



Midlatitude tropospheric ozone columns derived from the Aura Ozone Monitoring Instrument and Microwave Limb Sounder measurements

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[1] Tropospheric ozone columns derived from differences between the Dutch-Finnish Aura Ozone Monitoring Instrument (OMI) measurements of the total atmospheric ozone column and the Aura Microwave Limb Sounder (MLS) measurements of stratospheric ozone columns are discussed. Because the measurements by these two instruments are not spatially coincident, interpolation techniques, with emphasis on mapping the stratospheric columns in space and time using the relationships between lower stratospheric ozone and potential vorticities (PV) and geopotential heights (Z), are evaluated. It is shown that this PV mapping procedure produces somewhat better agreement in comparisons with ozonesonde measurements, particularly in winter, than does simple linear interpolation of the MLS stratospheric columns or the use of typical coincidence criteria. The OMI/MLS derived tropospheric columns are calculated to be 4 Dobson units (DU) smaller than the sonde measured columns. This mean difference is consistent with the MLS (version 1.5) stratospheric ozone columns being high relative to Stratospheric Aerosol and Gas Experiment (SAGE II) columns by 3 DU. Standard deviations between the derived tropospheric columns and those measured by ozonesondes are 9 DU (30%) annually but they are just 6 DU (15%) in summer. Uncertainties in the interpolated MLS stratospheric columns are likely to be the primary cause of these standard deviations. An important advantage of the PV mapping approach is that it works well when MLS data are missing (e.g., when an orbit of measurements is missing). In the comparisons against ozonesonde measurements, it provides up to twice as many comparisons compared to the other techniques.

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1. Introduction

[2] Ozone measurements using ozonesondes have been offering important information on tropospheric ozone climatology over the past several decades including at midlatitudes [Logan, 1999] and in the tropics [Thompson *et al.*, 2003]. Restricted by the limited and uneven spatial distribution of their measuring locations, it is, however, difficult for ozonesondes to provide detailed horizontal ozone distribution

information globally for nonclimatological studies. Satellite instruments, on the other hand, despite the increasing advancement in satellite measurements and retrieval techniques, still have difficulty measuring tropospheric ozone with good accuracy and precision. The combination of satellite measurements from several instruments, however, increasingly offers a good alternative to obtain at least tropospheric ozone column information with better horizontal resolution. Such measurements could, for example, provide useful information on ozone production resulting from the movement of air pollution from one country to another.

[3] Tropospheric ozone columns have already proved valuable for the study of ozone enhancement associated with dynamical and chemical processes such as biomass burning and El Niño events [e.g., Fishman *et al.*, 1990; Ziemke *et al.*, 1998]. An effective way to derive tropospheric ozone columns from satellite data has been the tropospheric ozone residual method which calculates the tropospheric ozone residual by subtracting the stratospheric ozone column from the total ozone column [e.g., Fishman and Larsen, 1987; Fishman and Brackett, 1997; Chandra *et al.*, 2003]. The

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Total Ozone Mapping Spectrometer (TOMS) has been providing the total ozone data necessary for this calculation of the ozone residuals for more than two decades. The limitation of the technique primarily has been that the necessary stratospheric column measurements all the way down to the tropopause have not been available with sufficient regularity and/or accuracy. Most of the early work therefore focused on the tropical regions where reasonable assumptions could be made about relatively small or relatively persistent variations in the stratospheric ozone columns (e.g., a longitudinal stationary wave one) [Hudson *et al.*, 1995; Kim *et al.*, 1996; Hudson and Thompson, 1998]. Ozone residual methods have evolved with improvements in satellite measurement techniques especially for ozone in the lower stratosphere.

[4] Using stratospheric ozone columns calculated from the Stratospheric Aerosol and Gas Experiment (SAGE) measurements, Fishman *et al.* [1990] use the residual method to study the climatological distribution and seasonal cycle of tropospheric ozone columns in the region between 50°N and 50°S for a 9-a period between 1979 and 1987. The major shortcoming of the residuals obtained using SAGE measurements was relatively poor spatial and temporal coverage, since, each day, SAGE measured only 15 sunrise and 15 sunset events in two narrow bands each approximately 2° of latitude wide.

