_c, h_{\oplus})/h_{\oplus}$ , which is unity except for Earth intersecting rays. Geocentric altitude,  $h_c$ , is computed from the hydrostatic equation, which depends on the temperature profile, tangent pressure, and a reference height. Its calculation is based on those described in section 3.8. A cubic spline computation [de Boor, 1987] interpolates the computed radiances to a grid of 1024 equally-spaced points (covering  $\pm 0.1$  radians) in the convolution. There will be a small contribution due to signals impinging on the antenna at absolute angles greater than 0.1 radians which is estimated and removed from the calibrated radiances produced by level 1 processing.

**3.7.2. Spectral integration** The spectral integration is

$$\bar{I}_{\text{ch}}^L(\chi, \mathbf{x}) = r_u \frac{\int_{\nu_{\text{lo}}}^{\infty} I(\nu, \mathbf{x}) \Phi(\nu) d\nu}{\int_{\nu_{\text{lo}}}^{\infty} \Phi(\nu) d\nu} + r_l \frac{\int_{-\infty}^{\nu_{\text{lo}}} I(\nu, \mathbf{x}) \Phi(\nu) d\nu}{\int_{-\infty}^{\nu_{\text{lo}}} \Phi(\nu) d\nu} \quad (15)$$

where  $I(\nu, \mathbf{x}) \equiv I(\nu, \Omega, \mathbf{x})$  but with  $\Omega \rightarrow \chi$  dependence dropped because it is dependent on  $h_{\oplus}$  and  $h_s$ , which are state vector elements, and  $h_c$  and  $n$  which are functions of the state vector quantities tangent pressure, temperature, water vapor, and reference geopotential height. The spectral integration uses a 161 point trapezoidal quadrature, based on the measured channel filter shape,  $\Phi(\nu)$ . The radiative transfer calculation is evaluated on far fewer points and interpolated to the frequencies represented by the 161 points. The spectral characteristics of the radiance signal within the channel determines the method and the frequency gridding used by the radiative transfer calculations. In some cases a ‘pre-frequency-averaging’ approximation can be made. With this, only one radiance calculation per channel is performed, using a precomputed filter-shape-weighted average absorption coefficient derivative with respect to mixing ratio (cross section). This can be implemented in cases where radiances are optically thin (total single sideband radiances less than 100 K), such as ozone in band 4, or in any band having very little spectral variation irrespective of absolute radiance, such as bands 2 and 3.

Bands 1, 5, and 6 have optically thick signals with large spectral variation. The signals from these are calculated with a filter-weighted average of radiance calculations. If there is only one spectral line in a channel, the radiances are computed on a 25 point equally-spaced frequency grid within the channel, and an Aitkin’s  $\Delta^2$  method is used [Isaacson and Keller, 1994]. The filter weighted integral is evaluated three times using radiance calculations at 7, 13 and finally all 25 frequencies. Each time the radiances are interpolated with cubic splines to the 161 point filter shape frequency grid and evaluated with a trapezoid quadrature. The results form a three term series converging to a better solution. This approach is used for center channels in band 1 and all the channels in bands 5 and 6.

Aitkin’s method does not work well in cases where there are multiple spectral lines within the channel (e.g., the  $^{18}\text{O}$  and  $^{17}\text{O}$  lines in the wing channels of band 1). In these cases an adaptive computation is implemented. The adaptive computation starts by evaluating the radiative transfer on  $2n+1$  frequency points, where  $n$  is the number of lines in the channel. The initial frequency grid consists of points at the channel boundaries, line centers, and midway between lines. This defines  $2n$  integration regions. In each region, two in-

tegrals are evaluated: one using only the two end points, the other also including a point at the center of the region. If the difference between these is below a specified threshold, the integration between these points has adequately converged and the calculation proceeds to the next unconverged region, otherwise the region is split at the center and the process starts again. When all regions between the channel boundaries are converged, the calculation is complete.

**3.7.3. Radiative transfer calculation** The MLS limb radiance,  $I(\nu, \mathbf{x})$ , is modeled with a nonpolarized, nonscattering, radiative transfer calculation for atmospheric emitters in local thermodynamic equilibrium. The integration, along the LOS path, beginning at the instrument ( $s = 0$ ), passing through the atmosphere ( $s > 0$ ) and out into space ( $s \rightarrow \infty$ ), is given by

$$\begin{aligned} I(\nu, \mathbf{x}) &= I_o(\nu) \Upsilon \tau(t, \infty, \nu, \mathbf{x}) \tau(0, t, \nu, \mathbf{x}) \\ &\quad + B[\nu, T(t, \mathbf{x})] (1 - \Upsilon) \tau(0, t, \nu, \mathbf{x}) \\ &+ \Upsilon \tau(0, t, \nu, \mathbf{x}) \int_{s=\infty}^{s=t} B[\nu, T(s, \mathbf{x})] \alpha(\nu, \mathbf{x}, s) \tau(t, s, \nu, \mathbf{x}) ds \\ &\quad + \int_{s=t}^{s=0} B[\nu, T(s, \mathbf{x})] \alpha(\nu, \mathbf{x}, s) \tau(0, s, \nu, \mathbf{x}) ds, \quad (16) \end{aligned}$$

where  $B[\nu, T(s, \mathbf{x})]$  is the Planck blackbody function,  $T(s, \mathbf{x})$  is the temperature, which is a part of state vector  $\mathbf{x}$ ,  $I_o$  is the cosmic space radiance,  $\tau(a, b, \nu, \mathbf{x})$  is the transmission function between points  $a$  and  $b$  along  $s$ ,  $\Upsilon$  is the Earth surface reflection coefficient,  $t$  is the tangent point on  $s$ , and  $\alpha(\nu, \mathbf{x}, s)$  is the absorption coefficient. The transmission function is given by

$$\tau(a, b, \nu, \mathbf{x}) = \exp \left[ - \int_{s'=a}^{s'=b} \alpha(\nu, \mathbf{x}, s') ds' \right], \quad (17)$$

where  $a$  and  $b$  are the integration limits and  $s'$  is path length. The Earth reflection coefficient,  $\Upsilon$  is 1 for non Earth intersecting rays and 0.05 for Earth intersecting rays. The limb viewing geometry used by MLS makes the measurements insensitive to  $\Upsilon$  for the LOS Earth intersecting paths, and therefore a model that considers surface reflectivity variations is not needed. Equation (16) is solved piece-wise by a summation over layers, using the discrete differential tem-

perature radiative transfer equation [Read et al., 1995],

$$\begin{aligned} I(\nu, \mathbf{x}) \approx I_t &= \sum_{i=t}^N \Delta B_i \prod_{k=i}^{N-1} \Delta \tau_{kt} \\ &- \Upsilon \left( \prod_{k=t}^{N-1} \Delta \tau_{kt} \right) \times \left[ \sum_{i=t}^N \Delta B_i \prod_{k=t}^{i-1} \Delta \tau_{kt} \right. \\ &\quad \left. - I_o \left( \prod_{k=t}^{N-1} \Delta \tau_{kt} \right) \right]. \quad (18) \end{aligned}$$

Equation (18) is applied to an  $N - 1$  layered atmosphere where each layer is separated by surfaces with indices running from 1 (the Earth surface) to  $N$  (top of the  $N - 1$  layer).

Radiances are computed for a set of LOS tangents that are a subset (indices  $t$ ) of the  $N$  surfaces (interpolation to the required tangents is performed later). Subscripts  $i$  and  $k$  indicate the surfaces used in the products and sums. The quantities  $\Delta B_i$  are  $(B_{i-1} - B_{i+1})/2$ , except when  $i = N$ , where  $\Delta B_N$  is  $(B_{N+1} + B_N)/2$ . The Planck blackbody function  $B_i$  is divided by Boltzmann's constant to yield radiances in units of Kelvins, as reported in the MLS Level 1 data [Jarnot et al., 1996]. The four product terms are the atmospheric LOS transmissions from (1) layer  $i$  to the instrument, (2) tangent layer  $t$  to the instrument, (3) layer  $i$  to the tangent layer  $t$ , and (4) from space to layer  $t$ . Any product for a term which does not exist (e.g., when  $i = N$  in the first term and  $i = t$  in the third term), is set to unity. The layer transmission  $\Delta \tau_{kt}$  is given by

$$\Delta \tau_{kt} = \exp \left[ - \frac{\Delta s_{k \rightarrow k+1}^{\text{refr}}}{\Delta s_{k \rightarrow k+1}} \int_{\zeta_k}^{\zeta_{k+1}} \alpha(\nu, \mathbf{x}, \zeta) \frac{ds}{dh} \frac{dh}{d\zeta} d\zeta \right] \quad (19)$$

where  $\Delta s_{k \rightarrow k+1}^{\text{refr}}/\Delta s_{k \rightarrow k+1}$  is the correction for refraction,  $ds/dh$  is the unrefracted path length derivative with respect to height, and  $dh/d\zeta$  is the height derivative with respect to negative logarithm of pressure, and  $\zeta$  is the negative base 10 logarithm of pressure along the LOS path  $s$ . The refraction correction is the ratio of the refracted layer path length to the unrefracted path length. The refracted path length is

$$\Delta s_{k \rightarrow k+1}^{\text{refr}} = \int_{\zeta_k}^{\zeta_{k+1}} \frac{(1+n)h}{\sqrt{((1+n)h)^2 - ((1+n_t)h_t)^2}} \frac{dh}{d\zeta} d\zeta. \quad (20)$$

The absorption coefficient is a sum over each species contribution,

$$\alpha(\nu, \mathbf{x}, \zeta) = \sum_l f^l \beta^l(T, \zeta, \nu), \quad (21)$$

where  $\beta^l(T, \zeta, \nu)$  is species  $l$  cross section (absorption coefficient per unit volume mixing ratio, vmr), and  $f^l$  is its vmr

expressed either as a linear function,

$$f^l = \sum_m F_m^l \eta_m^l(\zeta), \quad (22)$$

or for upper H<sub>2</sub>O at pressures greater than 100 hPa, as a logarithmic function,

$$f^l = \exp \left[ \sum_m \ln \left( F_m^l \right) \eta_m^l(\zeta) \right], \quad (23)$$

where  $F_m^l$  is a mixing ratio coefficient and  $\eta_m^l$  is a representation basis function. The representation basis,  $\eta_m^l$ , is a triangular shaped function with a unit value at the coefficient break-point pressure,  $\zeta_m$ , and linearly decreasing to zero at pressures  $\zeta_{m+1}$  and  $\zeta_{m-1}$ . These basis functions give a state vector describing linear interpolation in the vertical. Representation basis functions are discussed more fully in *Froidevaux et al.* [1996]. The linear function given in (22) is used for all species, including upper tropospheric humidity (UTH) which has units of relative humidity with respect to ice (%RH<sub>i</sub>). However, in order to evaluate radiances, UTH needs to be transformed into vmr represented by (23). The transformation between UTH and vmr is described more fully in *Read et al.* [2001].

The forward model calculation for all species except UTH is performed on a layered atmosphere having 91 integration layers (1 km thick between 0 and 70 km and 2 km thick from 70 to 110 km). Equation (19) is solved with 8-point Gauss-Legendre quadrature. Radiances are computed at 81 fixed tangent heights; 10 tangents are for rays intersecting or skimming the Earth (with tangent heights from 0.0 to -100 km below the Earth surface), and 71 tangents above the Earth. The UTH retrieval uses channel 30 at 202/204 GHz, in an atmospheric window that permits measurements into the troposphere. The UTH forward model uses 35 integration layers, 19 nearly equally-spaced levels between the surface and 18 km and 16 equally-spaced levels between 20 and 50 km. Radiances are computed at 34 tangent heights above the Earth's surface and 10 Earth surface skimming and intersecting rays.

The O<sub>2</sub> emission in the center of band 1 is polarized because of the Earth's magnetic field. While a polarized forward model has been developed, its computational requirements prohibited its use in v5. The radiances in the center 3 channels in band 1 are therefore not used except the lowest tangent heights, for which an empirical forward model has been implemented (see section 3.9).

**3.7.4. Cross section calculations** The cross section,  $\beta$ , in (21) is [*Pickett et al.*, 1992]

$$\beta = \sqrt{\frac{\ln 2}{\pi}} \frac{10^{-6}}{w_d k T} P \left[ \sum_j 10^{\delta_j} \text{LineShape}(x_j, y_j, z_j) \right], \quad (24)$$

where

$$\begin{aligned} \delta_j &= \delta_j(300) - \log \{ Q_{\text{ratio}} [Q(300, 225, 150), T] \} \\ &\quad + \frac{h E l_j}{k} \left( \frac{1}{300} - \frac{1}{T} \right) \\ &\quad + \log \left\{ \frac{1 - \exp[-h v_j / k T]}{1 - \exp[-h v_j / k(300)]} \right\}, \quad (25) \end{aligned}$$

$T$  is temperature in Kelvins,  $P$  ( $= 10^{-\zeta}$ ) is pressure in hPa,  $\delta_j(300)$  is the logarithm of the integrated intensity in nm<sup>2</sup> MHz at 300 K,  $v_j$  is the pressure shifted line center frequency in MHz,  $E l_j$  is the ground state energy in cm<sup>-1</sup>,  $\log \{ Q_{\text{ratio}} \}$  is the logarithm of the ratio of the partition function at  $T$  to the partition function at 300 K,  $Q(300, 225, 150)$  are values of the partition function at 300, 225, and 150 K, and  $h$  and  $k$  are Planck and Boltzmann constants, respectively. The partition function ratio is evaluated with a linear interpolation of the logarithm of the partition functions and the logarithm of temperature. The spectral parameters,  $\delta_j(300)$ ,  $Q(300, 225, 150)$ ,  $E l_j$ , and  $v_j$  are taken from the JPL submillimeter, millimeter, and microwave spectral line catalog [*Pickett et al.*, 1992].

$$w_d = \frac{v_j \sqrt{2 \ln 2 k T}}{c \sqrt{\mathcal{M}}} \quad (26)$$

is the Doppler width in MHz,  $\mathcal{M}$  is the absorber molecular mass in amu, and  $j$  identifies the individual lines or quantum states in the molecule, and  $c$  is the speed of light. The line shape function is a convolution of a collision contribution [*van Vleck and Weisskopf*, 1945] and a velocity broadening (Doppler) contribution,

$$\begin{aligned} \text{LineShape}(x_j, y_j, z_j) &= \left( \frac{v}{v_j} \right)^2 \\ &\times \left\{ \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{[y_j - Y_j(x_j - t)] \exp(-t^2)}{y_j^2 + (x_j - t)^2} dt \right. \\ &\quad \left. + \frac{1}{\sqrt{\pi}} \frac{y_j - Y_j z_j}{z_j^2 + y_j^2} \right\}, \end{aligned}$$

where

$$\begin{aligned} x_j &= \frac{\sqrt{\ln 2} (v_j - \nu)}{w_d}, \\ y_j &= \frac{\sqrt{\ln 2} w_{cj} P \left( \frac{300}{T} \right)^{n_j}}{w_d}, \\ z_j &= \frac{\sqrt{\ln 2} (v_j + \nu)}{w_d}. \end{aligned}$$

$w_{cj}$  is the collision width in MHz hPa<sup>-1</sup> at 300 K,  $n_j$  is its temperature dependence,  $Y_j$  is an intramolecular line mixing contribution,  $\nu_j$  is the line position frequency in MHz,  $\nu$  is the radiation frequency in MHz. The  $(\nu/\nu_j)^2$  term, which is virtually constant over a Doppler width, has been pulled outside the integral, giving the well studied Voigt integral [Shippony and Read, 1993]. The line center frequency is pressure shifted according to

$$\nu_j = \nu_{j0} + \Delta\nu_{j0} P \left( \frac{300}{T} \right)^{\frac{1+6n_j}{4}}, \quad (27)$$

where  $\nu_{j0}$  is the ‘zero pressure’ line center frequency in MHz,  $\Delta\nu_{j0}$  is the pressure shift parameter in MHz hPa<sup>-1</sup> at 300 K, and the temperature dependence is related to  $n_j$  [Pickett, 1980]. The intramolecular line mixing contribution is parameterized according to

$$Y_j = P \left[ \delta_j \left( \frac{300}{T} \right)^{0.8} + \gamma_j \left( \frac{300}{T} \right)^{1.8} \right] \quad (28)$$

where  $\delta_j$  and  $\gamma_j$  are line-mixing coefficients in hPa<sup>-1</sup> at 300 K. The linewidth parameters,  $w_{cj}$  and  $n_j$ , are based on laboratory measurements. The line shift,  $\Delta\nu_{j0}$ , is nonzero only for H<sub>2</sub>O [Pumphrey and Bühler, 2000]. The line mixing coefficients are nonzero only for O<sub>2</sub> [Liebe et al., 1992].

For the pre-frequency-averaged calculations, cross sections and their derivatives are computed on a molecule by molecule basis. The contributions from each line inside the MLS band, wings of strong lines outside the band, and background continuum [Read et al., 2001] are summed and weighted by the MLS filter response function to produce a cross section for each channel. These are computed for a range of temperatures and pressures and stored in tables. The temperature dependence of  $\beta$  is approximated with

$$\beta = \beta (300) (300/T)^n \quad (29)$$

and  $n$  is computed and stored. Also computed is  $d\beta/d\nu$ , the derivative of channel cross section with respect to molecular velocity. This is used for computing the radiance derivative with respect to molecular velocity.

For situations where the radiances are frequency averaged, the cross section is computed as needed. The calculation includes only those lines within the band for each species under consideration and approximates contributions from strong line wings and its continuum with an empirical polynomial function of pressure and temperature. The line data,  $\nu_{j0}$ ,  $El_j$ ,  $\mathcal{I}_j$  (300),  $w_{cj}$ ,  $n_j$ ,  $Y_j$ , and  $\Delta\nu_{j0}$ , and coefficients of the polynomial function are stored in a file cataloged by species and MLS band that is read by the forward model program if needed.

The UTH forward model described in detail by Read et al. [2001] uses spectrally averaged cross sections. H<sub>2</sub>O and air cross sections are empirical continuum functions determined from in-orbit data. Cross sections for the contributing species O<sub>3</sub>, N<sub>2</sub>O, and HNO<sub>3</sub> are computed from a line by line calculation.

**3.7.5. Forward model derivatives** In addition to radiances, the forward model computes radiance derivatives with respect to the state vector for use in the retrieval. As with radiance calculations, this is a three part calculation including FOV and spectral integrations and a radiative transfer derivative calculation.

The FOV radiance derivative calculation is given by:

$$\begin{aligned} \frac{\partial \hat{I}_{\text{ch}}^{\text{L}}}{\partial x_j} &= \int_{-\infty}^{\infty} \left[ \frac{\partial \overline{T}_{\text{ch}}^{\text{L}}}{\partial x_j} + \overline{T}_{\text{ch}}^{\text{L}} \frac{\partial}{\partial \chi} \left( \frac{\partial \chi}{\partial x_j} \right) \right] G(\chi_p - \chi) d\chi \\ &\quad + \frac{\partial \chi_p}{\partial x_j} \int_{-\infty}^{\infty} \overline{T}_{\text{ch}}^{\text{L}} \frac{\partial G(\chi_p - \chi)}{\partial (\chi_p - \chi)} d\chi \\ &\quad - \int_{-\infty}^{\infty} \overline{T}_{\text{ch}}^{\text{L}} \frac{\partial \chi}{\partial x_j} \frac{\partial G(\chi_p - \chi)}{\partial (\chi_p - \chi)} d\chi. \quad (30) \end{aligned}$$

The multiterm equation involving derivatives of the antenna gain function,  $G(\chi_p - \chi)$ , as well as the spectrally integrated derivatives of the radiative transfer function,  $\overline{T}_{\text{ch}}^{\text{L}}$ , is a consequence of using pressure as the independent vertical coordinate. The FOV function varies with some state vector components when projected onto the vertical coordinate system. Referring to (13), the elements of  $\mathbf{x}$  which have nonzero  $\partial \chi / \partial x_j$  are temperature,  $T$ , Earth radius,  $h_{\oplus}$ , and satellite radius,  $h_s$  (the dependencies of  $n$  are neglected). The partial

derivatives for these terms are,

$$\begin{aligned}
\frac{\partial \chi}{\partial T_m} &= \frac{\tan \chi}{h_c} \frac{dh_c}{dT_m}, \\
\frac{\partial}{\partial \chi} \left( \frac{\partial \chi}{\partial T_m} \right) &= \frac{2 + \tan^2 \chi}{h_c} \frac{dh_c}{dT_m} + \frac{\eta_m^T}{T}, \\
\frac{\partial \chi}{\partial h_s} &= -\frac{\tan \chi}{h_s}, \\
\frac{\partial}{\partial \chi} \left( \frac{\partial \chi}{\partial h_s} \right) &= -\frac{1}{h_s \cos^2 \chi}, \\
\frac{\partial \chi}{\partial h_\oplus} &= \frac{\tan \chi}{h_c}, \text{ and} \\
\frac{\partial}{\partial \chi} \left( \frac{\partial \chi}{\partial h_\oplus} \right) &= \frac{1}{h_c \cos^2 \chi}.
\end{aligned} \tag{31}$$

The derivative,  $\partial G[\chi_p - \chi] / \partial [\chi_p - \chi]$  is evaluated using the Fourier transform derivative property

$$\mathcal{F} \left( \frac{\partial G(\chi)}{\partial \chi} \right) = iq \mathcal{F}(G(\chi)) \tag{32}$$

where  $q$  is the aperture coordinate (number of wavelengths) and  $i = \sqrt{-1}$ . This property is convenient because the pattern is stored as  $\mathcal{F}[G(\chi)]$  which ensures internal consistency between the pattern and its derivative. The integrals in (30) are evaluated with Fourier transforms analogous to (12).

The spectral integration of the radiance derivatives is identical to (15) used for radiances, with  $\frac{\partial \bar{I}_{\text{ch}}^L}{\partial x_j}$  and  $\frac{dI_t}{dx_j}$  replacing  $\bar{I}_{\text{ch}}^L$  and  $I_t$ . However, derivatives for all channels are computed with spectrally integrated ‘cross sections’ (i.e., ‘pre-frequency-averaging approximation’). This reduces the number of spectral radiance derivative calculations to one per channel. For those cases where the radiances are computed from a spectral integration of multiple radiative transfer calculations per channel, the derivative is scaled by the ratio of the spectrally integrated radiance to the radiance computed from a spectrally integrated cross section. In most cases, this approximation produces very good results.

The derivative of the radiative transfer equation with re-

spect to a state vector element is

$$\begin{aligned}
\frac{dI_t}{dx_j} &\equiv K_{tj} = \\
&\sum_{i=t}^N \left( \frac{d\Delta B_i}{dx_j} + \Delta B_i \sum_{k=i}^{N-1} \frac{d\Delta \delta_{kt}}{dx_j} \right) \prod_{k=i}^{N-1} \Delta \tau_{kt} \\
&\quad - \Upsilon \left( \prod_{k=t}^{N-1} \Delta \tau_{kt} \sum_{k=t}^{N-1} \frac{d\Delta \delta_{kt}}{dx_j} \right) \\
&\times \left[ \sum_{i=t}^N \left( \frac{d\Delta B_i}{dx_j} + \Delta B_i \sum_{k=i}^{i-1} \frac{d\Delta \delta_{kt}}{dx_j} \right) \prod_{k=i}^{i-1} \Delta \tau_{kt} \right. \\
&\quad \left. - 2I_o \left( \prod_{k=t}^{N-1} \Delta \tau_{kt} \right) \right], \tag{33}
\end{aligned}$$

where  $d\Delta \delta_{kt}/dx_j$  is the derivative of the layer opacity (the argument of the exponential function in (19)) with respect to state vector element  $x_j$ , and  $d\Delta B_i/dx_j$ , is the derivative of  $\Delta B_i$  with respect to state vector element  $x_j$  which is zero except for temperature. The layer opacity derivatives are evaluated analytically for each state vector element. The value for any sum term that does not exist (e.g., when  $i = N$  in the second sum in the first term and  $i = t$  in the second sum of the third term) is zero.

### 3.7.6. Forward model implementation in retrievals

The forward model described above is a very complex and time consuming calculation. It was not possible to implement this full calculation operationally for all the MLS channels. Instead a linear approximation to (11) from a Taylor series is implemented, according to:

$$\begin{aligned}
\hat{I}_{\text{ch}}^{L_0} &= r_u \left[ I_{\text{ch},u}^{*L_0} + \mathbf{K}_{\text{ch},u}^{*L_0} (\hat{\mathbf{x}} - \mathbf{x}^*) \right] \\
&\quad + r_l \left[ I_{\text{ch},l}^{*L_0} + \mathbf{K}_{\text{ch},l}^{*L_0} (\hat{\mathbf{x}} - \mathbf{x}^*) \right]. \tag{34}
\end{aligned}$$

The full forward model is used to compute values of  $I^{*L_0}$  and  $\mathbf{K}^{*L_0}$  for upper and lower sidebands of each channel, corresponding to observations made at a fixed set of tangent pressures (denoted by  $L_0$ ), based on a representative state  $\mathbf{x}^*$ . In the v5 processing, the Taylor series in (34) is computed, followed by an interpolation of the resulting radiance profile to the tangent pressures given in the state vector. In addition, the pretabulated  $\mathbf{K}^*$ s are combined by sideband, and also interpolated to the state vector tangent pressures. While the radiances are interpolated using a cubic spline [de Boor, 1987], the weighting function matrix values are interpolated linearly.

The accuracy of the linear approximation is dependent on the proximity of the state  $\mathbf{x}$  to the representative state

$\mathbf{x}^*$ . Accordingly, representative values of  $\mathbf{x}^*$  have been chosen for several ‘bins’. The divisions of the bins are the ‘UARS months’ (10 per calendar year); 8 latitude regions with boundaries at the equator,  $\pm 20^\circ$ ,  $\pm 40^\circ$ , and  $\pm 60^\circ$ ; and the ascending and descending sides of the orbit (needed to account for Doppler shift effects). The values for species abundances and temperatures in the  $\mathbf{x}^*$  vector for each bin are taken from appropriate climatology.

One indicator of the accuracy of the linear forward model is optical depth  $\tau_{\text{ch}}^L$ , which is estimated for each sideband from calculated radiances using

$$\tau_{\text{ch}}^L \simeq -\ln \left[ 1 - \frac{\hat{I}_{\text{ch}}^L}{T^L} \right], \quad (35)$$

where  $T^L$  is the atmospheric temperature at the tangent point. Section 4 discusses the use of the optical depth in more detail.

For most of the MLS observations the linear forward model is a very good approximation, particularly for bands 2–4, where the signals are never optically thick in the stratosphere. It is also a good approximation for band 1, despite the very optically-thick nature of these signals, because temperature and pressure—not mixing ratio—are retrieved from the band 1 radiances. Temperature has a fairly linear effect on the signals, and the nonlinear effects of tangent pressure are described well by the cubic spline interpolation in tangent pressure performed by the linear forward model.

Bands 5 and 6 show significant nonlinear effects, as the channels become increasingly optically thick closer to the line center, and lower in the atmosphere. Radiances in channels close to the line centers are ignored when the optical depth in either sideband is greater than a certain threshold. However, information can still be obtained from other channels at the same tangent altitudes, as the linear forward model is still applicable to channels further out. In the lower stratosphere however, the only information comes from the ‘wing’ channels, and the full nonlinear model has to be used. For reasons of efficiency, only the radiances are computed with the nonlinear model; derivatives of radiance with respect to mixing ratio are still obtained from the linear model.

In the case of the retrieval of UTH, the system is sufficiently nonlinear that the full forward model computation, with derivatives, is implemented [Read et al., 2001]. However, as only a single channel is used, over a small vertical range, the computational effort required is not prohibitive.

### 3.8. The ‘scan residual’ model

In addition to radiance information, the estimates of limb tangent point altitudes (based on the MLS antenna position encoder readouts and the spacecraft attitude determination

system) are used as additional measurements in the retrieval of temperature, pressure, and geopotential height. The forward model for these measurements consists of a hydrostatic calculation of the tangent point altitudes, based on the values of tangent point pressure, temperature and the geopotential height of a reference pressure surface, taken from the state vector. Complexities in this approach arise from the variations in the Earth’s gravitational field with altitude and latitude.

Expressing the problem in terms of geopotential rather than geometric height simplifies the calculations. The hydrostatic calculation assumes that the atmosphere is an ideal gas with the gas ‘constant’ described as a function of  $\zeta = -\log_{10} [\text{pressure} / \text{hPa}]$  by

$$R(\zeta) = \frac{R_0}{M}, \quad (36)$$

where  $R_0$  is the ideal gas constant and the molecular mass

$$M = 0.0289644 \cos(0.2[\zeta' - \zeta_c]), \quad (37)$$

with  $\zeta' = \max(\zeta, \zeta_c)$ , and  $\zeta_c = 2.5$ , describes a fit to the US Standard Atmosphere. An exact integration is used, although for simplicity linear variation in  $RT$  across each layer is assumed, (as opposed to linear variation in  $T$  with the variation in  $R$  described in full by (36)). The same integration is used to produce vertical profiles of geopotential height from the reference geopotential height and temperature information that are the geopotential height product.

There are many well-documented expressions for converting geometric height to geopotential height [Tscherning, 1984]. V5 uses the expression

$$H_\infty = \frac{GM}{g_0 h_c} \left[ 1 - J_2 P_2(\lambda_c) \left( \frac{a}{h_c} \right)^2 - J_4 P_4(\lambda_c) \left( \frac{a}{h_c} \right)^4 \right] + \frac{\omega^2 h_c^2 \cos^2(\lambda_c)}{2g_0}, \quad (38)$$

with

$$P_2(\lambda_c) = \frac{1}{2} (3 \sin^2(\lambda_c) - 1)$$

$$P_4(\lambda_c) = \frac{1}{8} (35 \sin^4(\lambda_c) - 30 \sin^2(\lambda_c) + 3),$$

where  $a = 6378137.0 \text{ m}$  is the assumed Earth major axis,  $\omega = 7.292115 \times 10^{-5} \text{ s}^{-1}$  is the Earth angular velocity, and  $GM = 3.986005 \times 10^{14} \text{ m}^3 \text{ s}^{-2}$ .  $J_2 = 1.0826256 \times 10^{-3}$ , and  $J_4 = -2.3709122 \times 10^{-5}$  are polynomial terms,  $\lambda_c$  is geocentric latitude,  $h_c$  is geocentric altitude, and  $g_0 = 9.80665 \text{ ms}^{-2}$  is the nominal gravitational acceleration. The

geodetic altitude reported by the UARS orbit/attitude software is simply the difference between the geocentric altitude and local Earth radius, not the ‘proper’ geodetic altitude. Its conversion back to geocentric altitude therefore makes the same approximation.

A further subtlety arises from the effects of refraction on the limb rays. The tangent point pressure terms in the state vector describe the pressure at the true tangent points, whereas the level 1 tangent point altitudes that are input to the level 2 processing refer to fictitious rays not affected by refraction. The effects of refraction are taken into account and the altitudes modified according to

$$h_c = \frac{h_c^u}{1+n}, \quad (39)$$

where  $n$  is given by (14).

The effects of refraction make the mathematical construction of the retrieval problem a little more complex. If we still wish to consider the system in terms of geopotential height, a conceptual problem arises, as the ‘measurements’ of refracted geopotential height are dependent on the contents of the state vector (through the temperature and water vapor terms in (14)). Typically, only the forward model estimates are affected by changes in state vector values. To avoid this (merely semantic) issue, we reformulate the problem in terms of a ‘scan residual’, defined as the difference between the geometric and hydrostatic geopotential heights. The ‘measurements’ of this quantity are defined to be zero, with appropriate precisions based on estimates of pointing uncertainty. In addition to computing this residual quantity, the model also computes its derivative with respect to temperature, tangent pressure and reference geopotential height for use in the retrieval algorithm.

Including the scan model requires that the retrieval algorithms not use virtual measurements for tangent pressure, as these would be based on the same tangent height information being used by the scan model. It can be shown that the scan model provides enough information to ensure that the matrix in (5) can be inverted.

These calculations also form the basis of a ‘first guess’ solver for the tangent pressure. This takes the a priori temperature and reference geopotential height and computes an initial guess for the tangent pressure, based on the observed tangent point geometric altitudes.

### 3.9. An empirical model for mesospheric temperature.

The v5 algorithms implement an approximate retrieval of mesospheric temperature, using an empirically-derived forward model for the saturated (sometimes described as ‘blacked out’) radiances at the lowest tangent point in each

scan from the three center channels (7–9) in the 63-GHz radiometer. The value of a saturated radiance reflects the temperature in the region of the mesosphere where the saturation occurs. However, the altitude of saturation (i.e., where the temperature weighting function peaks) is strongly affected by the geomagnetic field, due to the Zeeman splitting of the 63-GHz O<sub>2</sub> emission. A full polarized forward model calculation was too computationally expensive to implement for v5. Instead an empirical model was developed.

Study has shown that the temperature weighting function mimics the shape of the radiance derivative with respect to tangent height, offset by ~8 km. This can be used to obtain an empirical estimate of the weighting function directly from the limb radiances observed within each scan. First, the radiance derivatives with respect to tangent height are computed in the normal scan range (0–90 km). Above this range a linear decay to zero at 120 km is assumed. Secondly, the derivative profile is normalized and shifted down by ~8 km, giving an approximate weighting function. Thirdly, weighting functions are multiplied by the state vector temperature profile to determine the model saturated radiances. No mesospheric retrieval is attempted if the scan does not reach 80 km.

Using this empirical model, v5 produces estimates of mesospheric temperature that represent an improvement on the a priori information (based on the UARS zonal mean climatology). The empirical approach is not, however, applicable to unsaturated radiances, as these are more strongly influenced by the Zeeman effect.

## 4. Implementation of algorithms

This section describes in detail how the algorithms discussed in Section 3 are implemented in the UARS MLS version 5 data processing. There are several configurations of the algorithms, corresponding to different operational modes of the MLS instrument. The changes in instrument operation that necessitated configuration modifications were the April 1993 failure of the 183-GHz radiometer and the cessation in June 1997 of 63-GHz radiometer observations (to conserve spacecraft power). This section first describes factors common to all configurations, and then discusses specific details of each configuration.

### 4.1. V5 products and data files

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<sub>3</sub> 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 tropo-

spheric H<sub>2</sub>O observations were not affected by the 183-GHz failure. The stratospheric H<sub>2</sub>O values (pressures of 100 hPa or less) for the post-April 1993 period are set a priori and should not be used in scientific study. Sulfur dioxide data were produced by the MLS v4 algorithms, but are not in v5, because of the similarity between the spectra of sulfur dioxide and methyl cyanide.

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 can be found in *Burke and Lungu* [1996] (available from the MLS web site <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.

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.

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 ( $\chi^2$  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 UTH software and is discussed in section 9.

#### 4.2. Sources of a priori data

Each element of the state vector is initialized before the retrieval with a priori information. This includes not only the retrieved species such as temperature, ozone, etc., but also contaminating species such as N<sub>2</sub>O. Furthermore, for all the retrieved species, except tangent pressure (for reasons described above), this a priori value is also used as a virtual measurement, as described in section 3.2. The a priori

data for most species come from the UARS standard climatology. Note that this is the climatology constructed before launch for UARS data processing, not the climatology later constructed using UARS data (the UARS Reference Atmosphere Project, [*Wang et al.*, 1999, for example]). In the case of water vapor, the climatology has been modified in the manner described in *Pumphrey et al.* [1998].

The temperatures in the state vector are initialized with data from the National Centers for Environmental Prediction (NCEP, formerly NMC) Global Data Assimilation System (GDAS) Stratospheric Analysis data. These data are typically available up to  $\sim 0.3$  hPa, though occasionally they are missing at lower altitudes, or for a complete day. Where NCEP data are not available, data from the UARS climatology are used. The 100-hPa geopotential height is also initialized with NCEP data if available, with the UARS climatology as a fall back.

#### 4.3. Retrieval ranges and a priori covariances

Table 1 describes the vertical range over which each species was retrieved and gives the values used for the a priori error estimates. The latter were chosen based on lenient estimates of the amounts of variability to be expected in the atmosphere and an examination of the results of many test runs of the data processing algorithms.

The choice of 20 km for most products for the correlation length scale in Equation 7, described in section 3.2 (strictly speaking this is a length in pressure space) deserves some explanation. Firstly, this is not a smoothing length or vertical resolution that is directly imposed on the retrieval. It is a parameter applied to the a priori covariance matrix that makes vertically-correlated solutions more attractive. The correlation length was initially chosen based on studies of the ClO product, which has a fairly poor signal-to-noise ratio. A 20-km length was found to yield ClO profiles with suitable signal to noise for daily mapped products, with vertical resolution of 4–5 km in the lower stratosphere (see section 13).

Tests showed that the choice of correlation length had little impact on retrievals of other species, except for temperature and upper tropospheric humidity. Therefore, for simplicity, 20 km was chosen as the length scale for all the remaining species. However, this length scale was later found to have some undesirable side effects. In particular, as described in section 6.1, the retrieval overestimates the random uncertainty in the data.

#### 4.4. Data from launch to June 14, 1997

For the processing of data from launch to June 14, 1997, the configuration of the v5 algorithms is essentially the same

**Table 1.** Retrieval ranges and a priori information for all the v5 data products.

Species	Vertical range <sup>a</sup>	A priori source	A priori precision	Correlation length
Temperature	68–0.0001 hPa	NMC data, with UARS climatology stacked above	10–46 K	~5 km
Upper Tropospheric Humidity	464–146 hPa	50% global RH <sub>i</sub>	150% RH <sub>i</sub>	~3 km
Stratospheric water vapor (H <sub>2</sub> O)	100–0.00046 hPa	UARS climatology	20 ppmv	~20 km
183-GHz Ozone (O <sub>3</sub> )	100–0.00046 hPa	UARS climatology	20 ppmv	~20 km
205-GHz Ozone (O <sub>3</sub> )	100–0.00046 hPa	UARS climatology	20 ppmv	~20 km
Nitric acid (HNO <sub>3</sub> )	100–0.46 hPa	UARS climatology	10 ppbv	~20 km
Chlorine monoxide (ClO)	100–0.46 hPa	UARS climatology	4 ppbv	~20 km
Methyl cyanide (CH <sub>3</sub> CN)	100–0.14 hPa	UARS climatology	500 pptv	~20 km

<sup>a</sup> This is the range for which data are retrieved, not the range over which they are considered useful, nor the range over which data are output.

for each day processed. The only major change in is that necessitated by the April 1993 failure of the 183-GHz radiometer, described below.

**4.4.1. 63-GHz retrievals** The first phase consists of a retrieval of temperature, tangent point pressure and 100-hPa geopotential height, using the radiances from the 63-GHz radiometer and the estimates of tangent point altitude obtained by the Level 1 processing.

This phase implements three forward models. The first is the linearized model, used for all radiances from channels 1–6 and 10–15. This includes emissions from <sup>16</sup>O<sub>2</sub>, <sup>18</sup>O<sup>16</sup>O, and <sup>17</sup>O<sup>16</sup>O. Continuum emissions from N<sub>2</sub> and H<sub>2</sub>O contribute negligibly to total emission in this band and are not included. The O<sub>2</sub> volume mixing ratio used is 0.2095 from the surface to 80 km with a linear decrease to 0.1447 at 110 km. The 18 and 17 isotopic forms have mixing ratios scaled by  $4.07 \times 10^{-3}$  and  $7.5 \times 10^{-4}$ , respectively.

The scan model described in section 3.8 is used. A 100 m precision is assumed for the input residuals. The empirical mesospheric model described in section 3.9 is also used for the radiances from the lowest tangent height minor frame for channels 7–9. A Gauss-Newton iterative retrieval with a maximum of four iterations is used.

As described in section 3.6, in addition to temperature, tangent pressure and reference geopotential height, a minor-frame-dependent radiance baseline is retrieved. This is different from the other bands, where scan-independent baselines are retrieved along with atmospheric extinction profiles. This method was chosen as it led to better radiance fits than the extinction approach.

**4.4.2. Upper tropospheric humidity retrievals** The retrieval of upper tropospheric humidity consists of three phases, the first two of which are performed immediately following the retrieval of temperature, tangent pressure and geopotential height. The first is a retrieval of a layer mean humidity from 464 to 147 hPa that provides an initial guess

for the UTH profile retrieval. The second phase is a retrieval of humidity on the four UARS surfaces from 464 to 147 hPa. The forward model for the UTH retrieval includes emissions from dry and moist continua and O<sub>3</sub>, HNO<sub>3</sub>, and N<sub>2</sub>O, taken from the state vector. In the first and second UTH retrieval phases, the O<sub>3</sub> and HNO<sub>3</sub> concentrations are given by a priori. Once better estimates of these have been obtained from later retrieval phases, a third UTH retrieval phase is performed to improve the accuracy of the UTH data.

The UTH retrieval is split in this manner because it was found that having good estimates of UTH improved the quality of the retrievals of other species in the lower stratosphere. Thus the first two phases are designed to obtain an interim estimate of UTH, sufficient for use in the stratospheric retrievals.

The UTH retrieval is generally similar to that of version 4.9 (v4.9) [Read et al., 2001]. The main differences are:

1. V4.9 software took temperature from the NCEP dataset, on the original NCEP pressure grid. V5 uses the state vector temperatures, which are NCEP data interpolated to the UARS pressure levels, with UARS climatology substituted in regions where NCEP data are unavailable. V4.9 did not perform retrievals where NCEP data were unavailable.
2. The off-diagonal terms in the a priori covariance matrix had a Gaussian form in v4.9, as opposed to the exponential form given in Equation (7).
3. V4.9 used a different form for the wet and dry continua from that used by v5. This is discussed in section 9.
4. The v4.9 retrieval constrained O<sub>3</sub> and HNO<sub>3</sub> values to version 4 data. V5 uses the retrieved v5 values of O<sub>3</sub> and HNO<sub>3</sub>, as described above.

5. V4.9 retrieved UTH only when there were four or more radiance observations at tangent pressures greater than 80 hPa. V5 undertakes retrievals whenever any radiances meet this criterion.

**4.4.3. 183-GHz retrievals** The next phase is the retrieval of stratospheric water vapor and 183-GHz ozone from the two 183-GHz bands. Band 5 (channels 60–75) is centered on the 183.310-GHz H<sub>2</sub>O line and band 6 (channels 76–90) on the 184.377-GHz O<sub>3</sub> line. Channel 69, close to the center of the H<sub>2</sub>O line, is not used because of unexplained systematic biases seen in the radiances, thought to be instrumental artifacts. Also, channel 76, on the ozone line wing, is not used as its radiances are unreliable. For the center 9 channels of each band, radiances are only used if the tangent point pressure is smaller than 100 hPa and the optical depth in each sideband is less than 1.0. For these channels, the linear forward model is used to estimate radiances and weighting functions. For the sets of three channels at either end of each band (excluding channel 76), a nonlinear forward model is used to compute radiances (weighting functions are still obtained from the linear forward model, and therefore do not change with each iteration). In these channels all radiances whose tangent point pressures are smaller than 100 hPa are used.

The forward model for both bands 5 and 6 includes emissions from H<sub>2</sub>O, O<sub>3</sub>, dry air continuum, and extinction. The dry continuum function is the same as that used for the UTH retrieval with an empirical frequency adjustment based on N<sub>2</sub> collision-induced absorption data [Read et al., 2001]. In addition to retrieving stratospheric water vapor and 183-GHz ozone, spectrally-flat extinction coefficients and baselines are retrieved independently for bands 5 and 6, as described in section 3.6.

The forward models take the temperature, pressure and upper tropospheric humidity from the results of earlier phases. As described in section 3.3, the effects of uncertainty in retrieved temperature and tangent pressure are propagated into the band 5 and 6 radiance error budgets. For speed, off-diagonal terms in the covariance matrices are ignored for all radiances having tangent pressures smaller than 22 hPa. An iterative retrieval based on the Marquardt-Levenberg method is used.

This phase is not invoked for data obtained after the April 1993 failure of the 183-GHz radiometer. The stratospheric water vapor and 183-GHz ozone elements of the state vector for these later days are therefore unchanged from their a priori values.

**4.4.4. 205-GHz retrievals** The 205-GHz radiometer data (bands 2–4, channels 16–60) are used to retrieve 205-GHz ozone, chlorine monoxide, nitric acid, and methyl

cyanide. In addition to these species, baseline and extinction are retrieved jointly for the overlapping bands 2 and 3, with a separate baseline and extinction retrieved for band 4. All the radiance observations in each band with tangent pressures less than 150 hPa are used in the retrieval. As this system is very linear, only a single iteration is performed.

The forward model includes the following species for bands 2 and 3: <sup>35</sup>ClO, HNO<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, SO<sub>2</sub>, O<sub>3</sub>, <sup>18</sup>O<sub>2</sub>, H<sub>2</sub><sup>18</sup>O, N<sub>2</sub>O, O<sub>3</sub>(ν<sub>2</sub>), CH<sub>3</sub>CN, wet continuum, dry continuum, baseline and spectrally flat extinction. The band 4 forward model includes emissions from O<sub>3</sub>, HNO<sub>3</sub>, SO<sub>2</sub>, O<sup>18</sup>OO, N<sub>2</sub>O, HO<sub>2</sub>, O<sub>3</sub>(ν<sub>2</sub>), wet continuum, dry continuum, spectrally flat extinction and baseline. A priori values are used for species not retrieved. Note that this includes species that are related to retrieved quantities, such as ozone isotopes and excited states. This is a departure from version 4, in which such species were appropriately constrained to the retrieved value of the ‘parent’ species.

In earlier versions of the MLS software, SO<sub>2</sub> was retrieved from these bands [Read et al., 1993]. However, the algorithms retrieved small but unrealistic SO<sub>2</sub> mixing ratios in the atmosphere at times when SO<sub>2</sub> abundance was not enhanced by volcanic injections. Studies showed that the band 2 and 3 radiances exhibited a persistent spectral feature, similar to SO<sub>2</sub>, that accounted for this bias. This spectral feature is now known to be emission from CH<sub>3</sub>CN [Livesey et al., 2001]. It was decided in v5 to retrieve CH<sub>3</sub>CN instead of SO<sub>2</sub>, constraining SO<sub>2</sub> to a climatological field. Their spectral similarity in the UARS MLS bands makes simultaneous retrieval of SO<sub>2</sub> and CH<sub>3</sub>CN inappropriate.

As in the previous phase, the effects of uncertainty in retrieved temperature and tangent pressure were propagated into the error budget, with full propagation for radiances in bands 2 and 4 with tangent pressures greater than 22 hPa, and diagonal propagation elsewhere. In addition the uncertainty in stratospheric H<sub>2</sub>O (taken from the previous phase, or a priori following the 183-GHz failure) was also accounted for.

Note that the forward model and retrieval consider emission from the <sup>35</sup>ClO isotope. Before output, the retrieved mixing ratios are scaled by 1.32 to produce the combined abundance of the <sup>35</sup>ClO and <sup>37</sup>ClO isotopes. This phase is followed by the final UTH retrieval phase, as described above.

#### 4.5. Data from 15 June 1997 onward

From June 1997, in order to conserve spacecraft power, the instrument was operated with only the 205-GHz radiometer. The data from this period are processed in v5

by implementing a single-phase retrieval of all the 205-GHz products as described above, with the addition of limb tangent pressure, for which information is obtained from the width of the 206-GHz O<sub>3</sub> line. The temperature is constrained to a priori. In addition to the bands 2, 3 and 4 radiance measurements above 150 hPa, the tangent point altitudes are also used, with the scan model described in section 3.8. No constrained quantity error propagation is needed. A nonlinear iterative retrieval is used with the Marquardt-Levenberg methodology.

This is followed by a two-phase UTH retrieval, identical to the first and third UTH phases in the ‘standard’ processing.

## 5. Proper use of MLS data

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 3.4, 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.

The appropriate parameter files (Level 3PT or 3PL) should always be used in conjunction with MLS Level 3A data. These contain information for each Level 3AT/3AL profile. The MMAF\_STAT field is a single-character flag that indicates the status of the instrument, as it impacts each profile, according to Table 2. 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 3. Only profiles with QUALITY\_... = 4 should be used. In the case of nitric acid, QUALITY\_O3\_205 should be consulted, with QUALITY\_CLO used for methyl cyanide.

In addition to the information available from the data files, the MLS science team has inspected of the quality of the v5 dataset on a UARS-monthly basis. The study involves examination of timeseries 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 page at <http://mls.jpl.nasa.gov/>.

**Table 2.** The 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 data at pressures greater than 22 hPa.
T	Temperatures missing from NCEP 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.

**Table 3.** The 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.

## 6. Validation and characterization issues common to all species

### 6.1. Precision versus scatter

The Level 3AT and Level 3AL files contain, for all data points, uncertainty values. These are the square roots of the corresponding diagonal elements of the solution covariance matrix from Equation (6). These describe a combination of the projection of the radiance uncertainty into state space and the assumed a priori uncertainty.

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 being essentially constant (they are zonal mean or single profile data for all the fields except temperature and geopotential height, for which NCEP data are used). The size of the precision ‘overestimate’ is determined by both the diagonal terms in the a priori covariance matrix that describe the confidence in the a priori data, and the off-diagonal terms that lead to the preference for smoother solutions. The latter factor had a significant effect in v5.

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 quoted for each species reported in later sections of this paper.

## 6.2. Vertical resolution

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 (9). 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 September 17, 1992, which is typical of the dataset.

## 6.3. Accuracy of retrieval estimates

The accuracies of the data vary from species to species, and are described quantitatively in later sections. Sources of uncertainties in accuracy include:

- Uncertainties in spectroscopic parameters,
- Uncertainties in instrument calibration,
- Uncertainties in spacecraft attitude,
- Accuracy of, and biasing toward, a priori information.

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 datasets, or with a priori information (e.g., knowledge that nighttime lower stratospheric ClO abundances are negligible except in certain situations).

## 6.4. Effects of the cessation of 63-GHz radiometer observations

As stated in section 4.5, after June 1997 MLS was operated without the 63-GHz radiometer. The v5 algorithms were modified for data taken after June 1997. The impact this modification had on the data was assessed by running these modified algorithms on data from selected days earlier in the mission and comparing the results with the ‘standard’ v5 data. This was done for observations on January 4, January 29, March 16, April 25, June 12, July 5, August

20, September 27, October 23, and December 13, all 1996. These results are given in later sections describing individual species.

Although the use of a different retrieval configuration resulted in slight changes in behavior for  $\chi^2$  quantities, the criteria for setting values for flags such as QUALITY\_03\_205 were not changed between the two implementations of the algorithm. This results in less-cautious quality control (i.e., fewer profiles are rejected) for the post-June 1997 data.

## 6.5. Retrieved tangent pressure

As described in section 3.6, one of the most important components of the state vector for v5, apart from the ‘products’, is the set of tangent point pressures for each minor frame of observation. Figure 1 shows a summary of the estimated tangent pressure precision (from Equation (6)). It is clear that the precision of v5 tangent pressure is significantly improved over the v4 data. This is due to the introduction of the scan model (section 3.8). Also, it is interesting to note the change of behavior resulting from the switch to observations from the 205-GHz radiometer alone (section 6.4). The 63-GHz observations yield information mainly on the state of the upper stratosphere and lower mesosphere, whereas the 206-GHz ozone signal, being the emission from a weaker line, conveys useful tangent pressure information down to the lower stratosphere.