[5] More recently, Chandra *et al.* [2003] used the Upper Atmosphere Research Satellite (UARS) Microwave Limb Sounder (MLS) version 5 algorithm ozone retrievals to derive daily and monthly stratospheric ozone columns. The resulting tropospheric ozone columns then obtained by the residual method agreed well with the output of a three-dimensional model of chemistry in the tropics south of the equator for 1996–1997, showing similar zonal and seasonal characteristics. The ozone residuals derived by Chandra *et al.* [2003] were limited to latitudes within ±30° because the UARS MLS stratospheric retrievals only extended to 100 mbar and thus not to the tropopause at midlatitudes.

[6] The Convective Cloud Differential (CCD) technique is a variant of the tropospheric ozone residual technique that has also played a useful role in the characterization of tropospheric ozone columns in recent years (e.g., in the tropics [Ziemke *et al.*, 1998; Ziemke and Chandra, 1999; Chandra *et al.*, 2002]). Relying on the existence of high convective clouds near the tropopause, the technique calculates tropospheric ozone columns directly from TOMS measurements by taking the difference between the total ozone columns with TOMS reflectivities <0.2 and a nearby minimum in above cloud top ozone columns (determined from TOMS reflectivities >0.9) [Ziemke *et al.*, 1998]. The CCD technique has been applied to latitudes as high as 60° over the Pacific Ocean where there are frequent occurrences of deep convective clouds [Ziemke *et al.*, 2005]. Because of the sparseness of high reflectivity and nearby low reflectivity events, the CCD technique is likely to have difficulty producing a high-resolution global tropospheric ozone map on a daily or weekly basis.

[7] The recent launch of the Aura satellite has provided improved lower stratospheric and tropospheric ozone measurements. Aura carries two instruments in particular which

are resulting in improved global coverage of tropospheric ozone columns by the residual method. A new MLS instrument is providing good global coverage with improved ozone measurements in the lower stratosphere. Aura also carries a total ozone measuring instrument, the Dutch-Finnish Aura Ozone Monitoring Instrument (OMI), which is similar to the TOMS instruments but with much higher spectral resolution. OMI provides almost global coverage in a single day by observing in the nadir direction and scanning back and forth across the orbit track. MLS is looking forward along the orbit track, and there are therefore almost spatially coincident OMI and MLS measurements with time differences of less than 10 min. The most straightforward way to calculate tropospheric ozone columns is to subtract these MLS stratospheric ozone columns from the corresponding nadir OMI total ozone column measurements. A daily map of tropospheric ozone columns produced in this way, however, will have very large data gaps both because of the Aura orbit ground track separation of approximately 24.7° in longitude and because of cloud effects on the OMI measurements.

[8] Daily maps of tropospheric columns may be produced with smaller data gaps by interpolating the MLS derived stratospheric columns to the cross track locations of the OMI measurements. This may be accomplished, for example, by linear interpolation of MLS stratospheric ozone columns between consecutive orbits. However, in this paper we have emphasized a potential vorticity mapping approach to MLS interpolation at midlatitudes. This manuscript presents an assessment of the quality of the resulting OMI/MLS derived tropospheric ozone columns.

[9] A detailed description of the data and methodology is presented in section 2. The precision and accuracy of Aura MLS and the mapped stratospheric ozone columns are addressed in sections 3 and 4. Section 5 describes the estimation of column ozone between the tropopause and 215 mbar. Section 6 provides comparisons of the derived tropospheric ozone columns, and of MLS ozone columns in the lower stratosphere, against columns calculated from ozonesondes. Sections 7 and 8 briefly discuss possibilities for future improvements of these ozone columns.