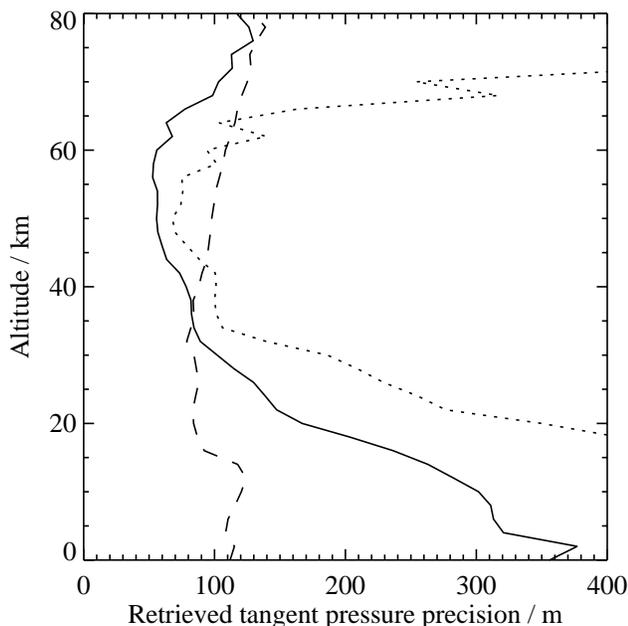
As described in section 4 these estimated precisions are carried forward into the error budgets for the retrievals of the atmospheric species.

## 7. Temperature

### 7.1. Changes in algorithms for v5 Temperature

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 4.2). The data at pressures of 68, 46 hPa, and less than 0.46 hPa are not scientifically useful, because of the poor MLS temperature sensitivity in these regions.

The MLS version 4 (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 3.2–0.0001 hPa (linearly changing with log pressure). Since the v4 algorithms re-



**Figure 1.** Typical precision for retrieved tangent pressure. The solid line shows the mean estimated precision in v5 retrieved tangent pressure on June 14, 1997 (converted into approximate meters using a 16 km  $\log_{10}$  scale height). The dotted line shows the same information for v4. The broken line indicates the same statistic for the v5 retrieval of data taken on July 14, 1997, a day when only the 205 GHz radiometer was activated.

**Table 4.** v5/v4 and v5/v3 Temperature Differences.

Pressure / hPa	Global	v5–v4 / K		v5–v3 Global
		Tropics	Polar winter	
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

trieved 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.

## 7.2. Comparison of v5, v4 and v3 temperatures

In the stratosphere, the v5 temperatures are generally warmer (by 1–3 K) than v4, but v5 is cooler than v4 (by  $\sim 1$  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 4 shows v5/v4 and v5/v3 differences, based on the first year of observations. The largest v5/v4 differences are in polar winter, where planetary wave activity is strong.

## 7.3. Estimated precision and accuracy of v5 temperatures

The estimated precision, accuracy and resolution (as defined in section 6.2) of v5 temperatures are given in Table 5. Precisions ( $1\sigma$ ) 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.

Accuracy is estimated from the error analysis described in Fishbein et al. [1996]. One observed artifact is a systematic error of  $\sim 0.5$  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 may cause serious problems for studies of short-period atmospheric waves. In the v5 temperature this artifact is reduced by about a half

from  $\sim 1$  K seen in v4, but users should be cautious about temperature variations near or below 0.5 K.

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. Mesospheric Temperature

As discussed in section 3.9 the v5 algorithm employs an empirical forward model to obtain information on mesospheric temperature (at pressures  $< 0.32$  hPa). Table 6 summarizes the estimated vertical resolution and single profile precision for the mesospheric temperature. The significant contributions of the a priori information to the mesospheric temperatures necessitate a different measure of the precision of the dataset from that used for other products. An approximate measure  $s_m$  can be obtained from

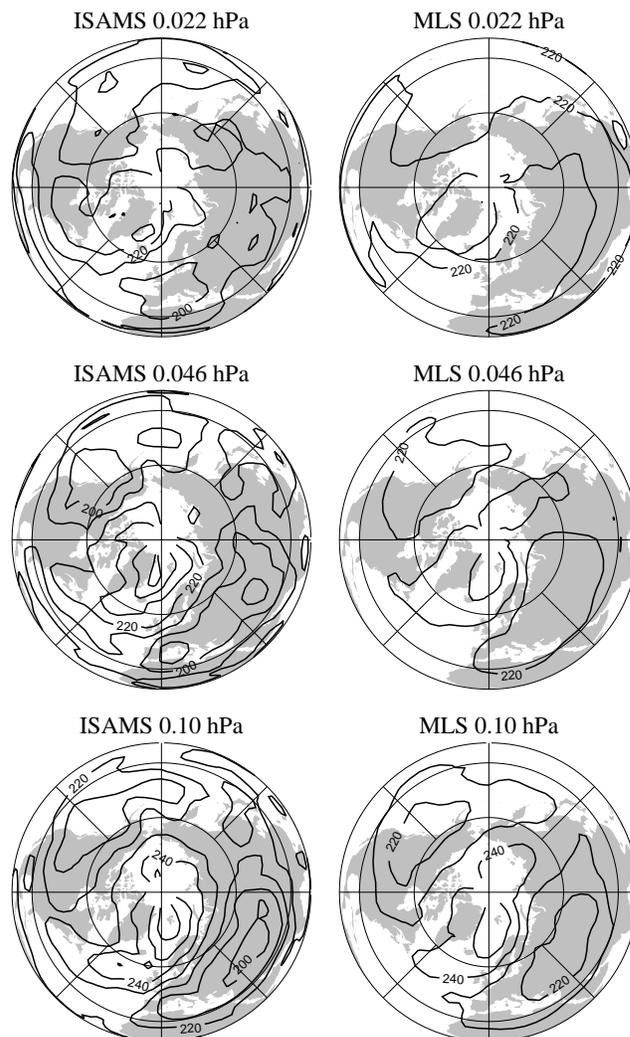
$$\frac{1}{s_m^2} = \frac{1}{s_x^2} - \frac{1}{s_a^2}, \quad (40)$$

where  $s_x$  is the precision quoted in the Level 3AT files, and  $s_a$  is the a priori precision.  $s_m$  is an approximate measure of the precision of the information MLS contributed to the retrieval system. The overall accuracy of these data remains to be assessed.

Table 6 also gives differences between v5 and the Improved Stratospheric and Mesospheric Sounder (ISAMS) v12 temperature observations during December 4, 1991 and January 14, 1992. The preliminary comparisons show that MLS temperatures are generally warmer than those from ISAMS at these altitudes. The warm bias increases with height from  $\sim 1$  K at 0.32 hPa to  $\sim 10$  K at 0.01 hPa. Comparisons of ISAMS data with Lidar observations [Dudhia and Livesey, 1996] indicate that there is good agreement between MLS and Lidar at 0.3–0.03 hPa.

Figure 2 compares MLS and ISAMS northern hemisphere temperature on January 8, 1992, when planetary waves were strong in the mesosphere. Both measurements reveal consistent patterns of the planetary disturbance, but MLS wave amplitudes are significantly weaker than those observed by ISAMS. This is probably due to the poorer sensitivity and vertical resolution of the MLS temperature data at these altitudes.

These data remain as a research product, in need of further validation before they can be confidently used in scientific studies. One reason for this is that there exists a large discontinuity in the v5 temperature sensitivity across  $\sim 0.4$  hPa, which can cause a problem in retrievals near this



**Figure 2.** ISAMS and MLS mesospheric temperature on January 8, 1992. Contour intervals are 10 K, and latitude circles are shown at the equator, 30°N and 60°N.

level. This discontinuity may not be evident in individual temperature profiles or averages. However, temperature perturbations can exhibit a sudden drop in amplitude, sometimes by as much as a factor of two, for pressures less than  $\sim 0.4$  hPa. This discontinuity is intrinsic to the retrieval configuration that aims to merge two differently-resolved temperature measurements. Scientists wishing to use MLS temperature data above 0.46 hPa are strongly advised to consult the MLS science team before embarking on scientific studies.

**Table 5.** Estimated vertical resolution, precision and accuracy of v5 temperature.

Pressure / hPa	Vertical resolution <sup>a</sup> / km	Typical precision / K	Precision ratio <sup>b</sup>	Estimated accuracy <sup>c</sup> / K	v5–NCEP / K
0.46	5.0	3.3	0.7	5	−1.5
0.68	7.0	2.1	0.5	5	+5.2
1.0	6.5	1.8	0.5	5	+8.6
1.5	7.0	1.7	0.5	5	+6.6
2.2	6.5	1.5	0.5	4	+4.3
3.2	6.5	1.5	0.5	4	+1.4
4.6	6.0	1.4	0.5	5	+0.9
6.8	6.5	1.4	0.5	4	+0.1
10	6.5	1.3	0.4	4	+1.1
15	6.0	1.2	0.4	4	+1.1
22	7.0	0.8	0.3	4	+1.6
32	6.5	0.9	0.3	6	+2.0

<sup>a</sup> As defined in section 6.2.

<sup>b</sup> Data file uncertainties should be multiplied by these numbers to obtain a better value for the ‘1 $\sigma$ ’ single profile precision (see text).

<sup>c</sup> Accuracies quoted here roughly represent a 95% confidence level (‘2 $\sigma$ ’ values).

**Table 6.** Characterization of MLS v5 mesospheric temperature.

Pressure / hPa	Vertical resolution <sup>a</sup> / km	Estimated precision <sup>b</sup> / K	MLS contribution <sup>c</sup> / K	v5–ISAMS / K
0.010	20	27	40	+9.7
0.015	20	26	44	+8.8
0.022	25	25	50	+7.1
0.032	25	24	55	+6.5
0.046	30	23	63	+5.5
0.068	20	22	57	+5.8
0.10	15	21	52	+3.4
0.15	10	19	48	+0.3
0.22	10	17	35	−0.4
0.32	10	15	29	+1.1

<sup>a</sup> As defined in section 6.2.

<sup>b</sup> Typical values given in Level 3AT files.

<sup>c</sup> See text.

### 7.5. Caveats in use of v5 temperature

- Only temperature data for pressures between 36–0.46 hPa should be used in scientific study.
- As stated in section 5, only data with MMAF\_STAT='G', 't', or 'T', QUALITY\_TEMP = '4', and positive uncertainties should be used.
- Temperature data following the deactivation of the 63-GHz radiometer (June 1997) should not be used.

## 8. Geopotential Height

Version 5 is the first MLS algorithm to produce geopotential height (GPH) as a standard output. 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 3.8, 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 (36)) and the retrieved temperature profile.

### 8.1. Accuracy and precision of GPH data

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.

The 100 hPa GPH precision depends mostly on knowledge of the MLS pointing. Random pointing errors are assumed 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  $\sim 26$  tangent points, the precision is expected to be better than the single-pointing precision.

Accuracy is harder to assess, as it is totally dependent on knowledge of UARS attitude, the uncertainty of which is hard to characterize. However, comparisons with correlative datasets can yield some insight into the accuracy of MLS GPH. Figure 3 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  $\sim 16.5$  km,

while the MLS values vary over 1 km and occasionally 2–3 km. The differences provide an estimate of MLS GPH accuracy. 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 Day 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.

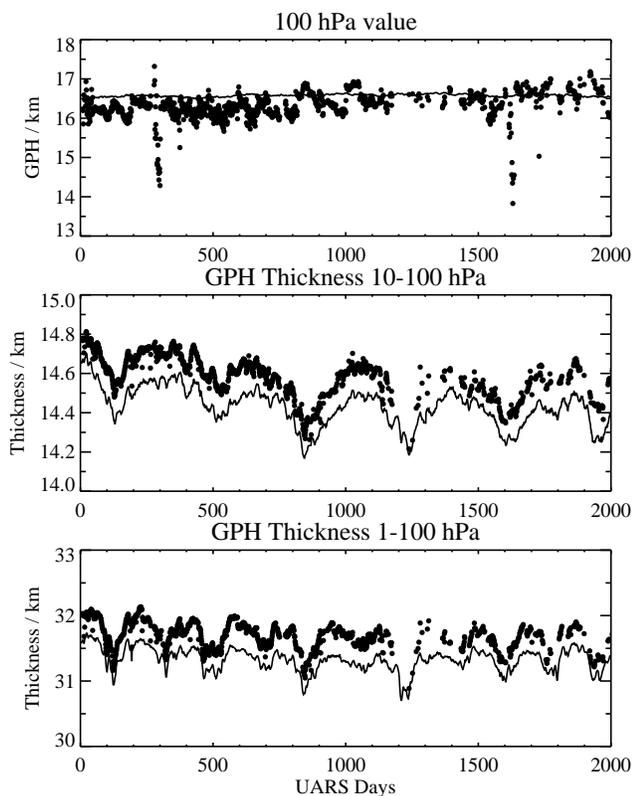
The estimated single-profile GPH precisions ( $1\sigma$ ) vary from 70 m at 100 hPa to 220 m at 0.01 hPa, based on the variability of MLS GPH measurements between 20°S–20°N from October 1991 to September 1992. The MLS GPH precision is much better than its accuracy, as is shown by the good tracking between the NCEP and MLS layer thicknesses in Figure 3. 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 mostly due to temperature differences between the two data sets.

### 8.2. Caveats for using GPH data

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 3 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 values here. GPH data after June 1997 should not be used as 63-GHz observations were not made during this period.

## 9. Upper tropospheric humidity

A full description of the MLS observations of upper tropospheric humidity (UTH) is given in *Read et al.* [2001]. This paper concentrates 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 to be measured from space using 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<sub>2</sub>O continuum function was derived by assuming the upper brightness boundary was in an atmosphere having a relative humidity of 100% with respect to ice (%RH<sub>i</sub>) and with no significant emissions from cirrus ice. The v5 water vapor continuum function used humidity measurements from Vaisala radiosonde measurements that



**Figure 3.** 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.

were coincident with MLS observations. Following the production of v5 data, the accuracy of Vaisala radiosonde observations of the uppermost troposphere has been 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 %RH<sub>i</sub>) over the poles during winter than had been seen in previous years which could be an artifact of the data processing.

### 9.1. Changes from UTH v4.9 to v5

The most significant change between v4.9 and v5 UTH was the manner in which the wet and dry spectroscopic continua were estimated. V4.9 estimated these from the MLS radiances alone. The dry continuum was obtained by assuming that the smallest MLS radiances on a few selected days correspond to 0%RH<sub>i</sub>. An appropriate pressure-squared function was fitted to the observed radiance profiles (allowing for the small contributions from minor species). The v4.9 wet continuum was similarly obtained, by assuming that the largest radiances corresponded to 100%RH<sub>i</sub>, and fitting an appropriate continuum function (again, accounting for the minor species and dry continuum contributions). The temperature and tangent pressures used in these fits were from the MLS version 4 dataset.

In v5 the dry and wet continua were obtained simultaneously from a larger sample of selected profiles. As in v4.9, the dry continuum was obtained by assuming the smallest radiances corresponded to 0%RH<sub>i</sub>. The v5 wet continuum, however, was obtained by using UTH profiles from coincident Vaisala radiosonde measurements. The MLS/sonde coincidence criteria applied for the fit were  $\pm 1^\circ$  longitude,  $\pm 1^\circ$  latitude, and  $\pm 3$  hours. The v5 UTH data produced using these fitted continua agree with the Vaisala radiosondes within 5% on average between 300–150 hPa.

V5 produces negative Level 3AT uncertainties where the estimated error is greater than 75%RH<sub>i</sub> or the observed radiances are detected as being contaminated by cloud scattering. The v4.9 Level 3AT data give negative uncertainties only when a retrieval is not performed (the ASCII Level 2 UTH file simply reports that no retrieval was attempted). These situations arise when too few good tropospheric radiances are observed in a scan.

The time values given in the Level 2 UTH files differ between v4.9 and v5. The v4.9 files quote time in UT hours, while v5 uses UT milliseconds. Also, the v5 times are quoted as being 32.786 s later than those in v4.9 (to be consistent with other MLS products).

Note that the v5 H<sub>2</sub>O files include data at pressures greater than 464 hPa, which should not be used in scientific investigations. The relative humidity at these levels is assumed to be the same as at 464 hPa, and a conversion to an H<sub>2</sub>O vmr is performed, using the temperature data in the state vector (essentially NCEP data).

## 9.2. Estimated precision, accuracy, and resolution for v5 UTH

The estimates for v5 UTH precision, accuracy and resolution are given in Table 7. The basis for these values is the same as that for v4.9 [Read et al., 2001]. Vertical resolution for 215 and 316-hPa levels are the full width at half maximum of the columns of the averaging kernel matrix, computed for the nominal MLS scan and radiance uncertainty. Outside the 464–147 hPa range, the forward model assumes constant relative humidity (constrained above to the 147 hPa and below to the 464 hPa values). This makes vertical resolution harder to define. We have chosen as a measure the distance between the lower (147 hPa) or upper (464 hPa) half maximum of the averaging kernel and the half maximum of the instrument weighting functions above (147 hPa) or below (464 hPa). The vertical resolution degrades with increasing moisture. The precisions in Table 7 account for the effects of radiance noise (0.1 K), tangent pressure precision (250 m), temperature uncertainty (2 K), and minor species corrections (0.4 ppmv for O<sub>3</sub>, 1.5 ppbv for HNO<sub>3</sub>, and 15 ppbv for N<sub>2</sub>O) projected onto the UTH profile.

The accuracy estimate is based on the mean residual of the radiance fit to the dry and wet continuum functions, described above, propagated into UTH space. The uncertainties given in the UTH data files are estimates of accuracy, not precision. The unusual latitude-dependent artifact of 8 ppmv observed in the v4.9 dataset [Read et al., 2001] is also observed in the v5 dataset. Therefore, any variability of a few ppmv seen in MLS UTH data may not reflect actual atmospheric conditions. As described in Read et al. [2001], care should be exercised when trying to ‘join’ the MLS UTH dataset to stratospheric H<sub>2</sub>O observations (e.g., from MLS), especially where the 147 hPa level is in the stratosphere. The values in Table 7 only apply to v5 UTH for data taken before June 16, 1997.

## 9.3. UTH after June 15, 1997

Table 8 shows the impact of the cessation of 63-GHz observations after June 1997, by comparing ‘standard’ data from earlier in the mission with data for the same days retrieved using the ‘no 63-GHz’ configuration.

The comparison shows significant differences between the two configurations, especially at high latitudes, and at 147 hPa globally. Since the ‘standard’ v5 data are a few %RH<sub>i</sub> drier than the v4.9 data, which are in turn mostly drier than correlative data [Read et al., 2001], the post-June-1997 v5 product—being drier still—probably underestimates the true humidity. However, the scatter plots shown in Figure 4 indicate that the morphology of the data is reasonable, as the scatter is reasonably tight, compared to the precision of either dataset, except at 147 hPa. The large scatter at 147 hPa is due to the strong sensitivity at this level to the retrieved tangent pressure, which typically show differences between the two configurations equivalent to ~200 m.

## 9.4. Caveats for v5 UTH data

The following caveats apply to the use of v5 MLS UTH data (note that these are different from those that apply to v4.9 data [Read et al., 2001]). The data at 147 and 215 hPa should only be used where the corresponding uncertainty in the data file is positive. Any UTH values greater than 125 %RH<sub>i</sub> are indicative of the presence of clouds or supersaturation. In the absence of ancillary data to detect clouds, such values should be reset to 100 %RH<sub>i</sub> for a better estimate of the true humidity. Note that this threshold value applies only to the data taken before June 1997; an equivalent threshold for the post-June-1997 data remains to be determined.

The 147 and 215 hPa level are thought to be reliable when the UTH uncertainty is positive. The quality of UTH at 464 and 316 hPa is crucially dependent on the humidity above these levels. In cases where the atmosphere is very moist in the 215–147 hPa region, MLS will have little or no sensitivity to the water vapor lower down. As part of the UTH retrieval, a simple retrieval of mean humidity over 464–147 hPa is performed. The results of this ‘initial guess’ prove to be a useful indicator of moist situations. Study has shown that MLS UTH data at 316 and 464 hPa are reliable when their values are greater than the ‘initial guess’ (quoted in the Level 2 UTH file) minus 5 %RH<sub>i</sub>, and the magnitude of their uncertainties (regardless of sign) is less than 110 %RH<sub>i</sub>. Even with this screen, data at the 464 and 316 hPa levels are expected to have a dry bias for humidities greater than 50 %RH<sub>i</sub>. However, information about the atmospheric morphology in these regions should still be useful.

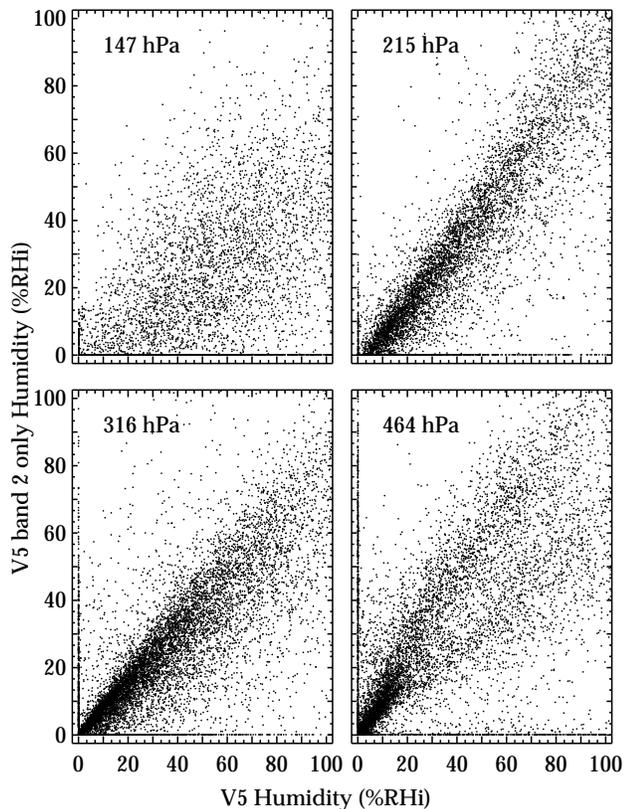
Care should be taken when converting the MLS data from

**Table 7.** Estimated resolution, precision, and accuracy of MLS v5 UTH.

Pressure / hPa	Typical vertical resolution / km	Global		30°S–30°N		30°–60°(S and N)		60°–81°(S and N)	
		Precision / %RH <sub>i</sub>	Accuracy / %RH <sub>i</sub>						
147	3–4	24	26	37	39	11	11	30	27
215	3	11	25	17	23	5	24	21	33
316	3	9	25	7	23	10	24	14	38
464	3–6	20	52	22	64	26	50	21	45

**Table 8.** Differences between ‘205-GHz only’ and standard v5 UTH for several 1996 days.

Pressure / hPa	Global		30°S–30°N		30°–60°N		60°–81°N	
	/ %RH <sub>i</sub>	/ %						
147	–23	–52	–56	–50	–4	–60	–9	–74
215	–4	–13	0	0	–6	–34	–6	–62
316	–7	–20	–4	–17	–7	–13	–14	–29
464	0	0	3	22	0	0	–7	–17

**Figure 4.** A comparison of UTH data taken over ten selected days given in section 6.4, processed with the ‘standard’ processing (x-axis) and the ‘R2 only’ mode (y-axis).

relative humidity to mixing ratio. As discussed in more detail in *Read et al.* [2001], the temperature data used for the conversion should be that planned for use in subsequent scientific analyses, rather than the NCEP/UARS climatology data used in the retrieval, as the latter could introduce biases.

## 10. Ozone from 205-GHz Radiometer Data

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.

Information on v3 O3\_205 data is in the MLS ozone validation paper [*Froidevaux et al.*, 1996] and in *Cunnold et al.* [1996a,b]. MLS v4 data quality and related studies have been presented in *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 (<http://mls.jpl.nasa.gov>) and distributed by the GSFC DAAC.

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 datasets.

### 10.1. Changes in Algorithms for v5 205-GHz Ozone

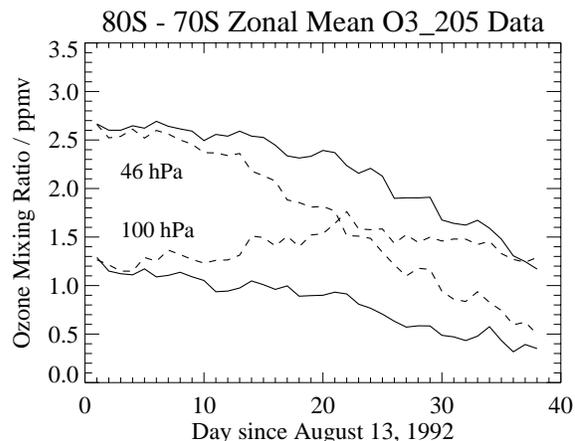
The main change in v5 O3\_205 is the use of a finer retrieval grid (see Introduction) below 0.1 hPa. While the finer retrieval grid can lead to better vertical discrimination, it also generally leads to somewhat poorer precision. Except at 100 hPa where the precision is better than v4, v5 stratospheric 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.

The v5 algorithm for setting the value of the quality flag ‘QUALITY\_O3\_205’ (which helps to screen bad data) was modified from that of v4 because of changes in the  $\chi^2$  statistic describing the fit to the radiances. The v5  $\chi^2$  statistic for this band is somewhat less correlated with anomalies in the retrieved O3\_205 values than in v4. More records in v5 data have been assigned quality flag values less than 4 (indicating bad radiances or bad radiance fits) than in v4. Except for unusual months, this parameter, together with the other general criteria for selecting ‘good’ O3\_205 retrievals, discards about 2% of v5 data, compared to about 1% for 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 the v5 data, even though the retrieved profiles do not appear obviously bad.

A factor that seems to contribute to the increased number of poor quality profiles is that v5 retrievals use radiances down to 150 hPa, which can lead to more contamination by clouds, especially in the tropics. Indeed, we find that the spatial distribution of flagged (poor quality) profiles 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.

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

Table 9 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 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



**Figure 5.** Zonal mean (80°S to 70°S) ozone changes during the August 14 to September 20, 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.

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.

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. The overall agreement in ‘trends’ between MLS and SAGE II data (discussed by *Cunnold et al.* [2000]) should remain essentially unchanged (within the error bars) for the main period of MLS measurements; some additional discussion of temporal changes in MLS/SAGE differences is given below, but long-term trends are not the main focus here.

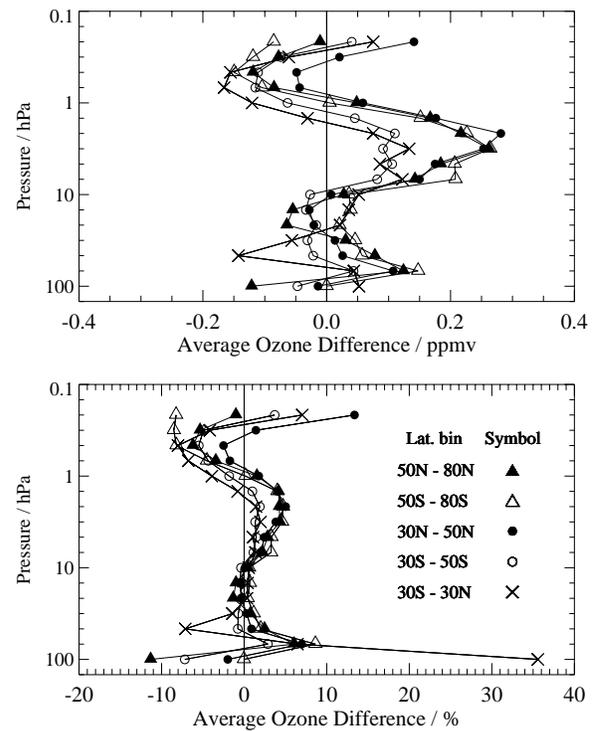
The vertical profile of the v5 ozone rate of decrease during Antarctic ozone hole conditions is different from v4. Figure 5 shows the zonal mean ozone changes for 80°S to 70°S at 46 and 100 hPa for the time period from August 14 to September 20, 1992. While the sum of the mixing ratios at these two levels 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). The south polar winter ozone variations shown here for 1992 are very similar to those measured by MLS for the 70°S to 60°S latitude bin at 100 hPa, but the 46 hPa total ozone decrease in the latter latitude range is about a factor of two smaller. 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 essentially 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.

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 each other better).

### 10.3. Validation of v5 205-GHz Ozone

**10.3.1. Comparison of 205-GHz Ozone Data with Other Datasets** We now discuss how the v5 MLS O3\_205 data compare with a few other ozone datasets, 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, 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 6. These years have much smaller potential impact from the Mt. Pinatubo vol-



**Figure 6.** 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).

**Table 9.** Average differences between 03\_205 data versions.

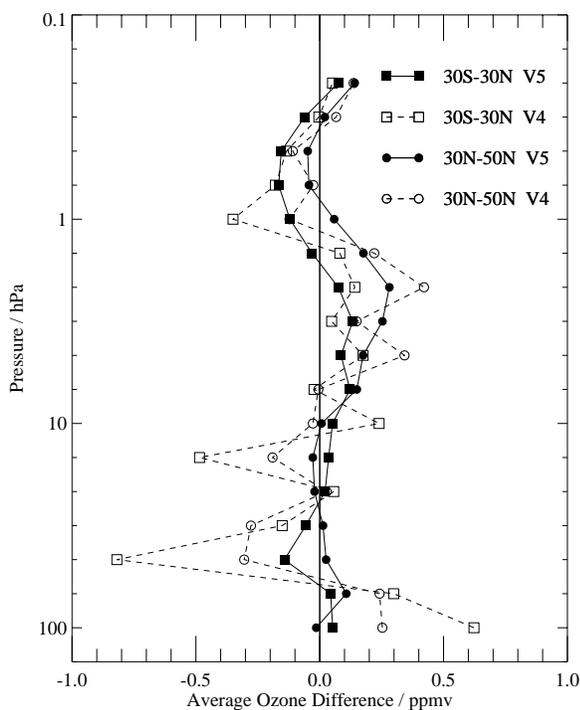
Pressure / hPa	Global <sup>a</sup>		v5/v4 Differences Tropical <sup>b</sup>		Midlatitude <sup>c</sup>		v5/v3 Differences Global	
	/ ppmv	/ %	/ ppmv	/ %	/ ppmv	/ %	/ ppmv	/ %
0.46	0.0	0	-0.05	-3	0.0	0	-0.03	-2
1.0	+0.2	+7	+0.3	+10	+0.2	+6	+0.1	+3
2.2	-0.1	-2	-0.1	-2	-0.1	-2	-0.2	-3
4.6	-0.1	-1	0.0	0	-0.1	-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

<sup>a</sup> Based on ~400,000 profiles from all latitudes for the first full year of data (October 91 through September

92).

<sup>b</sup> Based on ~60,000 profiles from 10°S to 10°N for the first full year of data.

<sup>c</sup> Based on ~25,000 profiles from 35° to 45°N and 35° to 45°S for the first full year of data.

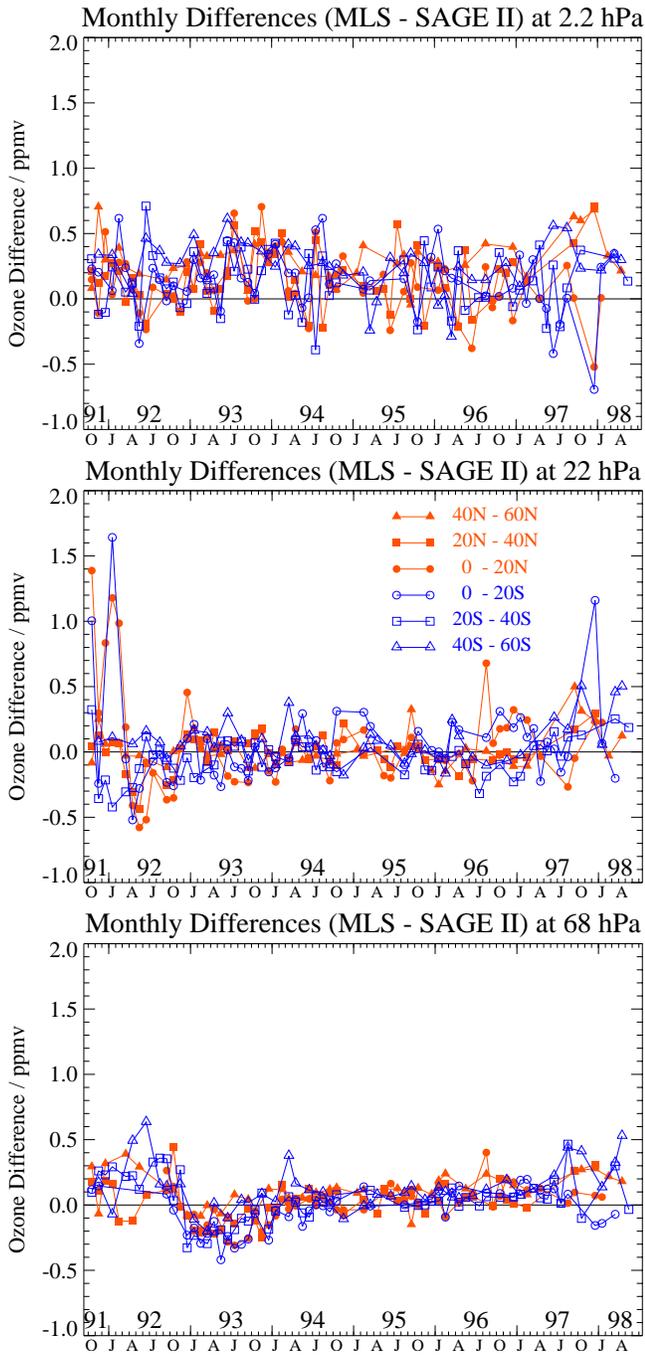


**Figure 7.** Average ozone differences (ppmv) for both MLS 03\_205 v5 and v4 retrievals versus coincident SAGE II version 6.1 profiles over two latitude ranges (see legend) during the 1995–1996 time period. The discrepancies that occurred primarily because of poorer v4 MLS data quality in the lower stratosphere, especially at low latitudes, have been largely removed by using v5 MLS data. Abundance differences are calculated as MLS minus SAGE II values.

canic 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 mid- to upper stratosphere (per a recommendation by R. Wang, private communication, 2001).

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 6 for 1995 through 1996). It is difficult to collect enough independent data in the tropics to categorically ascertain the relative merits of SAGE II and MLS lower stratospheric profiles compared to ozonesondes (the most reliable tropical data source having a significant number of ozone profiles). The MLS v5 profiles agree significantly better than v4, both in comparison to SAGE II profiles and to ozonesonde datasets (see below). MLS v4 differences with SAGE II coincident profiles are compared to the differences for v5 (during 1995–1996) in Figure 7. V5 shows a significant reduction in the average difference with SAGE II, particularly at low latitudes in the lower stratosphere. In general, v5 mid- 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].

At pressures less than 1 hPa, percentage differences be-

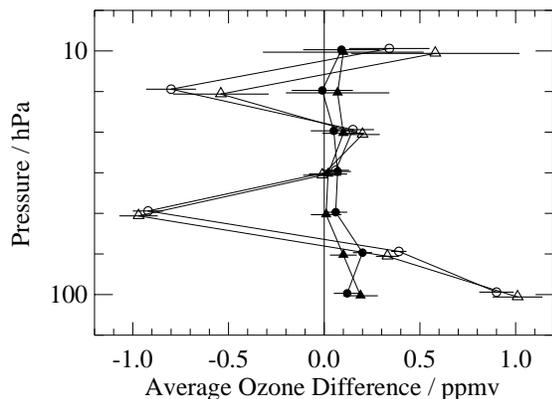


**Plate 1.** Time series of monthly mean differences between MLS v5 and SAGE II version 6.1 coincident ozone profiles; differences are calculated as MLS minus SAGE mean values. Six 20° wide latitude bins are shown, see legend in center panel; pressure levels are 2.2 hPa (top panel), 22 hPa (center panel) and 68 hPa (bottom panel).

tween the two datasets increase; this altitude range includes diurnal variability issues along the occultation ray path for SAGE II observations, so we are not surprised to see larger differences there (where we are not comparing quite the same quantities and accurate modeling would be needed to reduce this source of uncertainty). The total number of coincidences during 1995–1996 is about 650 for the high latitude bins and 1200 at low latitudes, but these numbers drop significantly for the lowest pressures reported for SAGE II data (0.1 hPa); also, the uncertainty for both measurements is larger at these pressures. However, these factors alone probably can not fully account for the observed 10 to 20% differences (or more) in average lower mesospheric profiles; these differences can change sign depending on whether SAGE II sunsets or sunrises are used, but no clear pattern emerges from the studies performed so far. We have illustrated in Figure 7 the MLS and SAGE II comparisons up to 0.22 hPa only, but we see no strong reason to degrade the MLS accuracy estimates for the lower mesosphere, compared to the upper stratosphere. With sufficient averaging of the MLS profiles, there is probably useful information at pressures less than 0.22 hPa, but we conservatively use 0.22 hPa as an upper limit.

Plate 1 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 Mt. 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 Plate 1 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).

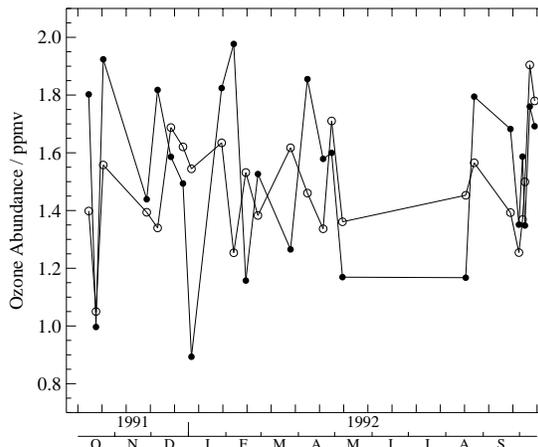
There are also significant improvements (over v4) in the agreement between v5 MLS profiles and tropical ozonesonde data from Ascension Island and Brazzaville. Figure 8 shows much smaller differences between MLS v5 and these ozonesonde averages (typically less than 0.1 ppmv for the average of about 25 total available coincidences for



**Figure 8.** Average differences between MLS 03\_205 retrievals and coincident ozonesonde profiles from the tropical sites Ascension Island (circles) and Brazzaville (triangles) for late 1991 and 1992; open symbols are for v4 and closed symbols are for v5. Differences are MLS minus ozonesonde values. There were 25 matching pairs of coincident profiles for Ascension Island and 24 for Brazzaville. Error bars give the standard errors for these average differences.

late 1991 through 1992) than for v4. V3 data, not shown here, are also in poorer overall agreement 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 no longer true for Ascension Island, as shown in Figure 9, nor for Brazzaville (not shown here). The MLS v5 ozone values between 46 and 10 hPa are larger than tropical ozonesonde values by  $2 \pm 2\%$ , well within the expected combined accuracies (of order 5%).

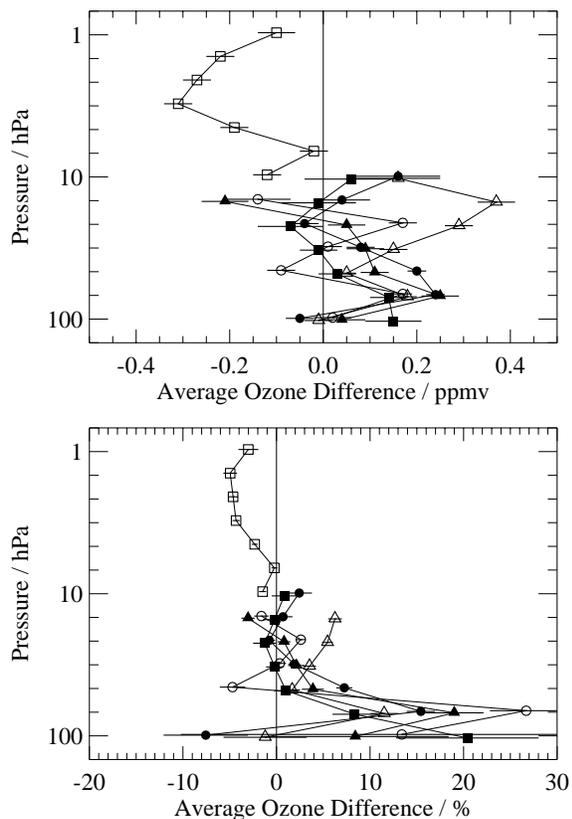
One possible source of differences between SAGE and MLS profiles at low latitudes is the positive bias introduced in MLS ozone data at 100 hPa by the presence of dense cloud. Not all the profiles affected by clouds have been flagged as ‘bad’ by the v5 software, and the amount of bias introduced by cloud remains to be 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 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



**Figure 9.** Time series comparison of 46 hPa ozone abundances from ozonesonde data (open circles) taken above Ascension Island in 1991 and 1992 with coincident (within  $2^\circ$  latitude,  $12^\circ$  longitude, same day) MLS v5 03\_205 values (dots). Estimated precision is about 0.25 ppmv for MLS values and better than 0.1 ppmv for the sonde data.

years of data) indicate average differences of about 5% or less for pressures less than or equal to 46 hPa, as shown in Figure 10. In the lower stratosphere, where mixing ratios can be small, the typical average differences between MLS and sondes are  $\sim 0.25$  ppmv or less for 68 hPa, and less than 0.15 ppmv for 100 hPa.

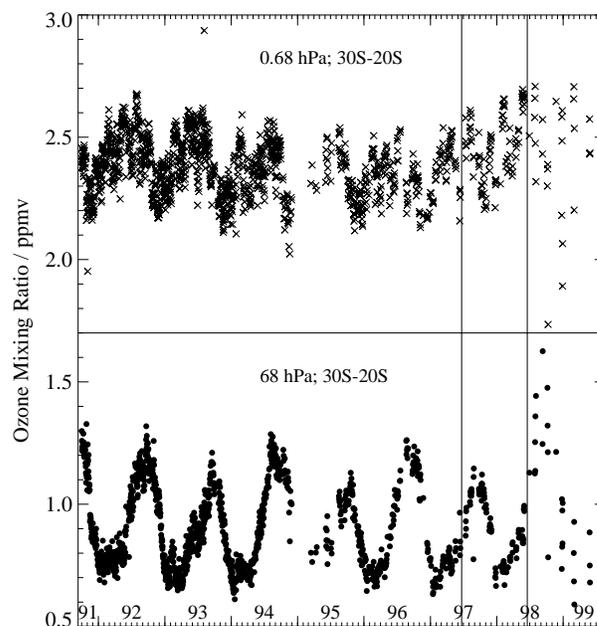
Based on the totality of the above MLS ozone comparisons with SAGE II and ozonesondes, we find a small positive bias (2 to 4%) in the v5 03\_205 data for mid- to upper stratospheric regions. This offset is within the combined absolute errors and is therefore of marginal significance. The Table Mountain 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 give excellent agreement (not shown here for brevity) over seasonal ozone variations of up to a factor of two. The remaining average offset for the mid- 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 ‘ $2\sigma$ ’ 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 accu-



**Figure 10.** Average differences (top panel: ppmv, bottom panel: percent) between MLS v5 03\_205 and various sets of coincident correlative profiles covering about 5 years or more: ozonesonde data from October 1991 through 1996 for Hilo ( $20^{\circ}$  N, 132 matched profiles, open circles), Boulder ( $40^{\circ}$  N, 70 matches, closed triangles), Uccle ( $50^{\circ}$  N, 126 matches, dots), Payerne ( $47^{\circ}$  N, 201 matches, open triangles), and Lauder ( $45^{\circ}$  S, 103 matches, closed squares), and lidar data from October 1991 through April 1996 for Table Mountain ( $34^{\circ}$  N, 289 matches, open squares). Error bars give the standard errors for these average differences. Coincidences were defined as having latitude differences less than  $2.5^{\circ}$  and longitude differences less than  $12^{\circ}$ .

racy estimates are summarized below in section 10.4.

Danilin et al. [2001] used trajectory calculations to increase the number of matches between MLS and other measurements during the northern winter of 1999/2000 (poleward of  $50^{\circ}$  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



**Figure 11.** Zonal mean time series of daily average ozone (205 GHz data) for 1991 through 1999 at  $30^{\circ}$  S to  $20^{\circ}$  S, for pressures of 68 hPa (dots) and 0.68 hPa (crosses). Vertical lines indicate the start of single radiometer mode in June 15, 1997, and the bad scan slip period on June 29, 1998.

Danilin et al. [2001], the MLS values are up to  $\sim 12\%$  larger than those from the Polar Ozone and Aerosol Measurement (POAM) III, and also 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  $\sim 5\%$  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 can be attributed to the MLS results themselves, except at 68 and 100 hPa.

**10.3.2. 205-GHz Ozone after June 15, 1997** The appendix summarizes significant changes in MLS operation for the entire MLS mission. A major change was the cessation of 63-GHz observations, as described in section 6.4. MLS ozone data have been scrutinized for any degradation

or discontinuities that may be tied to this deactivation or subsequent antenna scan slip problems; these slips led to significant loss of MLS data from mid-1998 through 1999. 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 40 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 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 are consistent overall with previous years' data, 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 August 18–25, 2001 data.

The mid-1998 to 1999 time period, when significant loss of data occurred because of scan slips, seems particularly affected in terms of data quality as well. Figure 11 shows that MLS zonal mean ozone data (shown for 30°S to 20°S) during this time period are noisier than in the preceding years for both lower (68 hPa) and upper (0.68 hPa) stratospheric levels. During this time period, the estimated ozone uncertainties show more extreme values (and scatter) as well. While these examples demonstrate some of the more obvious manifestations of poorer data quality because of the significant antenna scan slips during this time period, most other zonal means show smaller degradation.

Based on the loss and degradation of MLS data after mid-1998, we do not recommend using this time period as part of trend analyses, even if the ozone abundances appear reasonable to first-order. The time period from mid-1997 to mid-1998 yields seemingly much better results, but some caution should apply for this period as well. Although fewer profiles are flagged based on poor radiance fits in the single-radiometer mode, changes to the radiance fitting diagnostics should probably also have been made in order to optimize the profile rejection algorithms after mid-1997.

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

Our estimates of 03\_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 '(2 $\sigma$ )' 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 10 gives these v5 accuracies for 03\_205, along with the estimated vertical resolution and typical single-profile precision. These precisions are 1 $\sigma$  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 03\_205 data files should be multiplied by the values given in the fifth column of Table 10 to obtain the best estimate of precision.

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

1. A small positive MLS offset, of order 2 to 4% on average, is observed in average comparisons of v5 03\_205 MLS data versus ozonesonde profiles in the mid-stratosphere and SAGE II values in the mid- 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 mid- 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).
2. The uncertainties in the 03\_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 10 and section 6.1).

#### 10.6. Caveats in Use of v5 O3\_205

1. 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.
2. The Level 3 'parameter files' should be examined and only profiles with MMAF\_STAT = 'G', 't', or 'T' and QUALITY\_03\_205 = 4 should be used for scientific studies. Profiles with MMAF\_STAT set to 'T' (which occurs quite infrequently) may have slight biases at pressures larger than or equal to 46 hPa.
3. Only data having positive estimated uncertainties (the 'quality field' in Level 3A data files) should be used.

**Table 10.** Estimated Vertical Resolution, Precision and Accuracy of v5 03\_205.

Pressure / hPa	Vertical resolution <sup>a</sup> / km	Estimated precision		Precision ratio <sup>b</sup>	Estimated accuracy <sup>c</sup>
		/ ppmv	/ %		
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%

<sup>a</sup> As defined in section 6.2.

<sup>b</sup> Data file uncertainties should be multiplied by these numbers to obtain a better value for the ‘1 $\sigma$ ’ single profile precision (see text).

<sup>c</sup> Accuracies quoted here represent roughly a 95% confidence level (‘2 $\sigma$ ’ values).

- 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.

## 11. Ozone from 183-GHz Radiometer Data

Information on data quality and characteristics of previous versions of 03\_183 is in the work of *Froidevaux et al.* [1996] and *Ricaud et al.* [1996]. The data versions have been described in the MLS ‘Data Quality Documents’ available on the MLS web site (<http://mls.jpl.nasa.gov>). The previous software changes led to a typically small decrease (by less than 5 to 10%) in the v4 mixing ratios, compared to v3 data, whereas the v5 values are now typically increased somewhat over the v4 data. Here, we briefly summarize the changes that occurred for the v5 03\_183 data, and give our estimates of v5 precision and accuracy. This ozone dataset remains the recommended MLS dataset for mesospheric ozone, but the 03\_205 MLS data set is still recommended for the stratosphere. *Pumphrey and Harwood* [1997] have shown that the raw 183-GHz ozone radiances contain useful information and realistic signals 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

The main changes in v5 03\_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 4. 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. This was done using off-line retrievals of these parameters along with 03\_183 from zonal mean radiances [*Pumphrey and Bühler*, 2000].

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

Table 11 shows average differences between the three data versions for MLS 03\_183. V5 03\_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 versus 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 val-

**Table 11.** Average Differences Between 03\_183 Data Versions

Pressure / hPa	Global <sup>a</sup>		v5/v4 Differences Tropical <sup>b</sup>		Midlatitude <sup>c</sup>		v5/v3 Differences Global	
	/ 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

<sup>a</sup> Based on ~400,000 profiles from all latitudes for the first full year of data (Oct. 91 through Sep. 92).

<sup>b</sup> Based on ~60,000 profiles from 10°S to 10°N for the first full year of data.

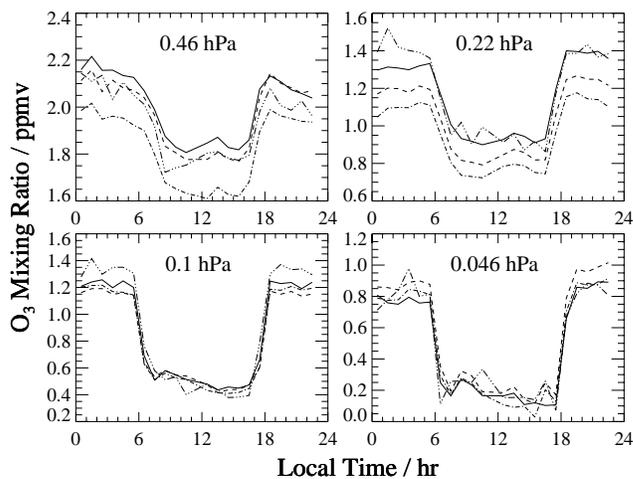
<sup>c</sup> Based on ~25,000 profiles from 35° to 45°N and 35° to 45°S for the first full year of data.

ues (these differences are not shown in the table).