2. Data and Methodology

2.1. Data

[10] The Aura spacecraft was launched in July 2004 for the study of atmospheric dynamics and chemistry. Its Sun-synchronous orbit has a 16-d repeat cycle. The Dutch-Finnish OMI is a nadir viewing, near-UV and visible spectrograph; it provides daily global maps of total column ozone with a pixel size of 13 × 24 km² at nadir and a swath width of 2600 km [Levelt *et al.*, 2006a, 2006b]. OMI total ozone columns have been retrieved by two methods: using a Differential Optical Absorption Spectroscopy (DOAS) algorithm [Veejkind *et al.*, 2006] and using an enhancement of the TOMS Version 8 algorithm [Bhartia and Wellemeyer, 2004], respectively. The OMI total ozone (OMTO3) products retrieved using the TOMS algorithm (available at http://disc.gsfc.nasa.gov/data/datapool/OMI/Level2_V002/) are used in this paper. Under cloudy conditions, variations in cloud height with respect to climatology

are estimated to produce a root-mean-squared error (rms) of $\sim 2\%$ in OMI TOMS total ozone products; however, errors may reach up to 10%, typically associated with the presence of very bright low clouds [Bhartia, 2006].

[11] The MLS is a forward looking limb sounding instrument which measures microwave thermal emission. Its wavelength characteristics allow the instrument to measure during both day and night, as well as to provide reliable measurements even in the presence of aerosols, thin cirrus or polar stratospheric clouds [Waters *et al.*, 2006]. MLS performs 240 limb scans per orbit, which provides coverage from 82°S to 82°N in latitude over a time span of 98.8 min. The ozone measurements for the standard product (which are based on the 240 GHz data) are performed along the suborbital track with a single profile spatial resolution of 6 km cross-track and approximately 200 km along track. On the basis of the MLS ozone vertical averaging kernels (full width at half maximum), the MLS version 1.5 ozone profiles have a vertical resolution ~ 3 km in the lower stratosphere [Livesey *et al.*, 2005]. MLS level 2, version 1.5, retrievals, which are reported on pressure levels which differ by $10^{1/6}$ in pressure, have been used for this study. The MLS ozone profiles possess their best accuracy from 146 mbar to 0.46 mbar, but there is evidence that the 146 mbar measurements are biased high by approximately 10% [Froidevaux *et al.*, 2006]. The expected result of updating to the newer MLS version 2.2 data set is discussed in section 8, on the basis of a limited number of provisional v2.2 MLS profiles.

2.2. Methodology Used to Derive Tropospheric Ozone Columns

2.2.1. Coincident Profiles

[12] Tropospheric ozone columns are first derived from coincident OMI and MLS measurements using the residual method. Except where otherwise stated, coincidence in this paper is defined as ± 12 h, and $\pm 1^\circ$ of latitude and $\pm 8^\circ$ of longitude for MLS, and $\pm 1.25^\circ \times 1.25^\circ$ longitude by latitude and latitude for OMI measurements. Thus, for example, for comparisons of coincident tropospheric columns against ozonesonde measurements both the OMI and the MLS measurements would have to satisfy their respective coincidence criteria with the ozonesonde measurements.

[13] Since the OMI only makes measurements during daytime and the MLS measures both day and night, it might be expected that tropospheric ozone columns with the best precisions would be produced by combining the OMI data and only the nearby MLS profiles with measurement time differences of less than 10 min. Although the tropospheric ozone columns produced with this more strict time coincidence criterion produce a reduction of approximately one Dobson unit (DU) in the mean differences versus ozonesonde data, these tropospheric ozone columns are not statistically different from the tropospheric ozone columns produced when nighttime MLS measurements are also included. Therefore the tighter temporal restriction has not been applied in our reported comparisons.