Figure 12 shows all three versions of the 03\_183 northern midlatitude mesospheric ozone diurnal cycle discussed for v3 by Ricaud et al. [1996]. For reference, Figure 12 also displays v5 03\_205, which is noisier than—but in agreement with (within about 10%)—the mesospheric 03\_183 data. Changes from v4 to v5 are larger for pressures greater than 0.1 hPa, mainly because of the finer v5 retrieval grid. The main 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). Ricaud et al. [1996] also showed that ground-based microwave data on mesospheric ozone agreed with the MLS results in the lower mesosphere; ground-based microwave data from Table Mountain (California) were shown to exhibit better agreement with models than MLS data at 0.1 hPa, but somewhat poorer agreement at 0.04 hPa. These conclusions are consistent with the newer v5 MLS data for mesospheric 03\_183.

### 11.3. Validation of v5 183-GHz Ozone

Our comparisons of zonal mean differences show that the v5 03\_183 values between 0.46 and 46 hPa are larger than the 03\_205 values by about 2 to 5%, within the combined estimated accuracies. Comparisons with SAGE II data and MLS 03\_205 profiles are shown in Figure 13, where the MLS 03\_183 zonal average profiles (coincident with SAGE II profiles, using the same criteria as for the 03\_205 validation) in 3 broad latitude bins exhibit higher values than both the MLS 03\_205 and the SAGE II profiles. Since



**Figure 12.** Comparison of the ozone diurnal variation at various mesospheric pressures for different MLS ozone retrieval versions: v5 03\_183 (solid lines), v4 03\_183 (dashed-dotted lines), v3 03\_183 (dashed lines), and v5 03\_205 (dashed-triple-dotted lines). These plots are for hourly averages and 40°N to 50°N zonal means for October 1991, the same conditions as for Figure 2 of Ricaud et al. [1996].

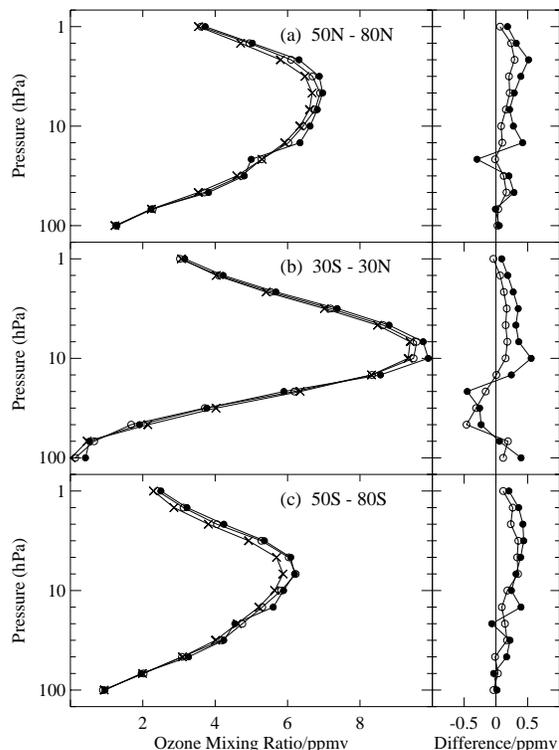
we have shown in section 10.3 that v5 O3\_205 values are on average a few % larger than several other accurate data sets, O3\_183 values are therefore a few % larger yet; this will be reflected in a poorer accuracy for O3\_183 (see section 11.4). While the two MLS ozone retrievals exhibit similar difference patterns from SAGE II, there are some suspicious O3\_183 features: abundances at 100 hPa are overestimated at low latitudes (see panel (b)), and notches in the profiles appear at higher latitudes (see panels (a) and (c)) 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.

The above discussion shows that we cannot recommend the use of O3\_183 data in the lower stratosphere (100 or 68 hPa), and that the quality of this data set is generally somewhat poorer than that of O3\_205 in the stratosphere, even if the two sets of profiles typically track each other very well, especially in the upper stratosphere. Likely reasons for the poorer data quality of O3\_183 profiles include our inability to use one of the wing channels in this band as a result of noisy performance and the poorer knowledge of sideband ratios (calibration) for the 183-GHz radiometer. These issues create limiting factors that the enhanced (iterative) v5 retrievals cannot overcome, despite some improvements for the lower stratospheric behavior in comparison to v4 data, which often had values too low at 46 hPa and a less constant average offset with height compared with SAGE II profiles (even if the v4 offset was smaller).

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  $\sim 5\%$  positive bias; however, relative changes should be represented very well in that region. Average differences (not shown here) near 0.1 hPa of order 20% exist between O3\_183 and SAGE II data (which are typically lower than the MLS data), but there are complications in interpreting the comparisons in that region because of the ozone (diurnal) variations along the ray paths sampled by SAGE II and the fact that the MLS ray paths are typically not close to those for SAGE II.

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

Table 12 gives the v5 O3\_183 estimated vertical resolution, precision and accuracy, obtained in the same manner as for O3\_205. Based on our mid- 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),



**Figure 13.** 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.

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 03\_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

1. MLS 03\_183 has a positive bias, averaging about 5 to 10%, based on comparisons with the v5 03\_205 data, as well as ozonesonde profiles and SAGE II values in the mid- to upper stratosphere.
2. Values are too large at 100 hPa at low latitudes and negative averages occur at 68 hPa.
3. There are also some pervasive notches in the profiles at 22 hPa, at high latitudes in particular.
4. The uncertainties in the 03\_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 12 and section 6.1).

### 11.6. Caveats in Use of v5 183-GHz Ozone

1. 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).
2. The Level 3 ‘parameter files’ should be examined and only profiles with MMAF\_STAT = ‘G’, ‘t’, or ‘T’ and QUALITY\_03\_183 = 4 should be used for scientific studies.
3. Only data having positive estimated uncertainties (the ‘quality field’ in Level 3A data files) should be used.
4. 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.

## 12. Stratospheric and mesospheric water vapor

Version 3 of the MLS stratospheric H<sub>2</sub>O product is described and validated by *Lahoz et al.* [1996]. Since that time,

**Table 13.** Average Differences Between H<sub>2</sub>O Data Versions

Pressure / hPa	v5–v104 / ppmv	v5–v104 / %
0.0100	+0.51	+29%
0.0215	+0.45	+12%
0.0464	+0.14	+2%
0.100	–0.09	–1%
0.147	–0.04	+0%
0.215	+0.01	+0%
0.316	+0.04	+0%
0.464	–0.01	+0%
0.681	–0.02	+0%
1.00	+0.08	+1%
1.47	+0.36	+6%
2.15	+0.22	+4%
3.16	+0.45	+9%
4.64	+0.33	+7%
6.81	+0.60	+13%
10.0	+0.50	+12%
14.7	+0.34	+8%
21.5	+0.30	+7%
31.6	+0.68	+16%
46.4	+0.35	+8%
68.1	+0.44	+12%
100	+0.23	+6%

version 4 has been released, followed by a development prototype known as version 104. 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 version 104 was a much better data set than was version 4 and it has gone on to be used in a number of scientific studies. The validation of version 4 and 104 water vapor is described in *Pumphrey* [1999]. Because version 104 is already validated and has been widely used, this section documents the differences between version 5 and version 104. Some of these differences are discussed in *Pumphrey et al.* [2000].

### 12.1. Changes in algorithms for v5 H<sub>2</sub>O

The relationship between H<sub>2</sub>O mixing ratio and radiance is somewhat nonlinear, so an iterative retrieval is used, as described in section 4.4.3. In addition, the sideband ratios and the pressure broadening and shift parameters for the water vapor line were changed to values retrieved from the measured radiances [*Pumphrey and Bühler*, 2000].

### 12.2. Differences between v5 and v104 H<sub>2</sub>O.

Differences between MLS v5 and v104 H<sub>2</sub>O are shown in Table 13. There is a significant break in the table at 1 hPa – above this the two versions agree to within 0.1 ppmv up to 0.04 hPa while below it, version 5 is about 0.5 ppmv wetter.

**Table 12.** Estimated Vertical Resolution, Precision and Accuracy of v5 O3\_183.

Pressure / hPa	Vertical resolution <sup>a</sup> / km	Estimated precision		Precision ratio <sup>b</sup>	Estimated accuracy <sup>c</sup>
		/ ppmv	/ %		
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%

<sup>a</sup> As defined in section 6.2.

<sup>b</sup> Data file uncertainties should be multiplied by these numbers to obtain a better value for the ‘1 $\sigma$ ’ single profile precision (see text).

<sup>c</sup> Accuracies quoted here represent roughly a 95% confidence level (‘2 $\sigma$ ’ values).

<sup>d</sup> 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.

### 12.3. Estimated vertical resolution, precision and accuracy of v5 H<sub>2</sub>O

Table 14 shows the estimated precision and accuracy of version 5 H<sub>2</sub>O. The precision is calculated as in section 6 from the variability in an equatorial latitude bin, over four 5-day periods in the middle of UARS yaw months. (Variability within those 5-day periods is considered, not variability between them.) The column labeled “Ratio” has the meaning explained in section 6.

The accuracy is estimated by comparisons with other instruments, including HALOE (HALogen Occultation Experiment) and ATMOS (Atmospheric Trace Molecule Spectroscopy Experiment), as described below, and also the ground-based microwave instrument WVMS (Water Vapor Millimeter-wave Spectrometer) [Nedoluha et al., 1997] and a balloon-mounted frost-point hygrometer (FPH). Comparisons of version 104 with FPH and WVMS are described in [Pumphrey, 1999] – the comparisons done here for v5 were carried out in the same manner and were consistent with the HALOE and ATMOS comparisons described below.

The version 5 data in the lower stratosphere contain two types of systematic error which vary on a timescale of days to weeks. The timing of these artifacts coincides with the UARS yaw cycle and with the ends of calendar months. The

first of these effects is caused by changes in the antenna temperature while the second is caused by the a priori, which changes from one calendar month to the next. These effects are illustrated in figure 14.

### 12.4. Correlative comparisons for v5 H<sub>2</sub>O

In this section we compare MLS data to data from two solar occultation instruments: HALOE [Harries et al., 1996] and ATMOS [Gunson et al., 1996]. For each profile used from each of these data sets the difference was taken with the closest MLS profile for the same day. The two profiles are typically separated by less than 15° in longitude and 2° in latitude. The difference was averaged over a number of pairs of profiles to yield a mean difference, which gives an indication of the systematic bias between the two instruments. This is shown in figures 15 and 16 as a solid line. We also calculated a root-mean-square (rms) difference which is shown in figures 15 and 16 as a dotted line. This will equal the absolute value of the mean difference if all of the difference is systematic and exceed it if some of the difference is random. To aid this comparison where the mean difference is negative, the absolute mean difference is shown in the figures as a dashed line. The rms difference should be of a similar size to the root-sum-square combined uncertainties of the two measurements, which is shown in the figures as a dot-dash line.

**Table 14.** Estimated Vertical Resolution, Precision and Accuracy of MLS v5 H<sub>2</sub>O.

Pressure / hPa	Vertical resolution <sup>a</sup> / km	Estimated precision		Precision ratio <sup>b</sup>	Estimated accuracy <sup>c</sup>	
		/ ppmv	/ %		/ ppmv	/ %
0.0100	8.4	0.39	19.3%	0.5	1.03	51.6%
0.0215	6.3	0.31	8.1%	0.5	1.02	26.8%
0.0464	6.1	0.23	4.5%	0.4	1.01	19.4%
0.100	5.7	0.26	4.9%	0.5	1.00	18.4%
0.147	7.5	0.18	3.2%	0.3	0.93	16.4%
0.215	5.3	0.22	3.9%	0.4	0.87	15.3%
0.316	7.0	0.18	3.1%	0.3	0.83	14.4%
0.464	3.5	0.18	3.1%	0.4	0.80	14.0%
0.681	3.7	0.19	3.3%	0.4	0.75	13.1%
1.00	4.3	0.20	3.5%	0.5	0.69	12.1%
1.47	3.7	0.20	3.5%	0.6	0.67	11.6%
2.15	3.3	0.16	3.2%	0.6	0.64	12.7%
3.16	3.2	0.15	3.1%	0.5	0.59	12.0%
4.64	3.1	0.14	3.2%	0.6	0.53	11.6%
6.81	3.1	0.14	3.0%	0.5	0.50	11.0%
10.0	3.1	0.13	2.9%	0.5	0.48	10.9%
14.7	3.2	0.12	3.0%	0.5	0.50	12.2%
21.5	3.4	0.15	3.7%	0.6	0.52	12.8%
31.6	3.4	0.16	3.6%	0.6	0.51	11.2%
46.4	3.7	0.17	4.0%	0.5	0.50	12.1%
68.1	5.3	0.28	7.5%	0.5	0.75	20.0%
100.	5.1	0.25	6.7%	0.2	1.00	27.0%

<sup>a</sup> As defined in section 6.2.

<sup>b</sup> Data file uncertainties should be multiplied by these numbers to obtain a better value for the ‘1 $\sigma$ ’ single profile precision (see text).

<sup>c</sup> Accuracies quoted here represent roughly a 95% confidence level (‘2 $\sigma$ ’ values).

**12.4.1. HALOE** This comparison uses every HALOE profile for which a co-located MLS profile is available. The results are shown in Figure 15. The MLS v5 profile is drier than HALOE in the mesosphere, but wetter than it in the stratosphere. This behavior contrasts somewhat with v104, which is uniformly drier than HALOE. On the whole, the differences between the two instruments are within the quoted errors (the dotted line in Figure 15 is inside the dot-dash line). In the mesosphere the difference is much smaller than the quoted errors: this is because the error bars supplied with the HALOE data are very large.

**12.4.2. ATMOS** ATMOS is a solar occultation instrument which flew on several space shuttle missions of which two occurred during the operational life of the MLS 183-GHz radiometer. We have collected all the profiles from these two missions for which a coincident MLS profile exists and carried out a comparison as described above. The results are shown in Figure 16. The ATMOS data used were version 3, described in *Michelsen et al.* [2002].

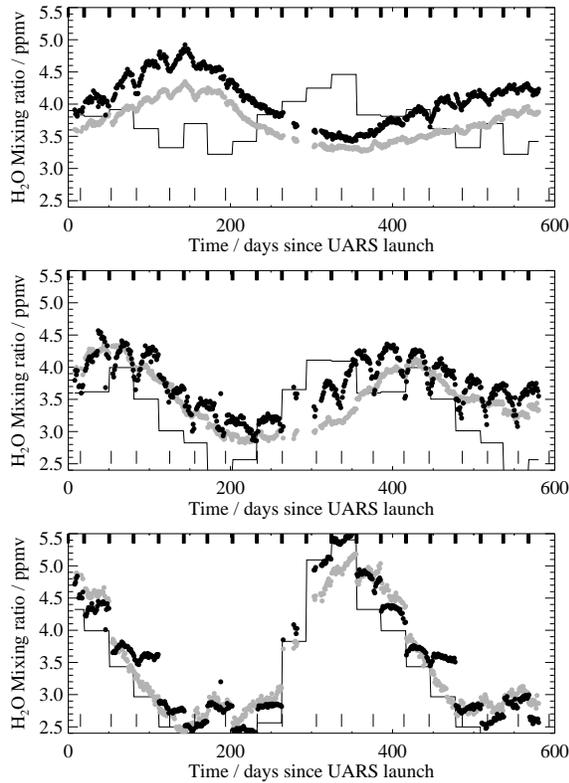
MLS v5 agrees well with ATMOS in the stratosphere but is considerably drier in the mesosphere. The similarity between Figures 15 and 16 suggests that the bias in MLS v5 H<sub>2</sub>O changes rapidly with altitude near 1 hPa. MLS v104, on the other hand, has a fairly uniform dry bias throughout

the stratosphere and lower mesosphere.

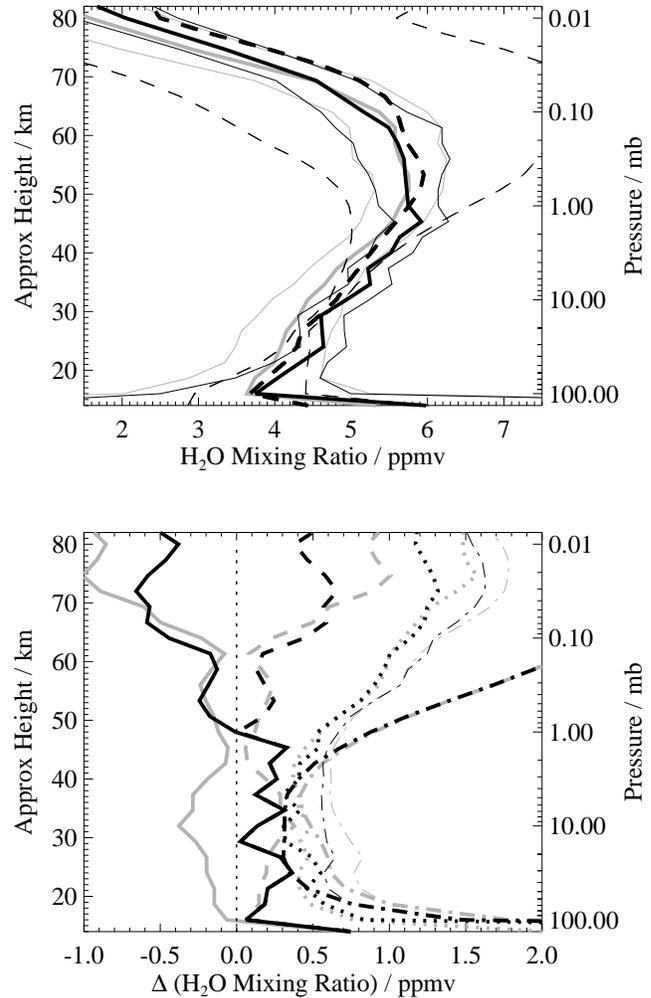
In the light of these comparisons and the systematic errors discussed in section 12.3 we recommend v104 H<sub>2</sub>O be used in preference to v5, especially in the lower stratosphere, taking note of the v104 dry bias of 0.2–0.5 ppmv. In the mesosphere, there is little difference between v104 and v5.

### 13. Chlorine monoxide (ClO)

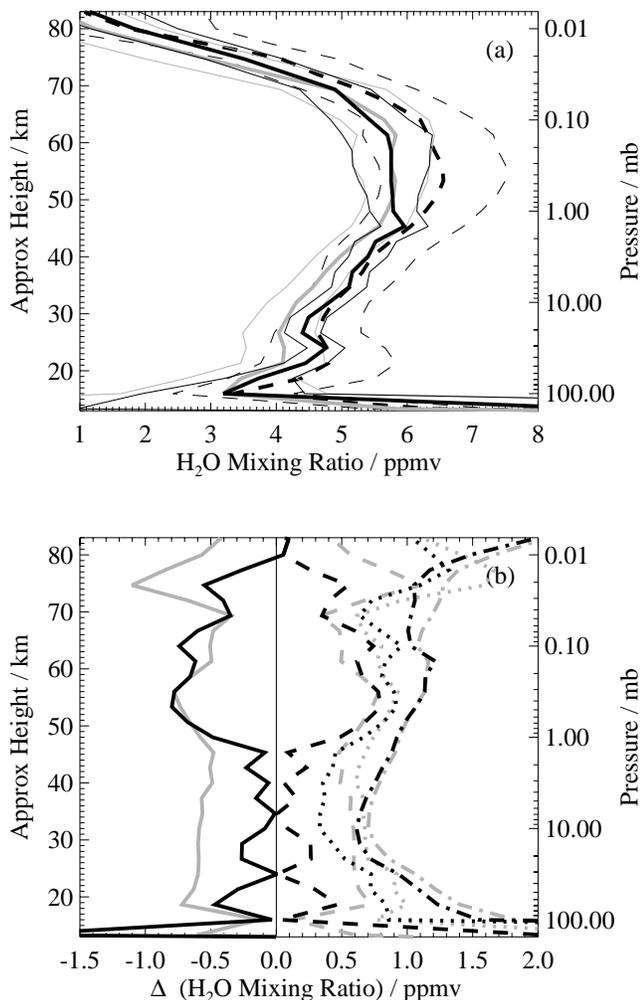
*Waters et al.* [1996], describing validation of MLS v3 ClO data, provides background for the material in this section and a general reference for the MLS ClO measurements. Major changes from v3 to v4 ClO were: (1) correction of the ‘old’ line strength that was inadvertently used in v3 processing [*Waters et al.*, 1996], with the expected 8% lowering of ClO values from v3, and (2) retrieval of HNO<sub>3</sub>, 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 (<http://mls.jpl.nasa.gov>). 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



**Figure 14.** Time series of equatorial MLS H<sub>2</sub>O, at 100 hPa (bottom), 68 hPa (middle) and 46 hPa (top). The black dots are version 5, the grey dots are version 104. The thin line is the a priori used for both versions. The ticks near the bottom of each panel mark the UARS yaws; the ticks near the top of each panel mark calendar month boundaries.



**Figure 15.** Comparison of MLS and HALOE H<sub>2</sub>O. Top panel: mean profiles of MLS v5 (solid) and HALOE v19 (dashed). MLS v104 is shown in grey for comparison. Thin lines are error bars for the thick lines of the same color and dash pattern. Bottom panel: Solid lines show differences between MLS and HALOE v19; dashed lines are the absolute value of this difference. Black lines are MLS v5 and grey are MLS v104. The dotted line is the rms difference between MLS and HALOE and the dot-dash line is the rms of the quoted errors of the two instruments. (The thin dot-dash line is the same as the thick one except that the standard deviation of the HALOE data is used in place of the quoted errors which seem unduly pessimistic in the mesosphere.)



**Figure 16.** As Figure 15 but for MLS and ATMOS H<sub>2</sub>O.

recommended for scientific studies.

Several papers published since *Waters et al.* [1996] further attest the general validity of MLS ClO. *Feist et al.* [2000] found that Millimeter-wave Atmospheric Sounder (MAS) ‘day’ minus ‘night’ ClO measurement agreement with MLS is typically better than 0.1 ppbv in the tropical and midlatitude regions where the two instruments have coincident measurements. *Khosravi et al.* [1998] found that MLS ClO measurements agree well with model results in the upper stratosphere when the model methane is constrained by HALOE measurements and a 6% HCl + O<sub>2</sub> channel for the ClO + OH reaction is included per laboratory measurements of *Lipson et al.* [1997]. *Ricaud et al.* [2000] found that the MLS ClO diurnal variations in the middle and upper stratosphere agree with model calculations to within 5–10%, and that seasonal variations imply an evolution essentially dictated by the variation in partitioning with HCl, together with partitioning within the ClO<sub>x</sub> family above 40 km. The differences found above 50 km are attributable to uncertainties in reaction rates. The analyses of *Froidevaux et al.* [2000] show that the increase in MLS upper stratospheric ClO during 1992–1995 is larger than expected from CFC increases, and is consistent with implications from the decline in CH<sub>4</sub> observed by HALOE during this period as found by *Siskind et al.* [1998]. *Froidevaux et al.* [2000] further show that the decrease in lower stratospheric ClO observed by MLS during 1991–1997 is consistent with relaxation from chemical perturbation induced by the Pinatubo volcanic eruption. *Massie et al.* [2000] show that the variations in 5-day vortex averages of v4 MLS lower stratospheric ClO are consistent with heterogeneous model calculations for the 1995–1996 Arctic early winter. ClO enhancement observed during the Arctic winter of 1996–1997 is qualitatively reproduced in the study of *van den Broek et al.* [2000], but the model underestimates v4 MLS ClO abundances by as much as 1 ppbv. *Danilin et al.* [2000] examine v5 MLS ClO data and find that they generally agree with results from a box model run along air parcel trajectories; however, the model tends to underpredict ClO values slightly. *Waters et al.* [1999] summarize results of additional analyses of the MLS ClO data.

### 13.1. Changes in algorithms for v5 ClO

The major changes for ClO in v5 are because of (1) retrievals on each UARS surface, and (2) retrieving CH<sub>3</sub>CN instead of SO<sub>2</sub>.

Although v5 retrievals are done on each UARS surface (between 100 and 0.46 hPa), the vertical resolution of v5 ClO (see Table 15 later in this section) is approximately the same as for v4 and v3. The version 5 algorithms introduce off-diagonal terms in the a priori ClO covariance matrix to favor smoother profiles (see section 3.2). The additional free

parameters in v5, however, allow better definition of the profile, and the v5 profiles are generally smoother. As in previous versions, the retrieved points should be interpreted as values at the breakpoints of a piecewise-linear representation of the vertical profile.

The CH<sub>3</sub>CN retrievals in v5 allow a better fit of the measured radiances in MLS bands 2 and 3 when there is negligible volcanically-injected SO<sub>2</sub> 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.

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 top panels of Figure 20 in Waters et al. [1996]), although averages of these data exhibit the expected diurnal behavior (more ClO at night).

The algorithm for setting QUALITY\_C1O to ‘4’ (indicating good radiances and retrievals) has been changed in v5 because of changes in the v5 radiance-fit  $\chi^2$  statistic, which is not as highly correlated with ‘spikes’ in retrieved ClO as in v4 or v3. This algorithm was determined empirically by choosing  $\chi^2$  criteria (including bands 2, 3 and 4) that eliminate most unreasonable ‘spikes’ while not throwing out an excessive amount of good data. With the exception of a few months when unusual instrument or satellite problems occurred, the general criteria for selecting good ClO retrievals (QUALITY\_C1O=‘4’, MMAF\_STAT = ‘G’, ‘T’ or ‘t’) discard ~3% of all v5 data compared to ~1.5% for v4. For records with MMAF\_STAT = ‘G’, ‘T’ or ‘t’ (the same in v5 and v4), more records in v5 (~2%) are assigned quality flags less than ‘4’ than in v4 (~0.2%).

As in v4, retrievals of HNO<sub>3</sub> are done in v5 that reduce or eliminate the HNO<sub>3</sub>-caused bias in enhanced polar lower stratospheric v3 ClO described by Waters et al. [1996]. Also, as in v3 and v4, a linear forward model is used in the v5 ClO retrievals; thus the ~10% scaling uncertainty due to lack of radiance ‘closure’, and possible ~5–10% overestimates of lower stratospheric ClO in the cold polar vortices, described in Waters et al. [1996] are not expected to be significantly reduced in v5, although the v5 retrievals on every surface may have somewhat reduced them. The algorithms have no constraints that force retrieved ClO values to be positive; doing so would cause undesirable bias artifacts in averages made from individual profiles. Individual retrieved values will often be negative because of instrument noise. (Note that this

is a different issue than the reduction of negative values in averages of nighttime ClO data mentioned earlier due to retrieval of CH<sub>3</sub>CN in v5.)

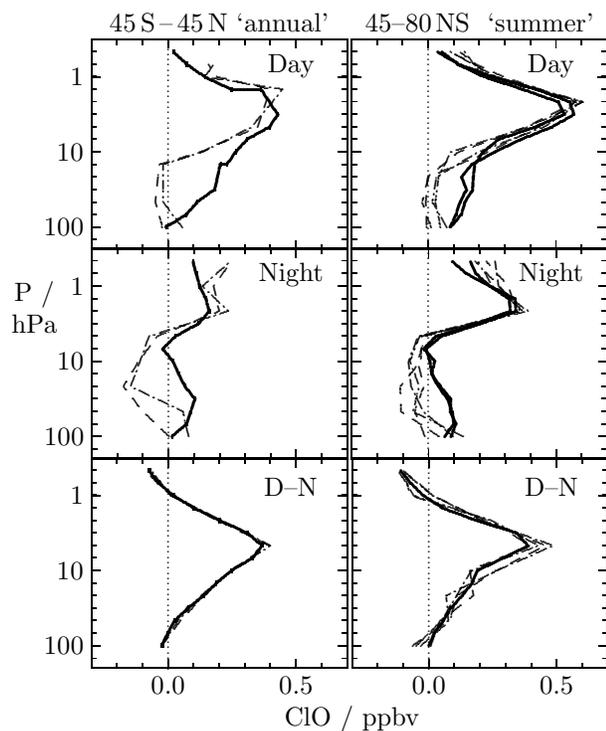
### 13.2. Comparison of v5, v4 and v3 ClO

We compare v5, v4 and v3 ClO for three categories of observations: (1) low and mid latitudes, 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\_C1O=‘4’, MMAF\_STAT = ‘G’, ‘T’ or ‘t’, and positive uncertainties in the data files. All v3 data values shown here for comparison have been multiplied by 0.92 to correct the known line-strength error in v3 data.

**13.2.1. Low to mid latitude annual, and high latitude summer** Figure 17 compares averages of retrievals from 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<sub>3</sub>CN. 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]) due to decreased nighttime atomic O and ClO loss through ClO + O.

**13.2.2. Antarctic vortex** Figure 18 compares averages of retrievals from measurements made in the Antarctic 1992 winter vortex where lower stratospheric ClO was enhanced.

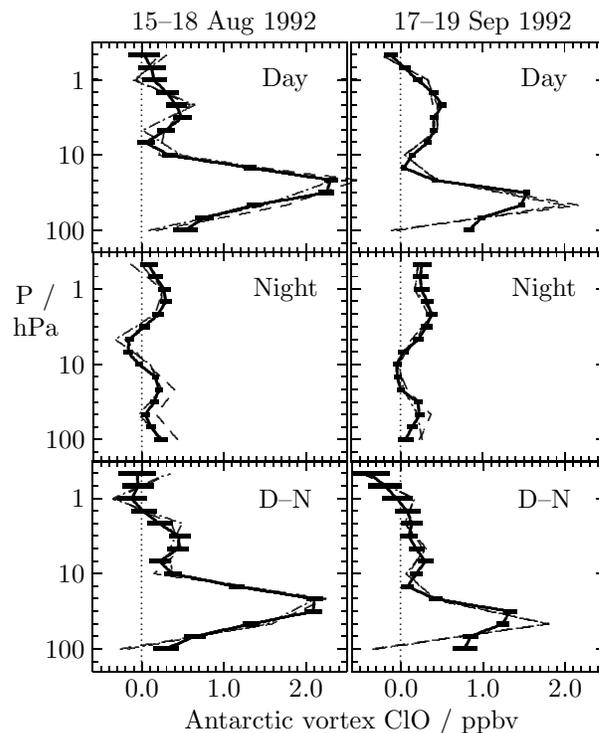
The mid-August 1992 v5 Antarctic daytime peak value of 2.3 ppbv at 22 hPa agrees to within 0.04 ppbv with that of v4, both of which are 0.3 ppbv less than v3. V5 has 0.4 ppbv more ClO at 100 hPa than v4 or v3. Other mid-August 1992 daytime changes are generally less than 0.2 ppbv and within the noise 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



**Figure 17.** Averages of CIO retrievals from MLS 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’ averages are for sza > 90°, and local solar times between midnight and 6 a.m. The 45°S–45°N measurements were made between September 21, 1991 and September 20, 1992, and are averages of ~80,000 individual profiles for day and ~90,000 for night; predicted 1 $\sigma$  precisions for these averages are better than 0.003 ppbv at all altitudes. The 45°–90° measurements were made between May 2 and October 28, 1992, in the north and between November 4, 1991 and April 30, 1992, in the south (averages of ~25,000 day profiles and ~4000 night profiles each for N and S); 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 located at breakpoints of the piecewise-linear representation of the profile.

by 0.21 ppbv.

The mid-September 1992 Antarctic v5 profile has significantly more daytime CIO at 100 hPa (0.84 ppbv) than does v4 (–0.04 ppbv) or v3 (–0.11 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 ver-

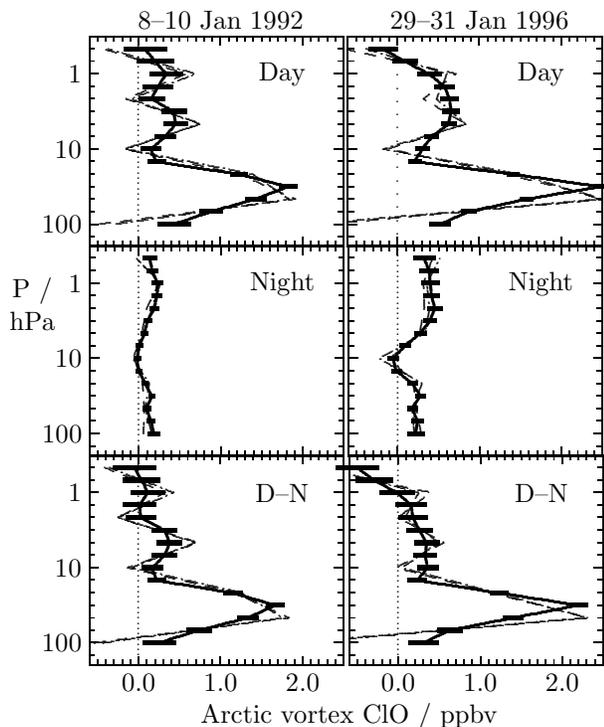


**Figure 18.** Average of CIO retrievals from measurements made in the 1992 Antarctic winter vortex at locations of greatest CIO enhancement in the lower stratosphere. Solid thick lines are v5 data with horizontal bars indicating the  $\pm 1\sigma$  predicted precision of the averages; dash-dot-dash are v4 and dashed are v3 (in places these merge). The August 15–18 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 September 17–19, 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 19, for distinguishing ‘night’ and ‘day’ is the number of measurements that were available at different zenith angles.)

sions. The altitude of the enhanced lower stratospheric CIO 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.

**13.2.3. Arctic vortex** Figure 19 compares averages of retrievals from measurements made in the Arctic winter vor-

tex in January 1992 and January 1996, at times and locations where lower stratospheric ClO was enhanced [Waters et al., 1996; Santee et al., 1996].



**Figure 19.** As in Figure 18 but for Arctic January measurements. The January 8–10, 1992 measurements (left panels) are from  $60^{\circ}$ – $80^{\circ}$ N and  $30^{\circ}$ W– $60^{\circ}$ E: ‘Day’ is for  $\text{sza} < 87^{\circ}$  and the average of 12–14 individual profiles; ‘Night’ is for  $\text{sza} > 100^{\circ}$ , and the average of 116–118 profiles. The January 29–31, 1996 measurements (right panels) are from  $60^{\circ}$ – $80^{\circ}$ N and  $45^{\circ}$ – $105^{\circ}$ E: ‘Day’ is for  $\text{sza} < 90^{\circ}$ , and the average of 28–30 profiles; ‘Night’ is for  $\text{sza} > 110^{\circ}$ , and the average of 51–52 profiles.

Day ClO mixing ratios at the profile peak agree to better than 0.1 ppbv among the three 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 ( $\sim -1$  ppbv, which are representative of the individual profiles that went into the average, and not the result of a single very bad profile) are not present in v5, which has  $\sim 0.5$  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 at 10 hPa in January 1996 where v4 and v3 are more unrealistically negative ( $-0.2$  ppbv) than v5 ( $-0.05$  ppbv).

### 13.3. ClO data after June 15, 1997

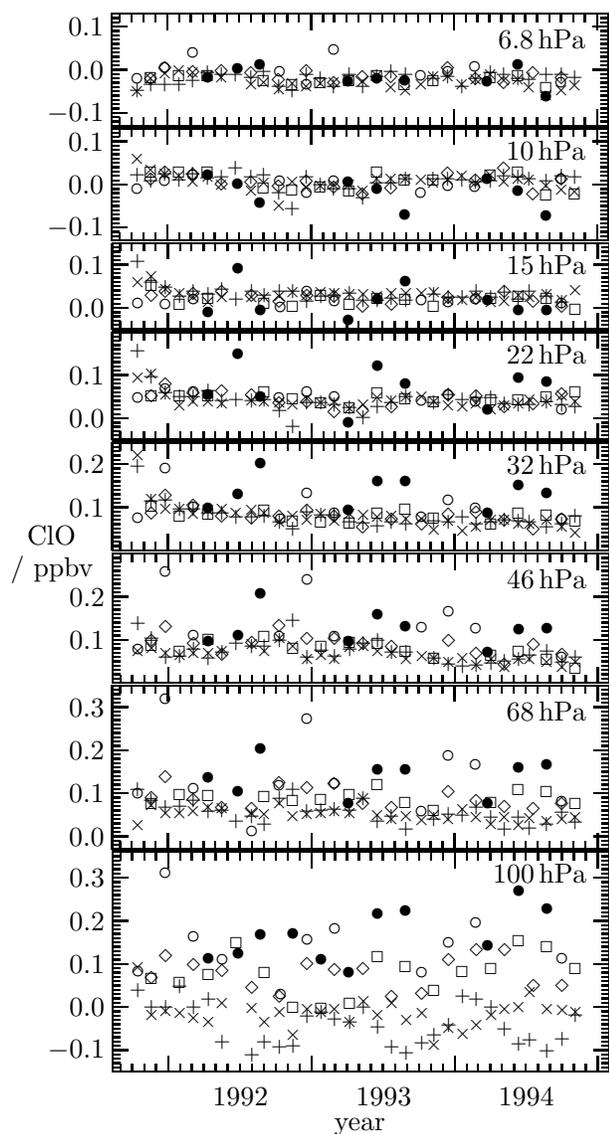
After June 15, 1997, MLS was operated using only the 205-GHz radiometer, in order to conserve spacecraft power. As described in section 6.4, the impact of this change was characterized by reprocessing data from earlier in the mission using the algorithms modified for ‘205-GHz only’ operation, and comparing the results with the output of the standard data processing.

Daily zonal means show that the ‘205-GHz only’ values differ from the standard v5 values by  $\sim 0.05$  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  $\sim 0.2$  ppbv. This offset is not necessarily removed by taking day/night differences. Thus, the ClO data after June 15, 1997 are expected to have systematic differences from earlier data by up to  $\sim 0.2$  ppbv at 100 hPa and in polar enhanced situations, and up to  $\sim 0.05$  ppbv elsewhere.

### 13.4. Determining biases in lower stratospheric ClO data

Limitations of the data processing algorithms to account for curvature and features in the observed limb spectra with an accuracy better than  $\sim 0.1$  K brightness introduce a (thought to be mostly ‘bias’) uncertainty of  $\sim 0.1$  ppbv in retrieved ClO. This uncertainty is believed less severe in the upper stratosphere than in the lower stratosphere, where there are more spectral features and the broader lines make measurement of ‘baseline’ more difficult. However, a robust feature of models that predict diurnal behavior of ClO [Ko and Sze, 1984; Froidevaux et al., 1985; Ricaud et al., 2000, for example] is that—at heights below  $\sim 35$  km (pressures greater than  $\sim 5$  hPa) and away from enhanced chlorine chemistry in the winter polar vortices—the ClO abundances at night decrease essentially to zero. Very accurate *in situ* measurements of midlatitude ClO at 20 km show nighttime abundances of no more than the detection threshold of  $\sim 0.001$  ppbv, in agreement with the model predictions [Brune et al., 1990]. By assuming that nighttime lower stratospheric ClO abundances are zero away from the winter polar vortices, we can infer biases in the MLS ClO data.

Figure 20 shows time series of monthly average v5 nighttime ClO on each retrieval surface between 6.8 hPa and 100 hPa for the first  $\sim 3$  years of the UARS mission in which MLS operations were mostly normal. The nighttime values were selected for local solar time between midnight and 6 a.m. (to avoid including evening measurements when ClO can still be decaying from daytime values) and for solar zenith angles greater than  $90^{\circ}$  (to avoid including measurements where sunrise occurs before 6 a.m.). The points



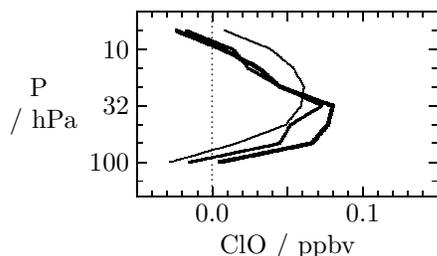
**Figure 20.** Time series of v5 ClO monthly zonal means for measurements between midnight and 6 a.m., and solar zenith angles greater than  $90^\circ$ :  $60^\circ\text{N}$ – $80^\circ\text{N}$ ( $\circ$ ),  $30^\circ\text{N}$ – $60^\circ\text{N}$ ( $\diamond$ ),  $0^\circ$ – $30^\circ\text{N}$ ( $+$ ),  $0^\circ$ – $30^\circ\text{S}$ ( $\times$ ),  $30^\circ\text{S}$ – $60^\circ\text{S}$ ( $\square$ ),  $60^\circ\text{S}$ – $80^\circ\text{S}$ ( $\bullet$ ). Each point is the average for one UARS month. A point is included only if there are more than 300 individual retrievals meeting the standard quality criteria, corresponding to expected precision due to instrument noise of better than  $\sim 0.02$  ppbv ( $\sim 0.04$  ppbv at 100 hPa). Typically, 1000–4000 individual profiles were used in each average, corresponding to better than  $\sim 0.01$  ppbv expected precision. Small ticks on the horizontal axis are calendar month boundaries.

in Figure 20 generally lie in a tight cluster at each level (other than 100 hPa) with relatively few outside the cluster. The two  $60^\circ\text{N}$ – $80^\circ\text{N}$  points at 6.8 hPa in late February 1992 and 1993 are where examination of the profiles indicates that descent of more abundant ClO from higher altitudes reached 6.8 hPa. The larger  $0^\circ$ – $30^\circ\text{N}$  and  $0^\circ$ – $30^\circ\text{S}$  values in October 1991 at 15, 22, and 32 hPa can be explained by residual effects of Pinatubo  $\text{SO}_2$  not accounted for in v5. The  $60^\circ\text{N}$ – $80^\circ\text{N}$  and  $60^\circ\text{S}$ – $80^\circ\text{S}$  points lying above the general cluster all occur during winter and are thought due to increased nighttime ClO from thermal decomposition of enhanced ClOCl. MLS ClO maps for January 1992 show enhanced nighttime ClO in warmer regions of the Arctic vortex with abundances that are consistent with the amounts expected for thermal decomposition of ClOCl [Waters et al., 1993]. Examination of the nighttime ClO abundances for the Antarctic and Arctic winter vortices given in Figures 18 and 19 show that they vary with temperature and pressure in reasonable agreement with predictions for ClOCl thermal decomposition provided by R.J. Salawitch. The  $\sim -0.05$  ppbv  $60^\circ\text{S}$ – $80^\circ\text{S}$  values at 10 hPa in August 1992, 1993 and 1994 that stand out from the cluster are unexplained, and probably—since they all occur at the same month in each of the three years—represent some atmospheric effect that is not accounted for in the retrievals. Nothing unusual occurred in MLS operations during these months that is thought to affect the ClO, nor that would cause the ‘abnormal’ values (differing by  $\sim 0.05$  ppbv from the cluster) at  $0^\circ$ – $30^\circ\text{N}$  and  $0^\circ$ – $30^\circ\text{S}$  at 10 hPa in October–November 1992, at  $60^\circ\text{S}$ – $80^\circ\text{S}$  and 15 hPa in April 1993, and at  $0^\circ$ – $30^\circ\text{N}$  and 22 hPa in November 1992—so these observations may indicate some atmospheric effect that is not included in the MLS forward model. The scatter in the points at 100 hPa, and to a much lesser extent at 68 hPa, is significantly larger than can be explained by instrument noise and varies with latitude. This is likely to be due to some unaccounted-for atmospheric phenomenon that is affecting the radiances, and thus the retrievals, at these low altitudes.

Figure 21 shows, for altitudes where nighttime ClO is thought to be ‘zero’, the profile of retrieved nighttime ClO averaged over  $60^\circ\text{S}$ – $60^\circ\text{N}$  for three portions of the UARS mission as indicated in the caption. We interpret the curves in Figure 21 as bias artifacts in MLS v5 ClO.

### 13.5. Estimated vertical resolution, precision and accuracy of v5 ClO

Table 15 gives the v5 ClO vertical resolution, typical single profile precision, known bias, and estimated accuracy. The vertical resolution given in the table is the full width at half maximum of the rows of the averaging kernel ma-



**Figure 21.** Bias in MLS v5 ClO determined from average of nighttime (0–6 a.m.,  $\text{sza} > 90^\circ$ ) retrievals between  $60^\circ\text{S}$  and  $60^\circ\text{N}$ . Thick line: average of 323,630 individual retrieved profiles from measurements taken between October 1, 1991 and November 19, 1994, when MLS operations were mostly normal. Medium line: average of 70,454 profiles between February 1, 1995, and June 15, 1997, in which antenna scan slips occurred. Thin line: average of 33,136 profiles between June 15, 1997, and March 30, 2000, when only the 205-GHz radiometer was operated and scan slips also occurred. The expected precision, based on instrument noise, varies from  $< 0.001$  ppbv for the thick line to  $\sim 0.002$  ppbv for the thin line.

trix computed for the nominal MLS operational scan and radiometer noise.

The typical single profile precisions in Table 15 are  $1\sigma$  values, based on the minimum monthly rms variability in individual night retrievals from measurements equatorward of  $45^\circ$  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 estimated precision (uncertainty) given in the data files overestimates the actual precision (i.e., is conservative), as mentioned in section 6.1. The estimated uncertainties given in the ClO data files should be multiplied by the ‘ratio’ values in the fourth column of Table 15 to obtain a better value for the precision. The ClO precision can be improved by averaging together 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.

Values in the ‘known bias’ column of Table 15 are from the thick line of Figure 21 for the first  $\sim 3$  years of the mission, which differ negligibly from values for the additional period up to June 15, 1997, when both the 205 and 63-GHz radiometers were operated, and thus apply to the majority of MLS data. More appropriate values for data taken after June

15, 1997, when the 63-GHz radiometer was turned off, are given by the thin line of Figure 21.

The ‘estimated accuracy’ column of Table 15 gives the ‘bias’ (i.e., additive) uncertainty in ppbv after subtracting the ‘known bias’ and the ‘scaling’ (i.e., multiplicative) uncertainty in %. Values given in the table represent 90–95% confidence levels (roughly  $2\sigma$ ). 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 20. The bias uncertainty is increased to 0.15 ppbv for winter polar vortex conditions because, as shown in Figure 18, 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. Users of the data should remove biases by taking day/night differences whenever possible. We have put 0.1 ppbv bias uncertainty on ClO 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.

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 in the last column of Table 15). The scaling uncertainty in v5 data, based on the arguments given in Waters *et al.* [1996], is  $\sim 15\%$  (at the  $\sim 90$ – $95\%$  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.

The estimated overall absolute 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  $1\sigma$  values, whereas accuracies are 90–95% confidence (roughly  $2\sigma$ ) values.

### 13.6. Known artifacts in v5 ClO

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 15 from the values given in the v5 MLS data files. For data after June 15, 1997, a better correction for the biases is obtained from the thin curve in Figure 21.

**Table 15.** Vertical resolution, precision, known bias and accuracy for V5 ClO. See text for further explanation.

Pressure / hPa	Vertical resolution <sup>a</sup> / km	Typical precision / ppbv	Precision ratio <sup>b</sup>	Known bias / ppbv	Estimated accuracy <sup>c</sup> , after subtracting known bias (values in parentheses apply to polar winter vortex data)
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%

<sup>a</sup> As defined in section 6.2.

<sup>b</sup> Data file uncertainties should be multiplied by these numbers to obtain a better value for the '1 $\sigma$ ' single profile precision (see text).

<sup>c</sup> Accuracies quoted here represent roughly a 95% confidence level ('2 $\sigma$ ' values).

- ClO low-latitude values at  $\sim$ 46–4.6 hPa are artificially high in September and October 1991 (by up to  $\sim$ 0.5 ppbv in September and decaying through October to less than 0.1 ppbv). This is due to residual contamination by Pinatubo SO<sub>2</sub> which is not accounted for in the v5 retrievals. Day/night differences remove this artifact.
- A negative bias of  $\sim$ 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.
- 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 15.
- 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 15.

### 13.7. Caveats for using v5 ClO

Caveats for the v5 ClO data are:

- the general caveats described in section 5,

- the artifacts described in the previous subsection,
- values in the files for pressures greater than 100 hPa should never be used,
- values in the files for pressures smaller than 1 hPa are not necessarily reliable.

## 14. Nitric Acid

Although measurement of HNO<sub>3</sub> was not initially an MLS objective, a significant HNO<sub>3</sub> 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<sub>3</sub> in the lower stratosphere. MLS HNO<sub>3</sub> measurements from precursory retrieval algorithms were presented by *Santee et al.* [1995]. HNO<sub>3</sub> became a standard MLS data product in v4; general information on the v4 HNO<sub>3</sub> quality, resolution, and suitability for various scientific studies (in particular investigations of polar stratospheric clouds) can be found in *Santee et al.* [1998] and *Santee et al.* [1999].

After the MLS v5 data set was produced it was discovered that emissions from the HNO<sub>3</sub>  $\nu_9$  and  $\nu_7$  excited vibrational states, which were omitted from the v5 retrieval system, are significant in the spectral region in which MLS HNO<sub>3</sub> is being retrieved. Neglecting the contributions from these lines caused the retrieved MLS v5 values to significantly overestimate HNO<sub>3</sub> abundances at some levels in the stratosphere. An empirical correction to the MLS v5 HNO<sub>3</sub> data set has been derived and is described below in sufficient detail to

allow its application to the v5 HNO<sub>3</sub> Level 3A files by the user. Alternatively, corrected (v501) HNO<sub>3</sub> Level 3AT files may be obtained directly from the MLS team.

#### 14.1. Changes in Algorithms for v5 HNO<sub>3</sub>

More rigorous error propagation as well as improvements in the 03\_205 (retrieved in the same band as HNO<sub>3</sub>) and tangent point pressure retrievals have led to substantially less scatter in the HNO<sub>3</sub> values in v5 than in v4. Thus the precision for v5 is much better (by a factor of 2–3) than for v4 despite the fact that the v5 retrievals are performed on every UARS surface. In addition to the strong HNO<sub>3</sub> feature just outside band 4 (used for the 03\_205 measurement), emission from several weak HNO<sub>3</sub> lines in bands 2 and 3 (primarily used to measure ClO) is 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).

Because HNO<sub>3</sub> is retrieved in the 205-GHz ozone band, the relevant quality flag for HNO<sub>3</sub> data is QUALITY\_03\_205. In v5 the algorithm for setting this parameter was modified because of changes in the  $\chi^2$  statistic describing the fit to the radiances. The  $\chi^2$  statistic for this band is now less correlated with anomalies in the retrieved HNO<sub>3</sub> than it was in v4. The  $\chi^2$  criteria used to assign values to QUALITY\_03\_205 were determined empirically to eliminate most unreasonable ‘spikes’ without rejecting an excessive amount of good data. With the exception of a few months when unusual instrument or satellite problems occurred, the overall criteria for selecting good HNO<sub>3</sub> retrievals (QUALITY\_03\_205=‘4’, MMAF\_STAT=‘G’, ‘T’, or ‘t’) discard about 2% of all records in v5, about twice as many as in v4. Note that the MMAF\_STAT condition is unchanged between v4 and v5. In v5, more records (~1%) satisfying this criterion have been assigned quality flags less than ‘4’, indicating bad fits to the radiances and/or bad radiances, than were flagged bad in v4 (typically <0.1%). Thus in some cases individual profiles that passed the recommended quality control measures in v4 will be screened out using the same procedures with the v5 data, even though they do not appear obviously bad. One factor that appears to be contributing to the increased number of poor quality profiles in v5 is the use of radiances down to 150 hPa, which can lead to more contamination by clouds, especially in the tropics. This notion is supported by the spatial distribution of the poor-quality profiles, which appears to be correlated with regions of upper tropospheric convection and cloud ice, based on analyses not shown here. Only profiles with QUALITY\_03\_205 values of ‘4’ should be used for scientific studies. In addition, the HNO<sub>3</sub> data are generally ‘spikier’ in v5 than they were in v4, where ‘spikes’ are identified by comparison of their deviation from monthly zonal

means. Even though a significant fraction of the profiles are being flagged bad in v5, some of these apparent spikes still pass all of the recommended quality control measures and can be identified by inspection and removed on an individual basis.