[14] A code provided by the MLS team is used for the computation of ozone columns from the MLS profile measurements. The routine incorporates the MLS retrieval assumption that the ozone mixing ratio varies linearly in log

(pressure) between the reported levels. The column ozone amount (Y) between two adjacent MLS pressure levels is:

$$Y = \text{const} \times \left[-\chi_2 P_2 \left(1 - \ln \left(\frac{P_1}{P_2} \right) \right) - \chi_1 P_1 \left(1 + \ln \left(\frac{P_1}{P_2} \right) \right) + \frac{(\chi_1 P_2 + \chi_2 P_1)}{\ln(P_1/P_2)} \right] \quad (1)$$

where χ_1 , P_1 and χ_2 , P_2 are the ozone mixing ratios (χ) and the pressures (P) at the lower and upper levels respectively. For ozone mixing ratios in ppmv and pressures in mbar, $\text{const} = 0.789352$ results in Y in Dobson units. This algorithm is recommended for calculations of MLS ozone columns because it is consistent with the assumptions made in the MLS retrieval algorithm.

[15] Two other stratospheric ozone column computation approaches are:

$$Y = \int \chi P d(\ln P) = \text{const} \times \frac{(\chi_1 P_1 + \chi_2 P_2)}{2} \ln \left(\frac{P_1}{P_2} \right) \quad (2)$$

$$Y = \int \chi dP = \text{const} \times \frac{(\chi_1 + \chi_2)}{2} (P_1 - P_2) \quad (3)$$

where the integration symbol here indicates integration over a layer between two reported MLS levels. These two equations are based on the assumptions that χ^P (equation (2)) and χ (equation (3)) respectively vary linearly between MLS levels with $\ln P$ and with P respectively. Compared to equation (1), equations (2) and (3) applied to the vertically gridded MLS profiles yield mean differences of $\pm 1\%$ in the stratospheric ozone columns: the use of equation (2) underestimates individual columns by $1.0 \pm 0.1\%$, and the use of equation (3) overestimates columns by an equal amount. The sensitivity of the MLS stratospheric ozone column amounts to the different integration approaches is due to the large vertical spacing (a factor of $10^{1/6}$ in pressure or ~ 2.7 km) between two adjacent MLS vertical levels.

[16] MLS profile data have been used down to 215 mbar in altitude unless the tropopause is located above this; in that case the tropopause pressure is used as the lowest level for the stratospheric column. When the tropopause is below 215 mbar, ozone from measurements by the Stratospheric Aerosol and Gas Experiment (SAGE II) have been used to fill in the region between 215 mbar and the tropopause (more details are provided in the mapping section). To avoid the influence of corrections for clouds on OMI total column retrievals, we used only total ozone columns from OMI obtained under clear sky conditions because OMI contains climatologically based adjustments for the ozone that lies below the clouds. The clear sky condition is defined here by a reflectivity of less than 10% based on the OMI 360 nm reflectivity provided in the level 2 data set, but results are compared against other reflectivity conditions. The possibility of a scan angle effect in the OMI measurements on the tropospheric ozone column derivation has been investigated by grouping the derived tropospheric ozone columns into three groups according to the OMI scan angles. No noticeable scan angle effects have been detected.