#### 14.2. Correction to the v5 HNO<sub>3</sub>

We use Equation (5), with all radiances collapsed into one vector and the a priori term neglected, to formulate a ‘corrected’ HNO<sub>3</sub> retrieval,  $\mathbf{x}'$ , that accounts for the contributions from the  $\nu_9$  and  $\nu_7$  excited vibrational states:

$$\mathbf{x}' = \mathbf{x} + \left[ \mathbf{S}_a^{-1} + \mathbf{K}'^T \mathbf{S}_y^{-1} \mathbf{K}' \right]^{-1} \left( \mathbf{K}'^T \mathbf{S}_y^{-1} [\mathbf{y} - \mathbf{f}'(\mathbf{x})] \right)$$

where  $\mathbf{K}'$  is an improved weighting function matrix ( $\mathbf{K}' = \mathbf{K} + \mathbf{k}$ ) that includes the contributions from the ground state,  $\mathbf{K}$ , and the excited vibrational states,  $\mathbf{k}$ . Similarly, a corrected forward model is defined to be the original v5 forward model plus the extra terms from the excited vibrational states:  $\mathbf{f}'(\mathbf{x}) = \mathbf{f}(\mathbf{x}) + \mathbf{kx}$ . Assuming that the retrieval, based on the flawed forward model  $\mathbf{f}(\mathbf{x})$ , fits the radiances optimally (i.e., zero mean radiance residual), then in the mean  $\mathbf{y} - \mathbf{f}(\mathbf{x}) = 0$ . Substituting in for  $\mathbf{f}'(\mathbf{x})$ , defining a new solution covariance,  $\mathbf{S}'_x$ , similar in form to that in Equation (6), and rearranging leads to:

$$\begin{aligned} \mathbf{x}' &= \mathbf{x} + \mathbf{S}'_x \mathbf{K}'^T \mathbf{S}_y^{-1} [-\mathbf{kx}] \\ &= \mathbf{C}(\mathbf{T})\mathbf{x}. \end{aligned} \quad (41)$$

Thus the correction is a linear scaling of the original profile. The correction matrix,  $\mathbf{C}(\mathbf{T})$ , is temperature dependent because of the strong temperature dependence of the HNO<sub>3</sub> excited vibrational states.  $\mathbf{C}(\mathbf{T})$  is nearly tridiagonal and is assumed to be so in order to derive an easily-implemented empirical correction. The full model was run for eight representative temperature profiles that spanned the variations that occur in stratospheric temperatures for different seasons and latitudes.  $\mathbf{C}(\mathbf{T})$  was computed for each case, and the eight results for each tridiagonal element were combined and fitted with a simple linear polynomial in temperature. The tridiagonal elements of the correction matrix are thus represented by:

$$\begin{aligned} C_{l-1,l}(T_{l-1}, T_l) &= \alpha_{l-1,l} + \beta_{l-1,l} T_{l-1} + \gamma_{l-1,l} T_l, \\ C_{l,l}(T_l, T_l) &= \alpha_{l,l} + \beta_{l,l} T_l, \\ C_{l,l+1}(T_l, T_{l+1}) &= \alpha_{l,l+1} + \beta_{l,l+1} T_l + \gamma_{l,l+1} T_{l+1}, \end{aligned} \quad (42)$$

where  $\alpha$ ,  $\beta$  and  $\gamma$  are the coefficients of a linear polynomial in temperature,  $T$  is the atmospheric temperature, and  $l$  denotes a particular altitude level in the HNO<sub>3</sub> profile. The

coefficients  $\alpha$ ,  $\beta$  and  $\gamma$  for the diagonal and the two adjacent sets of elements are provided in Table 16. This polynomial correction is valid over the height range 100 to 4.6 hPa and provides an estimated rms accuracy of 0.2 ppbv for corrected  $\text{HNO}_3$  throughout the domain.

The tabulated coefficients can be used along with an associated temperature profile (either from MLS or from some meteorological analyses interpolated to the MLS measurement location) to correct each  $\text{HNO}_3$  profile in the MLS data set. Applying the correction leads to reductions in the reported v5  $\text{HNO}_3$  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, particularly at the upper levels; it has not eliminated them entirely, however. For example, a comparison of the corrected (v501) v5  $\text{HNO}_3$  data with both the original v5  $\text{HNO}_3$  retrievals and simultaneous, colocated  $\text{HNO}_3$  measurements (version 9) from the UARS Cryogenic Limb Array Etalon Spectrometer (CLAES) [Kumer et al., 1996] is shown in Plate 2 for the 22-hPa level, where the discrepancy between CLAES and MLS v5 was largest (more than 5 ppbv in the zonal mean at some seasons/latitudes). With the correction applied to the MLS v5 data, the disagreement between CLAES and MLS  $\text{HNO}_3$  values at 22 hPa is reduced below  $\sim 2$  ppbv under conditions of wintertime enhancement in the polar vortices and below  $\sim 1$  ppbv at most other seasons/latitudes. The agreement is also within  $\sim 1$  ppbv (and frequently much better) everywhere at and above 15 hPa, throughout the tropics at all altitudes, and during the summer season at all latitudes and altitudes (not shown). At 46 and 68 hPa, however, the correction factors are small and differences of up to 4–5 ppbv remain in the winter polar regions. (Note that the presence of enhanced levels of  $\text{SO}_2$ , which is not retrieved in v5, in the stratosphere following the eruption of Mount Pinatubo caused a high bias in the MLS  $\text{HNO}_3$  retrievals in the equatorial regions for the first 100 days or so of the mission.)

Because this correction leads to increases in the values of the weighting function matrix, it also produces a smaller error covariance. Thus it is necessary to correct not only the mixing ratios, but also the associated uncertainty values given in the Level 3AT files. Although the uncertainty values change with altitude, they are fairly constant over both latitude and time (over the course of the UARS mission), so the correction for them is simpler than for the highly variable mixing ratios. Tests comparing the elements of  $\mathbf{S}'_x$  and  $\mathbf{S}_x$  (i.e., the original v5 solution covariance) indicate that the uncertainty values are changed negligibly at 100, 68, and 46 hPa, but the values reported in the Level 3AT files should be decreased by 0.1 ppbv at 32 and 22 hPa, by 0.2 ppbv at 15 and 10 hPa, and by 0.3 ppbv at 6.8 and 4.6 hPa.

### 14.3. Comparison of v501 and v4 $\text{HNO}_3$

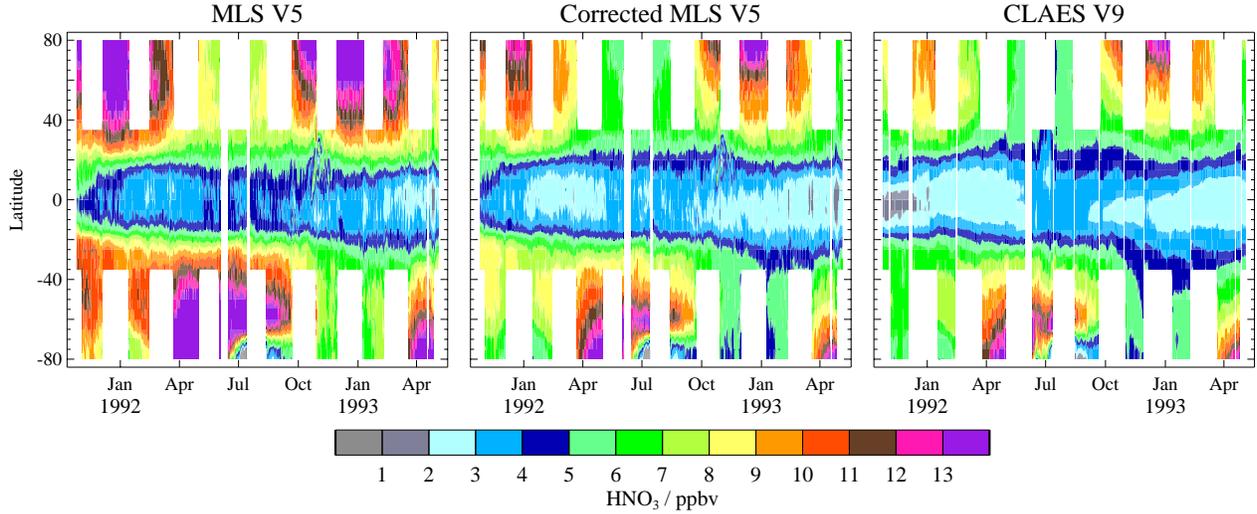
Differences between v501 and v4 average profiles are summarized in Table 17. Because v4 retrievals were performed on the even UARS surfaces and were reliable at and below 22 hPa, only the differences on these surfaces are tabulated. Also, because the distribution of  $\text{HNO}_3$  in the lower stratosphere 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  $\text{HNO}_3$  mixing ratio shifts in altitude from year to year.

V501  $\text{HNO}_3$  global average mixing ratios are slightly larger than those in v4, except at 100 hPa, where v501 values are smaller at all latitudes. V501 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, although a weak negative bias (about 1 ppbv or less) is now present in this region at 100 hPa. However, at mid and high latitudes, especially during early winter when  $\text{HNO}_3$  is enhanced inside the vortex, v501 values are substantially smaller at 22 hPa than in v4.

### 14.4. Estimated Vertical Resolution and Precision of v501 $\text{HNO}_3$

Best precision in the  $\text{HNO}_3$  retrievals is attained at 68 hPa. The general range of useful sensitivity is given in Table 18. While the absolute v5  $\text{HNO}_3$  precision is, to first order, independent of latitude and season, the scientific utility of the data (i.e., signal to noise) can vary due to variations in  $\text{HNO}_3$  abundance. For example, at 100 hPa the single-profile precision greatly exceeds the average  $\text{HNO}_3$  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  $\text{HNO}_3$  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 is not recommended for use in scientific studies.

The typical single-profile precisions given in Table 18 are  $1\sigma$  values. They were obtained by computing (for the first full year of measurements) the minimum monthly rms variability in the corrected  $\text{HNO}_3$  profiles retrieved in a  $10^\circ$  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



**Plate 2.** Time series of daily zonal-mean MLS v5, MLS v501 v5, and CLAES version 9 HNO<sub>3</sub> 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.

**Table 16.** Correction coefficients for computing  $C_{l-1,l}(T_{l-1}, T_l)$ ,  $C_{l,l}(T_l)$ , and  $C_{l,l+1}(T_l, T_{l+1})$

Pressure / hPa	$\alpha_{l-1,l}$	$\beta_{l-1,l}$	$\gamma_{l-1,l}$	$\alpha_{l,l}$	$\beta_{l,l}$	$\alpha_{l,l+1}$	$\beta_{l,l+1}$	$\gamma_{l,l+1}$
4.6	-0.013	$-1.0 \times 10^{-5}$	$-2.2 \times 10^{-4}$	1.009	$-5.0 \times 10^{-4}$	—	—	—
6.8	-0.007	$-5.0 \times 10^{-5}$	$-2.5 \times 10^{-4}$	1.036	$-7.2 \times 10^{-4}$	-0.009	$-5.0 \times 10^{-5}$	$-2.5 \times 10^{-4}$
10	0.014	$-1.2 \times 10^{-4}$	$-3.1 \times 10^{-4}$	1.097	$-1.07 \times 10^{-3}$	0.013	$-1.2 \times 10^{-4}$	$-3.1 \times 10^{-4}$
15	0.055	$-2.8 \times 10^{-4}$	$-3.3 \times 10^{-4}$	1.168	$-1.40 \times 10^{-3}$	0.054	$-2.8 \times 10^{-4}$	$-3.3 \times 10^{-4}$
22	0.081	$-2.2 \times 10^{-4}$	$-4.4 \times 10^{-4}$	1.220	$-1.57 \times 10^{-3}$	0.081	$-2.2 \times 10^{-4}$	$-4.4 \times 10^{-4}$
31	0.066	$-2.2 \times 10^{-4}$	$-2.8 \times 10^{-4}$	1.198	$-1.35 \times 10^{-3}$	0.066	$-2.2 \times 10^{-4}$	$-2.8 \times 10^{-4}$
46	0.034	$-2.2 \times 10^{-4}$	$-5.0 \times 10^{-5}$	1.158	$-1.05 \times 10^{-3}$	0.035	$-2.2 \times 10^{-4}$	$-5.0 \times 10^{-5}$
68	0.013	$-1.3 \times 10^{-4}$	$1.0 \times 10^{-5}$	1.117	$-7.6 \times 10^{-4}$	0.013	$-1.3 \times 10^{-4}$	$1.0 \times 10^{-5}$
100	—	—	—	1.084	$-5.4 \times 10^{-4}$	0.006	$-7.0 \times 10^{-5}$	$1.0 \times 10^{-5}$

**Table 17.** Differences between v501 and v4 HNO<sub>3</sub>

Pressure / hPa	Global <sup>a</sup>		Tropical <sup>b</sup>		Mid-latitude <sup>c</sup>		Polar enhanced <sup>d</sup>		Polar depleted <sup>e</sup>	
	/ ppbv	%	/ ppbv	%	/ ppbv	%	/ ppbv	%	/ ppbv	%
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

<sup>a</sup> Based on ~400,000 profiles from all latitudes for the first full year of data.

<sup>b</sup> Based on ~60,000 profiles from 10°S to 10°N for the first full year of data.

<sup>c</sup> 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.

<sup>d</sup> Based on ~5,000 profiles from 70°N to 80°N during the period from December 1992 to mid-January 1993.

<sup>e</sup> Based on ~4,500 profiles from 70°S to 80°S during the period from mid-August to mid-September 1992.

Differences are v501 – v4; percentages are relative changes from v4.

**Table 18.** Estimated vertical resolution and precision of v501 HNO<sub>3</sub>

Pressure / hPa	Vertical resolution <sup>a</sup> / km	Typical precision / ppbv	Precision ratio <sup>b</sup>
4.6	10.5	1.5	0.7
6.8	10.0	1.4	0.7
10	9.3	1.3	0.7
15	8.7	1.2	0.7
22	7.4	1.2	0.7
31	6.4	1.1	0.7
46	5.7	1.0	0.7
68	5.7	0.8	0.6
100	4.3	1.3	0.9

<sup>a</sup> As defined in section 6.2.

<sup>b</sup> Data file uncertainties should be multiplied by these numbers to obtain a better value for the '1 $\sigma$ ' single profile precision (see text).

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.

The theoretical precision values provided in the HNO<sub>3</sub> 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 18, 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 18 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. As mentioned above, the precision of v501 HNO<sub>3</sub> has been substantially improved (by a factor of 2–3) over that of v4.

The estimated accuracy of the v501 MLS HNO<sub>3</sub> has not yet been quantified; a comprehensive validation effort is currently underway. *Santee et al.* [manuscript in preparation, 2002] will quantify the accuracy.

#### 14.5. Caveats in Use of v5 HNO<sub>3</sub>

1. Omission of some HNO<sub>3</sub> excited vibrational state lines from the retrieval system caused v5 HNO<sub>3</sub> values

to overestimate stratospheric abundances. To correct for this, the linear scaling described in section 14.2 should be applied to the v5 HNO<sub>3</sub> profiles. Alternatively, corrected (v501) HNO<sub>3</sub> Level 3AT files may be obtained directly from the MLS team.

2. Only data between 100 hPa and 4.6 hPa are considered sufficiently reliable for general use in scientific studies.
3. The Level 3 'parameter files' should be examined and only profiles with MMAF\_STAT = 'G', 't', or 'T' and QUALITY\_03\_205 = 4 should be used for scientific studies. Profiles with MMAF\_STAT set to 'T' (which occurs quite infrequently) may have slight biases at pressures larger than or equal to 46 hPa.
4. Only data having positive estimated uncertainties (given in the 'quality field' in Level 3A data files) should be used.
5. These quality control measures are not always sufficient to filter out large ( $> 5\sigma$ ) 'spikes' in the retrieved HNO<sub>3</sub> mixing ratios; occasional anomalous retrievals can be identified by inspection and removed on an individual basis.
6. The uncertainties in the HNO<sub>3</sub> Level 3A files overestimate the actual precision of the measurements. To obtain a more realistic estimate of the true precision, the uncertainties in the data files should be multiplied by a factor of  $\sim 0.7$ , depending on altitude (see Table 18).

## 15. Methyl Cyanide

Methyl cyanide (CH<sub>3</sub>CN, also called acetonitrile) is a new product in v5. An overview of the MLS CH<sub>3</sub>CN dataset, along with a review of the current understanding of the role CH<sub>3</sub>CN plays in the atmosphere, is given in *Livesey et al.* [2001]. CH<sub>3</sub>CN is a trace constituent of the atmosphere, with abundances typically around 150 pptv (parts per trillion by volume) in the troposphere. The main source for CH<sub>3</sub>CN is biomass burning, and localized abundances as high as 10<sup>6</sup> pptv have been seen in some events. In the stratosphere, the role of CH<sub>3</sub>CN remains uncertain. It was thought to simply decay from reactions with OH and O(<sup>1</sup>D) with a lifetime of between 5 and 20 years. However, the MLS observations show an increase in mixing ratio with altitude in the tropics from  $\sim 30$  pptv at 68 hPa to  $\sim 50$  pptv at 30–10 hPa [*Livesey et al.*, 2001].

The MLS CH<sub>3</sub>CN data are scientifically useful between 68 and 1 hPa. While CH<sub>3</sub>CN abundance at 100 hPa is retrieved, it is believed that spectral features from H<sub>2</sub><sup>18</sup>O con-

**Table 19.** Estimated precision, vertical resolution and accuracy for v5 MLS CH<sub>3</sub>CN data. See text for details

Pressure / hPa	Vertical resolution <sup>a</sup> / km	Typical precision / pptv	Precision ratio <sup>b</sup>	Estimated accuracy <sup>c</sup>
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%

<sup>a</sup> As defined in section 6.2.

<sup>b</sup> Data file uncertainties should be multiplied by these numbers to obtain a better value for the ‘1 $\sigma$ ’ single profile precision (see text).

<sup>c</sup> Accuracies quoted here represent roughly a 95% confidence level (‘2 $\sigma$ ’ values).

taminate the CH<sub>3</sub>CN signal, leading to an unpredictable bias in the CH<sub>3</sub>CN at 100 hPa. CH<sub>3</sub>CN data at lower pressures are not significantly affected by H<sub>2</sub><sup>18</sup>O signals. The MLS data are not reliable above 1 hPa, because the spectral contrast in the radiance observations is approaching the accuracy limit of the instrument.

Individual profiles of MLS CH<sub>3</sub>CN data have a precision of 40–60 pptv. For scientific study, some form of averaging is generally required. For example, a monthly zonal mean dataset with a 10° latitudinal resolution will have a precision of 1 pptv at 10 hPa.

Occasionally, strong enhancements are seen in the MLS CH<sub>3</sub>CN dataset 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<sup>3</sup> pptv observed. A detailed study has concluded that they represent true enhancements in lower stratospheric CH<sub>3</sub>CN, 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) some days earlier [Livesey et al. in preparation]; the causes of the few similar events in the dataset are under investigation.

### 15.1. Estimated Vertical Resolution, Precision, and Accuracy in v5 CH<sub>3</sub>CN

Table 19 summarizes the precision, vertical resolution and accuracy of the MLS CH<sub>3</sub>CN dataset. The vertical resolution given is the full width at half-maximum of the rows of the averaging kernel matrix computed for a typical MLS

scan, with typical levels of radiometer noise.

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 19.

The accuracy is defined in terms of possible bias and scaling terms. The values given are believed to represent 60–70% confidence levels for these factors. These estimates were obtained by analogy with ClO (see section 13), accounting for the relative line strengths of CH<sub>3</sub>CN and ClO and assuming 10% uncertainty in the CH<sub>3</sub>CN 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<sub>3</sub>CN 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<sub>3</sub>CN

1. Data at 100 hPa are contaminated by emission from H<sub>2</sub><sup>18</sup>O and should not be used.
2. Data at pressures lower than 1 hPa are unreliable because of instrumental limitations.
3. The spectral signature of CH<sub>3</sub>CN in the MLS passband is very similar to that of SO<sub>2</sub>. As SO<sub>2</sub> is not retrieved in v5, the retrieval algorithms will interpret any enhancement in SO<sub>2</sub> as an enhancement in CH<sub>3</sub>CN. The high SO<sub>2</sub> resulting from the Pinatubo eruption leads to an unquantified high bias in the pre-1992 CH<sub>3</sub>CN data, which should not be used. A more short-lived, localized bias resulting from the Eruption of Mt. Lascar in Chile is seen from April 22–24, 1993.

### 15.3. Caveats in Use of v5 CH<sub>3</sub>CN

1. Data should only be used between 68 hPa and 1 hPa, and after Jan 1, 1992.
2. Profiles should only be considered if the MMAF\_STAT field is ‘G’, ‘T’ or ‘t’, and QUALITY\_C10 is 4. QUALITY\_C10 should be used as the CH<sub>3</sub>CN lines observed by MLS are in bands 2 and 3, used for the ClO observations.

3. Data points whose precision is set to a negative value are strongly affected by a priori measurements, and should not be used. Such biases will not be lessened by averaging, as they would be present for all points in the average.

## 16. Lessons learned from version 5

This paper describes the v5 algorithms as implemented and the data they produced. The work has also identified issues that may have benefited from more study during the development and configuration of the v5 software. The most notable of these are the issues associated with the use of off-diagonal terms in the a priori covariance matrices (see section 3.2) and their impact on the observed scatter in the data, described in section 6.1. In retrospect, the length scales chosen for O<sub>3</sub>, H<sub>2</sub>O, HNO<sub>3</sub> and CH<sub>3</sub>CN were probably longer than optimum. Another issue that may have warranted more investigation during the development of the v5 algorithms is the trade-off between the baseline and extinction elements of the state vector, described in section 3.6. The retrieval algorithms typically retrieved larger baseline terms than one might expect, using negative values for the extinction factor to compensate. More detailed study of the constraint of these terms may have lessened this rather inelegant effect. However, this issue had negligible impact on the main MLS data products.

There are no plans to perform further reprocessing on the UARS MLS dataset. However, the lessons learned from v5 are being applied to the algorithms and software for EOS MLS, planned for launch on the EOS Aura platform in 2004.

## 17. Summary and conclusions

We have shown that the MLS v5 algorithms produce data that are generally of higher quality than earlier versions. The doubling of vertical resolution 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 chemical enhancements in ClO. The 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.

For some species, there exist other versions of MLS data that are considered preferable. For upper tropospheric hu-

midity, 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 dataset [Pumphrey, 1999] is felt to be of superior overall quality to v5. The v5 nitric acid data exhibit a bias due to the omission of contributions from excited states. This bias can be easily corrected as described in section 14.2. Corrected (v501) HNO<sub>3</sub> data files are available from the MLS science team.

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.

## Appendix: MLS operations and data coverage

(Last edited March 6, 2002.)

Table A1 gives a chronology of MLS operations and events that significantly impacted data collection or quality. Plate A1 gives a calendar of daily data coverage from launch through the end of 1999.

UARS was launched on September 12, 1991, and the first full day of MLS data was obtained on September 21. During late September and October 1991, time was spent characterizing the MLS performance and ‘tuning’ its operations. On October 31, a limb scan was implemented with denser vertical spacing of measurements in the lower stratosphere than at other altitudes; this scan pattern has been used in normal operations for the remainder of the mission.

Problems with the UARS solar array caused MLS, or portions of it, to be turned off during much of the early southern winter observing period in June 1992. In late 1992, low voltage from the UARS power supply at spacecraft sunrise started affecting the MLS switching mirror movement during a few limb scans each orbit and caused excess noise in the measurements—most notably in ClO, as evidenced by the increased  $\chi^2$ -statistic for ClO (see Plate 1 of Waters et al. [1996]). The switching mirror was not moved during a few limb scans under spacecraft sunrise low voltage conditions from November 18, 1992 until August 9, 1993, when the low voltage problem was circumvented with a solution using the secondary commutator of the switching mirror motor.

In late 1992 the 183-GHz radiometer started showing increased noise, which eventually developed into erratic behavior. The last full day of useful 183-GHz measurements (stratospheric H<sub>2</sub>O and 183-GHz O<sub>3</sub>) was April 15, 1993. This radiometer was turned off on April 24, 1993 after analyses indicated failure of its mixer.