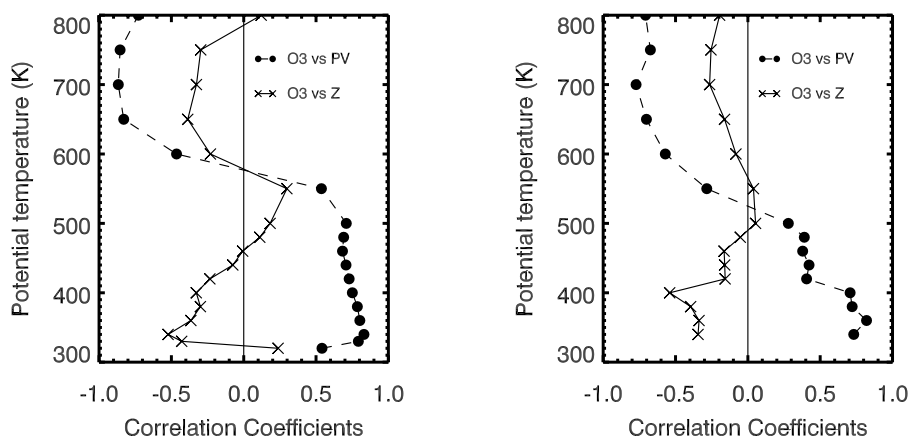


Figure 1. Correlations between potential vorticities (PV) and geopotential heights (Z) and MLS ozone mixing ratios at midlatitudes of the Northern Hemisphere on isentropic surfaces. The correlations shown are for 40–50°N for the months of (left) January and (right) September 2005.

2.2.2. Noncoincident Profiles

[17] Instead of requiring a coincident MLS profile within $\pm 8^\circ$ of longitude, tropospheric ozone columns also have been derived from the combination of OMI total ozone columns and potential vorticity (PV) mapped MLS stratospheric ozone columns. Potential vorticity mapping is capable of constructing a high-resolution stratospheric ozone field, and it has the potential to simulate the small-scale spatial structure in the stratospheric ozone columns up to about 250 km resolution (which is roughly the horizontal resolution of most of the meteorological assimilation models).

[18] Potential vorticity is a conserved quantity on isentropic surfaces during adiabatic transport. Using empirical relationships between PV and ozone to predict ozone on isentropic surfaces is one approach that has been used to study lower stratospheric dynamics [e.g., *Morgenstern and Marengo*, 2000; *Jing et al.*, 2004]. In our study, a two-predictor (PV and geopotential height (Z)) mapping has been applied to the MLS ozone measurements on isentropic surfaces. As shown in Figure 1, PV is highly correlated with ozone mixing ratio with the correlations changing from positive to negative at the height of the 550 K potential temperature surface at midlatitudes. Geopotential height (Z) shows lower correlations with ozone mixing ratio than with ozone number density, but on the lowest isentropic surfaces geopotential height is fairly well anticorrelated with ozone mixing ratio. The correlation coefficients of PV with ozone mixing ratio change with latitude, and they are fairly large for latitudes higher than 20°.

[19] MLS ozone has been mapped on 17 isentropic surfaces: 300, 310, 320, 330, 340, 360, 380, 400, 420, 440, 460, 480, 500, 550, 600, 650, and 700 K, which are approximately 1–2 km apart vertically. The interpolation of meteorological data to isentropic surfaces follows *Edouard et al.* [1997]. To be consistent with the assumption of the MLS retrievals, ozone mixing ratio (χ) was linearly interpolated on a logarithmic pressure scale. An isentropic ozone mapping relationship, expressed as in equation (4) below, is determined using linear regression of the interpolated natural logarithm of the MLS ozone mixing ratio ($\ln\chi$) and the

corresponding PV and geopotential height (Z) data on each of the isentropic surfaces:

$$\ln(\chi) = \alpha + \beta_1 \cdot PV + \beta_2 \cdot Z \quad (4)$$

Separate α , β_1 , and β_2 coefficients are determined from linear regressions for each 10° latitude band and using all the MLS measurements within $\pm 60^\circ$ longitude and ± 1.5 d of the desired location and time. Obtaining separate coefficients for different regions and different time periods was found to produce better mapping results particularly in winter. The meteorological data used in the mapping were either calculated or directly obtained from National Center for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis 1 data. Having determined the coefficients, the ozone mixing ratio for any location on each isentropic surface and any time is then determined using equation (4). The mapped ozone fields have a spatial resolution of $2.5^\circ \times 2.5^\circ$ (longitude by latitude) and a 6-h temporal resolution (i.e., the same as the NCEP/NCAR reanalysis data).