The MLS antenna scanning mechanism began exhibiting signs of wear by early 1994; the period from March to



1994		1995		1996	
6 Jan 94 - 4 Feb 94 D0848 - D0877	18457	1 Jan 95 - 30 Jan 95 D1208 - D1237	0	28 Dec 95 - 26 Jan 96 D1569 - D1598	13176
5 Feb 94 - 16 Mar 94 D0878 - D0917	44048	31 Jan 95 - 11 Mar 95 D1238 - D1277	5240	27 Jan 96 - 5 Mar 96 D1599 - D1637	20048
17 Mar 94 - 22 Apr 94 D0918 - D0954	23727	12 Mar 95 - 17 Apr 95 D1278 - D1314	4463	6 Mar 96 - 11 Apr 96 D1638 - D1674	21350
23 Apr 94 - 23 May 94 D0955 - D0985	19487	18 Apr 95 - 19 May 95 D1315 - D1346	0	12 Apr 96 - 14 May 96 D1675 - D1707	14790
24 May 94 - 4 Jul 94 D0986 - D1027	53664	20 May 95 - 30 Jun 95 D1347 - D1386	7744	15 May 96 - 24 Jun 96 D1708 - D1748	16487
5 Jul 94 - 4 Aug 94 D1028 - D1059	19763	1 Jul 96 - 30 Jul 96 D1389 - D1418	3522	25 Jun 96 - 24 Jul 96 D1749 - D1778	9572
5 Aug 94 - 12 Sep 94 D1060 - D1097	50018	31 Jul 96 - 8 Sep 96 D1419 - D1458	13160	25 Jul 96 - 9 Oct 96 D1779 - D1818	8712
13 Sep 94 - 20 Oct 94 D1098 - D1135	30229	9 Sep 96 - 15 Oct 96 D1459 - D1495	15510	3 Sep 96 - 9 Oct 96 D1819 - D1855	8776
21 Oct 94 - 20 Nov 94 D1136 - D1166	13839	16 Oct 96 - 16 Nov 96 D1496 - D1527	22626	10 Oct 96 - 11 Nov 96 D1856 - D1888	19979
21 Nov 94 - 31 Dec 94 D1167 - D1207	5702	17 Nov 96 - 27 Dec 96 D1528 - D1568	26639	12 Nov 96 - 22 Dec 96 D1889 - D1929	5880

Plate A1. Continued.

1997		1998		1999																							
23 Dec 96 - 20 Jan 97 D1930 - D1958	1123	27	28	29	30	31	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	9222
21 Jan 97 - 1 Mar 97 D1959 - D1988	1138	1166	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	11410
2 Mar 97 - 7 Apr 97 D1989 - D2005	1207	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	10358
8 Apr 97 - 10 May 97 D2006 - D2068	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	18009
11 May 97 - 20 Jun 97 D2069 - D2109	1189	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	7546
21 Jun 97 - 20 Jul 97 D2110 - D2139	1189	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	5321
21 Jul 97 - 29 Aug 97 D2140 - D2179	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	13058
30 Aug 97 - 4 Oct 97 D2180 - D2215	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	1198	5946
5 Oct 97 - 7 Nov 97 D2216 - D2249	1319	377	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	15453
8 Nov 97 - 18 Dec 97 D2250 - D2290	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	13135
19 Dec 97 - 16 Jan 98 D2291 - D2319	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	6591
17 Jan 98 - 25 Feb 98 D2320 - D2359	1318	1300	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	18835
26 Feb 98 - 2 Apr 98 D2360 - D2395	1318	1319	1304	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	13147
3 Apr 98 - 5 May 98 D2396 - D2428	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	9135
6 May 98 - 16 Jun 98 D2429 - D2470	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	13788
17 Jun 98 - 15 Jul 98 D2471 - D2499	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	742
16 Jul 98 - 25 Aug 98 D2500 - D2540	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1029
26 Aug 98 - 29 Sep 98 D2541 - D2575	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	211
30 Sep 98 - 2 Nov 98 D2576 - D2609	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	379
3 Nov 98 - 13 Dec 98 D2610 - D2650	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	127
14 Dec 98 - 11 Jan 99 D2651 - D2679	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	330
12 Jan 99 - 20 Feb 99 D2680 - D2719	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	265
21 Feb 99 - 28 Mar 99 D2720 - D2755	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	139
29 Mar 99 - 1 May 99 D2756 - D2789	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	43
2 May 99 - 11 Jun 99 D2790 - D2830	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	798
12 Jun 99 - 10 Jul 99 D2831 - D2859	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	28
11 Jul 99 - 20 Aug 99 D2860 - D2890	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	736
21 Aug 99 - 24 Sep 99 D2901 - D2935	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	0
25 Sep 99 - 29 Oct 99 D2936 - D2970	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	0
30 Oct 99 - 7 Dec 99 D2971 - D3009	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	1318	0

Plate A1. Continued.

July 1994 was primarily devoted to testing new operational modes, resulting in significantly reduced data. Continuing difficulties with the MLS scan system, and with the UARS batteries and solar array, caused severely limited data collection from October 1994 through July 1995. Various modifications to the operation of the antenna scanning mechanism were implemented in February 1995, and in June 1995 a schedule of (typically) 2 days of scanning followed by 1 day of ‘rest’ was adopted to conserve lifetime. During some of the period after March 1994, MLS was operated in a ‘limb-tracking’ mode to provide data on atmospheric gravity wave activity [Wu and Waters, 1996] while not causing excessive wear on the scan mechanism.

The UARS solar array, because of problems with its drive systems, was ‘parked’ in May 1995, resulting in a decrease of available power and an instrument power sharing mode with MLS periodically turned off. The UARS onboard computer experienced an anomalous shutdown in May 1997, stressing the batteries and leading to one (of three) being removed from service. Because of the degrading UARS power situation, MLS made measurements on only 6 days during May and June 1997. The 63-GHz radiometer that provided tangent pressure and temperature was turned off after June 14, 1997 to save power. MLS was put in ‘standby mode’ in July 1999 to conserve its remaining lifetime for possible overlapping measurements with the MLS to be launched on the EOS Aura mission in 2003. It was operated again for two brief periods in February and March 2000 to obtain Arctic observations [Santee et al., 2000], and briefly in mid-August 2001 to obtain correlative observations for the Odin satellite.

**Acknowledgments.** 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, and the staff of the UARS CDHF. We also wish to thank, R.R. Lay, and D. Flower for their support and management of UARS MLS operations, along with the UARS operations staff. We wish to 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 also acknowledge the assistance and contributions of the various investigators whose correlative datasets 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 processing team).

The research described in this paper was carried out by the Jet Propulsion Laboratory, California Institute of Technology, under a contract with the National Aeronautics and Space Administration. HCP acknowledges support from the Natural Environment Research Council.

## References

- Barath, F. T., et al., The Upper Atmosphere Research Satellite Microwave Limb Sounder Experiment, *J. Geophys. Res.*, **98**, 10,751–10,762, 1993.
- Brune, W. H., D. W. Toohey, S. A. Lloyd, and J. G. Anderson, The sunrise and sunset variation of ClO in the lower stratosphere, *Geophys. Res. Lett.*, **17**, 509–512, 1990.
- Burke, J. R., and T. A. Lungu, Upper Atmosphere Research Satellite, Microwave Limb Sounder Standard Formatted Data Units, distributed with MLS data, and available from the MLS home page <http://mls.jpl.nasa.gov/>.
- Cunnold, D. M., L. Froidevaux, J. M. Russell, III, B. Connor, and A. Roche, Overview of UARS ozone validation based primarily on intercomparisons among UARS and Stratospheric Aerosol and Gas Experiment II measurements., *J. Geophys. Res.*, **101**, 10,335–10,350, 1996a.
- Cunnold, D. M., H. Wang, W. P. Chu, and L. Froidevaux, Comparisons between Stratospheric Aerosol and Gas Experiment II and Microwave Limb Sounder ozone measurements and aliasing of SAGE II ozone trends in the lower stratosphere, *J. Geophys. Res.*, **101**, 10,061–10,075, 1996b.
- Cunnold, D. M., M. J. Newchurch, L. E. Floyd, H. J. Wang, J. M. Russell, III, R. McPeters, J. Zawodny, and L. Froidevaux, Uncertainties in upper stratospheric ozone trends from 1979 to 1996, *J. Geophys. Res.*, **105**, 4427–4444, 2000.
- Danilin, M. Y., M. L. Santee, J. M. Rodriguez, M. K. W. Ko, J. M. Mergenthaler, J. B. Kumer, A. Tabazadeh, and N. J. Livesey, Trajectory hunting: A case study of rapid chlorine activation in December 1992 as seen by UARS, *J. Geophys. Res.*, **105**, 4003–4018, 2000.
- Danilin, M. Y., et al., Comparison of ER-2 aircraft and POAM-III, MLS and SAGE-II satellite measurements during SOLVE using traditional correlative analysis and trajectory hunting technique, *J. Geophys. Res.*, 2001, in press.
- de Boor, C., *A Practical Guide to Splines*, Springer-Verlag, 1987, fourth edition.
- de Zafra, R. L., J. M. Reeves, and D. T. Shindell, Chlorine monoxide in the Antarctic spring vortex I. Evolution of midday vertical profiles over McMurdo Station., *J. Geophys. Res.*, **100**, 13,999–14,007, 1995.
- Dudhia, A., and N. J. Livesey, Validation of the Improved Stratospheric and Mesospheric Sounder temperature measurements, *J. Geophys. Res.*, **101**, 9795–9809, 1996.
- Feist, D. G., C. P. Aellig, N. Kämpfer, P. M. Solomon, J. W. Barrett, S. Zoonematkermani, P. Hartogh, C. Jarchow, and J. W. Waters, Validation of stratospheric ClO measurements from the Millimeter-wave Atmospheric Sounder (MAS), *J. Geophys. Res.*, **105**, 9053–9062, 2000.
- Fishbein, E. F., et al., Validation of UARS Microwave Limb Sounder temperature and pressure measurements, *J. Geophys. Res.*, **101**, 9983–10,016, 1996.
- Froidevaux, L., M. Allen, and Y. L. Yung, A critical analysis of ClO and O<sub>3</sub> in the mid-latitude stratosphere, *J. Geophys. Res.*, **90**, 12,999–13,029, 1985.
- Froidevaux, L., J. W. Waters, W. G. Read, P. S. Connell, D. E. Kinneson, and J. M. Russell, III, Variations in the free chlorine content of the stratosphere (1991–1997): Anthropogenic,

**Table A1.** Chronology of significant events relevant to UARS MLS operations and data.

Calendar day(s)	UARS day number(s)	Event
Sep 12, 1991	1	UARS launch
Sep 18, 1991	7	All MLS systems and subsystems on
Sep 21, 1991	10	First full day of MLS atmospheric data
Sep 23, 1991 – Oct 1, 1991	16-20	MLS initial characterization period, UARS roll-up on Sep 30
Oct 17–30, 1991	36–49	Some ‘tuning’ of MLS operations
Oct 31, 1991	50	Started using limb scan with denser sampling in lower stratosphere
Nov 1–2, 1991	51-52	UARS in safhold mode
Mar 17, 1992	188	UARS instruments inadvertently off at 10:36 UT; MLS back at 19:00 UT
Apr 8, 1992	210	MLS oblateness correction resolution changed from 2.5 to 1.25 km
Jun 2, 1992	265	UARS solar array drive anomaly; instruments turned off at 19:42 UT
Jun 14, 1992	277	MLS back on and fully operational
Jun 19 – Jul 9, 1992	282–302	183-GHz radiometer (and band 3 of 205-GHz) off to reduce power
Jul 14–17, 1992	307–310	MLS off; back on at 17:00 UT on 17 Jul
Oct 11, 1992	396	MLS moon scan for field-of-view calibration
Nov 18, 1992	434	Stopped moving switching mirror when voltage low at UARS sunrise
Mar 15, 1993	551	All UARS instruments off at 13:57 UT; MLS back operational at 22:45
Apr 6, 1993	573	UARS roll maneuver; nominal operations resumed at 05:00 UT
Apr 15, 1993	582	Last full day of data from 183-GHz radiometer
Apr 16–20, 1993	583–587	UARS in safhold mode; MLS scan stopped
Apr 24, 1993	591	183 GHz radiometer turned off, following failure of its mixer
Aug 9, 1993	698	Resumed moving switching mirror at sunrise via secondary commutator
Sep 18 – Oct 21, 1993	738–771	No limb scans much of this time because of UARS solar array problem
Dec 23, 1993 – Jan 25, 1994	834–867	Initial period of MLS scan slips
Jan 26, 1994	868	Start limb scanning in reverse (upward) direction: more motor torque
Jan 28 – Feb 4, 1994	870–877	MLS in safhold
Mar 1, 1994 – May 23, 1994	902–985	Reduced days of limb scans to conserve scan mechanism lifetime
Jul 5, 1994 – Aug 4, 1994	1028–1058	Limb scans on alternate days to conserve scan mechanism lifetime
Sep 19, 1994 – Oct 20, 1994	1104–1135	Limb scans on alternate days to conserve scan mechanism lifetime
Sep 23, 1994	1108	MLS moon scan for field-of-view calibration
Oct 1, 1994 – Jun 13, 1997	1116–2102	Intermittent limb scans during this period
Jun 14, 1997	2103	63 GHz radiometer turned off to reduce power drain on UARS
Jun 15, 1997 – 27 Jul 1999	2104–2876	Only occasional limb scans; MLS off most of the time
July 28, 1999	2877	MLS put in ‘standby’ to conserve lifetime for overlap with EOS Aura
Feb 2–12, 2000	3066–3076	MLS turned on for limited Arctic observations
Mar 27–30, 2000	3120–3123	MLS turned on for limited Arctic observations
Aug 18–25, 2001	3629–3636	MLS turned on for limited northern hemisphere observations and ODIN validation campaign

- volcanic, and methane influences, *J. Geophys. Res.*, *105*, 4471–4481, 2000.
- Froidevaux, L., et al., Validation of UARS Microwave Limb Sounder ozone measurements, *J. Geophys. Res.*, *101*, 10,017–10,060, 1996.
- Gunson, M. R., et al., The Atmospheric Trace Molecule Spectroscopy (ATMOS) experiment: Deployment on the ATLAS Space Shuttle missions, *Geophys. Res. Lett.*, *23*, 2333–2336, 1996.
- Harries, J. E., et al., Validation of measurements of water vapour from the halogen occultation experiment (HALOE), *J. Geophys. Res.*, *101*, 10,205–10,216, 1996.
- Harris, N., R. Hudson, and C. Phillips, Assessment of trends in the vertical distribution of ozone, *Tech. rep.*, World Meteorological Organization, Geneva Switzerland, 1998, SPARC Report 1.
- Isaacson, E., and H. B. Keller, *Analysis of Numerical Methods*, Dover, 1994.
- Jarnot, R. F., R. E. Cofield, J. W. Waters, and D. A. Flower, Calibration of the Microwave Limb Sounder on the Upper Atmosphere Research Satellite, *J. Geophys. Res.*, *101*, 9957–9982, 1996.
- Khosravi, R., G. P. Brasseur, A. K. Smith, D. W. Rusch, J. W. Waters, and J. M. Russell, III, Significant reduction in the stratospheric ozone deficit using a three-dimensional model constrained with UARS data, *J. Geophys. Res.*, *103*, 16,203–16,219, 1998.
- Ko, M. W. K., and N. D. Sze, Diurnal variation of ClO: Implications for the Stratospheric Chemistries of ClONO<sub>2</sub>, HOCl, and HCl, *J. Geophys. Res.*, *89*, 11,619–11,632, 1984.
- Kumer, J. B., et al., Comparison of correlative data with HNO<sub>3</sub> version 7 from the CLAES instrument deployed on the NASA Upper Atmosphere Research Satellite, *J. Geophys. Res.*, *101*, 9621–9656, 1996.
- Lahoz, W. A., et al., Validation of UARS microwave limb sounder 183 GHz H<sub>2</sub>O measurements, *J. Geophys. Res.*, *101*, 10,129–10,149, 1996.
- Liebe, H. J., P. W. Rosenkranz, and G. A. Hufford, Atmospheric 60-GHz oxygen spectrum: New laboratory measurements and line parameters, *J. Quant. Spectrosc. Radiat. Transfer*, *48*, 629–643, 1992.
- Lipson, J. B., M. J. Elrod, T. W. Beiderhase, L. T. Molina, and M. J. Molina, Temperature dependence of the rate constant and branching ratio for the OH + ClO reaction, *J. Chem. Soc. Faraday Trans.*, *83*, 2665–2673, 1997.
- Livesey, N. J., J. W. Waters, R. Khosravi, G. P. Brasseur, G. S. Tyndall, and W. G. Read, Stratospheric CH<sub>3</sub>CN from the UARS Microwave Limb Sounder, *Geophys. Res. Lett.*, *28*, 779–782, 2001.
- MacKenzie, I. A., R. S. Harwood, L. Froidevaux, W. G. Read, and J. W. Waters, Chemical loss of polar vortex ozone inferred from UARS MLS measurements of ClO during the Arctic and Antarctic springs of 1993, *J. Geophys. Res.*, *101*, 14,505–14,518, 1996.
- Manney, G. L., et al., Comparison of satellite ozone observations in coincident air masses in early November 1994, *J. Geophys. Res.*, *106*, 9920–9943, 2001.
- Massie, S. T., X. X. Tie, G. P. Brasseur, R. M. Bevilacqua, M. D. Fromm, and M. L. Santee, Chlorine activation during the early 1995–1996 Arctic winter, *J. Geophys. Res.*, *105*, 7111–7131, 2000.
- Michelsen, H. A., et al., ATMOS version 3 water vapor measurements: Comparisons with observations from two Lyman-alpha hygrometers, MkIV, HALOE, SAGE II, MAS and MLS, 2002, in press.
- Miloshevich, L. M., H. Vömel, A. Paukkunen, A. J. Heymsfield, and S. J. Oltmans, Characterization and correction of relative humidity measurements from Vaisala RS80-A radiosondes at cold temperatures, *J. Atmos. Oceanic Technol.*, *18*, 135–156, 2001.
- Nedoluha, G. E., et al., A comparative study of mesospheric water vapor measurements from the ground-based water vapor millimeter-wave spectrometer and space-based instruments, *J. Geophys. Res.*, *102*, 16,647–16,661, 1997.
- Pickett, H. M., Effects of velocity averaging on the shapes of absorption lines, *J. Chem. Physics*, *73*, 6090–6094, 1980.
- Pickett, H. M., R. L. Poynter, and E. A. Cohen, Submillimeter, millimeter and microwave spectral line catalog, *Tech. rep.*, Jet Propulsion Laboratory, 1992, 80-23, Rev. 3.
- Pumphrey, H. C., Validation of a new prototype water vapor retrieval for the UARS Microwave Limb Sounder, *J. Geophys. Res.*, *104*, 9399–9412, 1999.
- Pumphrey, H. C., and S. Bühler, Instrumental and spectral parameters: Their effect on and measurement by microwave limb sounding of the atmosphere., *J. Quant. Spectrosc. Radiat. Transfer*, *64*, 421–437, 2000.
- Pumphrey, H. C., and R. S. Harwood, Water vapour and ozone in the mesosphere as measured by UARS MLS, *Geophys. Res. Lett.*, *24*, 1399–1402, 1997.
- Pumphrey, H. C., D. Rind, J. M. Russell, III, and J. E. Harries, A preliminary zonal mean climatology of water vapour in the stratosphere and mesosphere, *Adv. Space. Res.*, *21*, 1417–1420, 1998.
- Pumphrey, H. C., H. L. Clark, and R. S. Harwood, Lower stratospheric water vapor as measured by UARS MLS, *Geophys. Res. Lett.*, *27*, 1691–1694, 2000.
- Read, W. G., L. Froidevaux, and J. W. Waters, Microwave Limb Sounder measurements of stratospheric SO<sub>2</sub> from the Mt. Pinatubo volcano, *Geophys. Res. Lett.*, *20*, 1299–1302, 1993.
- Read, W. G., J. W. Waters, D. A. Flower, L. Froidevaux, R. F. Jarnot, D. L. Hartman, R. S. Harwood, and R. B. Rood, Upper-tropospheric water vapor from UARS MLS, *Bull. Amer. Meteorol. Soc.*, *76*, 2381–2389, 1995.
- Read, W. G., et al., UARS Microwave Limb Sounder upper tropospheric humidity measurement: Method and validation, *J. Geophys. Res.*, *106*, 32,207–32,258, 2001.
- Reber, C. A., C. E. Trevathan, R. J. McNeal, and M. R. Luther, The Upper Atmosphere Research Satellite (UARS) mission, *J. Geophys. Res.*, *98*, 10,643–10,647, 1993.
- Ricaud, P., J. de la Noë, B. J. Connor, L. Froidevaux, J. W. Waters, R. S. Harwood, I. A. MacKenize, and G. E. Peckham, Diurnal variability of mesospheric ozone as measured by the UARS Microwave Limb Sounder instrument: Theoretical and ground-based validations, *J. Geophys. Res.*, *101*, 10,077–10,089, 1996.
- Ricaud, P., M. P. Chipperfield, J. W. Waters, J. M. Russell, III, and A. E. Roche, Temporal evolution of chlorine monoxide in the middle stratosphere, *J. Geophys. Res.*, *105*, 4459–4469, 2000.

- Rodgers, C. D., Retrieval of atmospheric temperature and composition from remote measurements of thermal radiation, *Rev. Geophys.*, *14*, 609–624, 1976.
- Rodgers, C. D., Characterisation and error analysis of profiles retrieved from remote sounding measurements, *J. Geophys. Res.*, *95*, 5587–5595, 1990.
- Rodgers, C. D., *Inverse methods for atmospheric science, theory and practice*, World Scientific, 2000.
- Santee, M. L., W. G. Read, J. W. Waters, L. Froidevaux, G. L. Manney, D. A. Flower, R. F. Jarnot, R. S. Harwood, and G. E. Peckham, Interhemispheric differences in polar stratospheric HNO<sub>3</sub>, H<sub>2</sub>O, ClO, and O<sub>3</sub>, *Science*, *267*, 849–852, 1995.
- Santee, M. L., G. L. Manney, W. G. Read, L. Froidevaux, and J. W. Waters, Polar vortex conditions during the 1995–96 Arctic winter: MLS ClO and HNO<sub>3</sub>, *Geophys. Res. Lett.*, *23*, 3207–3210, 1996.
- Santee, M. L., A. Tabazadeh, G. L. Manney, R. J. Salawitch, L. Froidevaux, W. G. Read, and J. W. Waters, UARS MLS HNO<sub>3</sub> observations: Implications for Antarctic polar stratospheric clouds, *J. Geophys. Res.*, *103*, 13,285–13,314, 1998.
- Santee, M. L., G. L. Manney, L. Froidevaux, W. G. Read, and J. W. Waters, Six years of UARS Microwave Limb Sounder HNO<sub>3</sub> observations: Seasonal, interhemispheric, and interannual variations in the lower stratosphere, *J. Geophys. Res.*, *104*, 8225–8246, 1999.
- Santee, M. L., G. L. Manney, N. J. Livesey, and J. W. Waters, UARS Microwave Limb Sounder observations of denitrification and ozone loss in the 2000 Arctic late winter, *Geophys. Res. Lett.*, *27*, 3213–3216, 2000.
- Shippony, Z., and W. G. Read, A highly accurate Voigt function algorithm, *J. Quant. Spectrosc. Radiat. Transfer*, *50*, 635–646, 1993.
- Siskind, D. E., L. Froidevaux, J. M. Russell, III, and J. Lean, Implications of upper stratospheric trace constituents observed by HALOE for O<sub>3</sub> and ClO from 1992 to 1995, *Geophys. Res. Lett.*, *25*, 3513–3516, 1998.
- Solomon, P., J. Barrett, B. Connor, S. Zoonematkermani, A. Parrish, A. Lee, J. Pyle, and M. Chipperfield, Seasonal observations of chlorine monoxide in the stratosphere over Antarctica during the 1996–1998 ozone holes and comparison with the SLIMCAT three dimensional model, *J. Geophys. Res.*, *105*, 28,979–29,001, 2000.
- Tscherning, C. C., ed., *The Geodesist's Handbook*, vol. 58, Bureau Central De L'Association Internationale De Géodésie, 1984.
- van den Broek, M. M. P., A. Bregman, and J. Lelieveld, Model study of stratospheric chlorine activation and ozone loss during the 1996/1997 winter, *J. Geophys. Res.*, *105*, 28,961–28,977, 2000.
- vanVleck, J. H., and V. F. Weisskopf, On the shape of collision-broadened lines, *Rev. Mod. Phys.*, *17*, 227–236, 1945.
- Wang, H. J., D. M. Cunnold, L. Froidevaux, and J. M. Russell, III, A reference model for middle atmosphere ozone in 1992–1993, *J. Geophys. Res.*, *104*, 21,629–21,643, 1999.
- Waters, J. W., L. Froidevaux, W. G. Read, G. L. Manney, L. S. Elson, D. F. Flower, R. F. Jarnot, and R. S. Harwood, Stratospheric ClO and ozone from the Microwave Limb Sounder on the Upper Atmosphere Research Satellite, *Nature*, *362*, 597–602, 1993.
- Waters, J. W., et al., Validation of UARS Microwave Limb Sounder ClO measurements, *J. Geophys. Res.*, *101*, 10,091–10,127, 1996.
- Waters, J. W., et al., The UARS and EOS Microwave Limb Sounder (MLS) experiments, *J. Atmos. Sci.*, *56*, 194–217, 1999.
- Wu, D. L., and J. W. Waters, Gravity-wave-scale temperature fluctuations seen by the UARS MLS, *Geophys. Res. Lett.*, *23*, 3289–3202, 1996.
- Wu, J., and A. E. Dessler, Comparisons between measurements and models of Antarctic ozone loss, *J. Geophys. Res.*, *106*, 3195–3201, 2001.

---

Jet Propulsion Laboratory, MS 183–701, 4800 Oak Grove Drive, Pasadena, CA 91109–8099

Not yet; revised Not yet; accepted Not yet.

---

This preprint was prepared with AGU's L<sup>A</sup>T<sub>E</sub>X macros v4, with the extension package 'AGU++' by P. W. Daly, version 1.5g from 1998/09/14.