[20] Stratospheric ozone columns are obtained by adding up the column amounts in three regions: between the tropopause and 215 mbar, between 215 mbar (or the tropopause pressure if it is less than 215 mbar) and 700 K potential temperature (~ 18 mbar), and above 700 K. The 700 K isentropic surface has been chosen to be the upper boundary of the middle region because the NCEP meteorological data are sometimes missing at higher potential temperatures and because of the reduced validity of the mapping procedure in the upper stratosphere. The 215 mbar lowest boundary for the middle region has been chosen because it is the lowest level that has typically been used to calculate MLS stratospheric columns [e.g., *Ziemke et al.*, 2006]. Note that because potential temperature and pressure coordinates are being mixed here, the stratospheric column integrations involve conversions between two sets of levels and vertical interpolation.

[21] The stratospheric ozone column amounts in the uppermost layer are calculated from MLS profiles satisfying

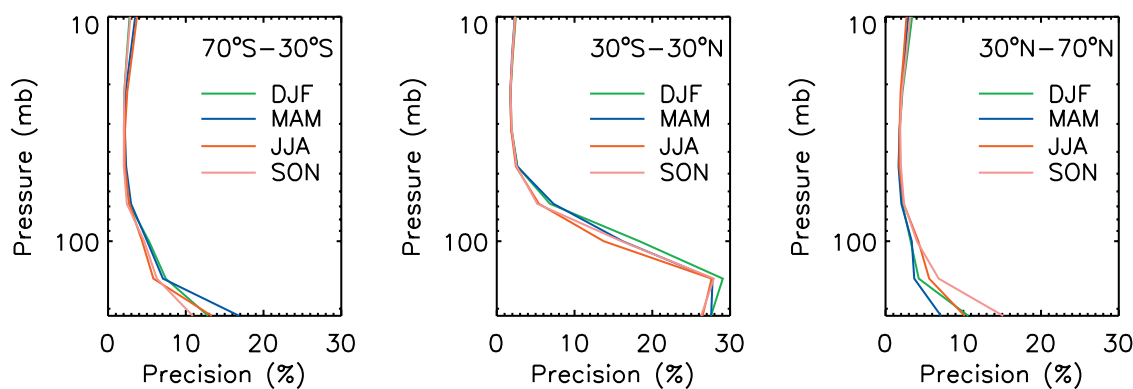


Figure 2. MLS ozone measurement precisions between 215 mbar and 10 mbar in 2005 estimated by season at 30°N – 30°S and at midlatitudes in the two hemispheres. Questionable profiles (MLS status flag $\neq 0$ or precision ≤ 0) are not included. The precisions are based on single profile precisions reported by the MLS ozone (version 1.5) level 2 software.

the coincidence criteria given in section 2.2.1. For the cases when the tropopause is below 215 mbar the column amounts in the lowest layer (tropopause to 215 mbar) are estimated by applying mapping to the Stratospheric Aerosol and Gas Experiment II (SAGE II) data. The necessary SAGE II ozone mapping relationships (in the form of equation (4)) are derived for each of the 12 months of the year on the basis of the SAGE II data from 1995 to 2005 (since the annual trend of stratospheric ozone has been small over that period); thus, for example, the January mapping relationships are derived using 11 a of January data.

[22] In the mapping approach, before subtracting the stratospheric ozone columns, the clear sky Level 2 OMI total ozone columns on the same day are averaged over an area of $2.5^{\circ} \times 2.5^{\circ}$ latitude by longitude in order to match the spatial resolution of the NCEP data.

[23] A more straightforward approach to obtaining stratospheric ozone columns as close as possible to the locations of the low reflectivity OMI total column measurements is to employ linear interpolation in longitude and latitude and to use only daytime MLS measurements. Tropospheric ozone columns obtained in this way are compared against those obtained by the coincidence and mapping procedures.

3. MLS Data Precision and Accuracy

[24] Because tropospheric ozone residuals represent small differences between two large numbers it is important to study the MLS ozone profile precisions and the accuracy of the MLS data. The MLS profile precisions based on the reported single-profile precisions, are presented in Figure 2 by season in the midlatitudes and tropics. In the upper stratosphere, the precisions are 2–3%. The precisions deteriorate with decreasing height in the lower stratosphere reaching 10–15% at 200 mbar at midlatitudes. The precisions expressed in percentage are smallest in winter and spring mostly because of higher mixing ratios in the lower stratosphere in those seasons.

[25] We have compared the MLS data against coincident SAGE II (version 6.2), SAGE III (version 3.0), and UARS Halogen Occultation Experiment (HALOE, version 19)

satellite data, as well as against the Southern Hemisphere Additional Ozonesonde (SHADOZ), the Climate Monitoring and Diagnostics Laboratory (CMDL), and the World Ozone and Ultraviolet Radiation Data Center (WOUDC) ozonesonde data at a total of 22 ozonesonde sites. The coincident measurements all possess a vertical resolution superior to that of the MLS measurements. The coincident measurements are therefore first interpolated to vertical levels differing by a factor of $10^{1/18}$ in pressure such that every third level coincides with an MLS retrieval level. The three mixing ratios closest to an MLS level are then averaged with log (pressure) weighting to provide the coincident profile values on the MLS vertical grid. This procedure is designed to be consistent with the MLS retrieval algorithm assumption that the ozone mixing ratio varies as log (pressure) between the MLS levels. The same vertical summation procedures have been used for all the data sets.

[26] Figure 3 shows the mean ozone differences in percentage between MLS and coincident data for midlatitudes and the tropics from August 2004 to July 2005. There is some indication that MLS is biased low between 1 and 3 mbar. However, these levels contribute little to the stratospheric ozone columns. From 3 mbar to approximately 100 mbar, the agreement between all the instruments is better than approximately 5%. Significant disagreement occurs below 100 mbar; however, the sondes suggest the MLS measurements are good to 10% at 146 mbar, and SAGE II measurements suggest differences larger than that only in the tropics where SAGE II values are known to be low in the troposphere [Wang *et al.*, 2002]. On the basis of ozonesonde comparisons, Wang *et al.* [2006] have shown that HALOE version 19 measurements are biased low below about two MLS layers above the tropopause, whereas SAGE II measurements are good to about 10% down to the tropopause. SAGE III measurements are less extensive in latitude than SAGE II and HALOE measurements, and Wang *et al.* [2006] have shown them to be biased high relative to SAGE II by 2–10%, depending on altitude. The most reliable ozone comparisons below 100 mbar for the selected latitude ranges should be against ozonesondes and against SAGE II at midlatitudes. On this basis, it seems that

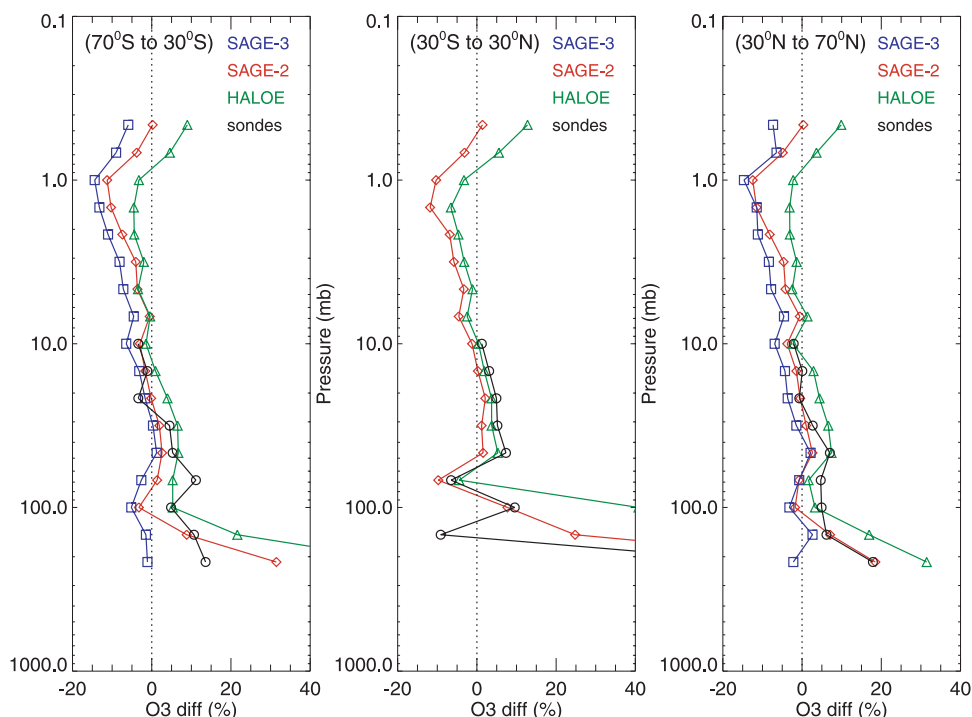


Figure 3. Mean ozone differences between MLS measurements and ozonesondes and SAGE II, SAGE III, and HALOE satellite measurements (MLS-coincident, expressed in % of the MLS values) in three latitude bands. The coincidence criteria are $\pm 1^\circ$ in latitude, $\pm 8^\circ$ in longitude, and ± 12 h. The data cover the period from August 2004 to July 2005. Ozonesonde data include measurements from 22 ozonesonde stations.

MLS measurements at 215 mbar are biased high by approximately 10–15% at midlatitudes.

[27] The MLS stratospheric ozone columns obtained by upward summation from the bottom of the 215, 146, and 100 mbar layers, respectively, are next compared against those from coincident SAGE II, SAGE III, and HALOE measurements. The means and standard deviations of the stratospheric ozone column differences are presented in Figure 4. The MLS column means are seen to be very consistent with the SAGE II measurements, and even the known tropospheric bias of the SAGE II measurements only significantly impacts the tropical columns when the 215 mbar layer is included. Excluding this layer the agreement is approximately 1 DU (about 0.4%). At midlatitudes when the 215 mbar layer is included the MLS columns are approximately 1% (2–3 DU) higher than the SAGE II columns. This difference reflects the bias of the MLS measurements at 215 mbar. Although the MLS mean differences with respect to SAGE III and HALOE measurements are significantly more variable, the standard deviations of the differences in the columns with respect to all three sets of measurements are $\leq 7\%$ at midlatitudes (and less than 5% with respect to SAGE II).

[28] The excellent agreement between the SAGE II and the MLS stratospheric columns suggests among other things that a consistent long-term record of the stratospheric ozone columns might be produced by extending the OMI/MLS derived tropospheric column time series backward in time

using the combination of SAGE II stratospheric columns obtained since 1984 and TOMS total columns.

4. Mapped Ozone Precision and Accuracy

[29] The prediction error of the ozone mapping has been estimated by comparing an ensemble of individually predicted profiles against observed MLS profiles using the mean square root of $press_p$ (predicted residual sums of squares):

$$press_p = \sqrt{\sum_{i=1}^n (y_i - \hat{y}_{i(i)})^2 / (n - 1)}, \quad (5)$$

where y_i is the i th case of the $n-1$ observed responses, and $\hat{y}_{i(i)}$ is the fitted value by deleting the i th case from $n-1$ cases, and then using the fitted regression function to obtain the predicted value $\hat{y}_{i(i)}$. The standard prediction error is calculated as one standard deviation of $press_p$ for all n cases. The solid lines in Figure 5 represent those prediction errors for 2 months of data in $40-50^\circ\text{N}$. The standard prediction error in each approximately 2.7 km thick layer (Figure 5, right) is typically less than 3 DU in summer and fall. However, for winter and early spring, the prediction errors are larger, mostly less than 5 DU on and above 50 mbar, and they increase downward reaching a maximum of approximately 7 DU at around 82 mbar. The green and red dashed

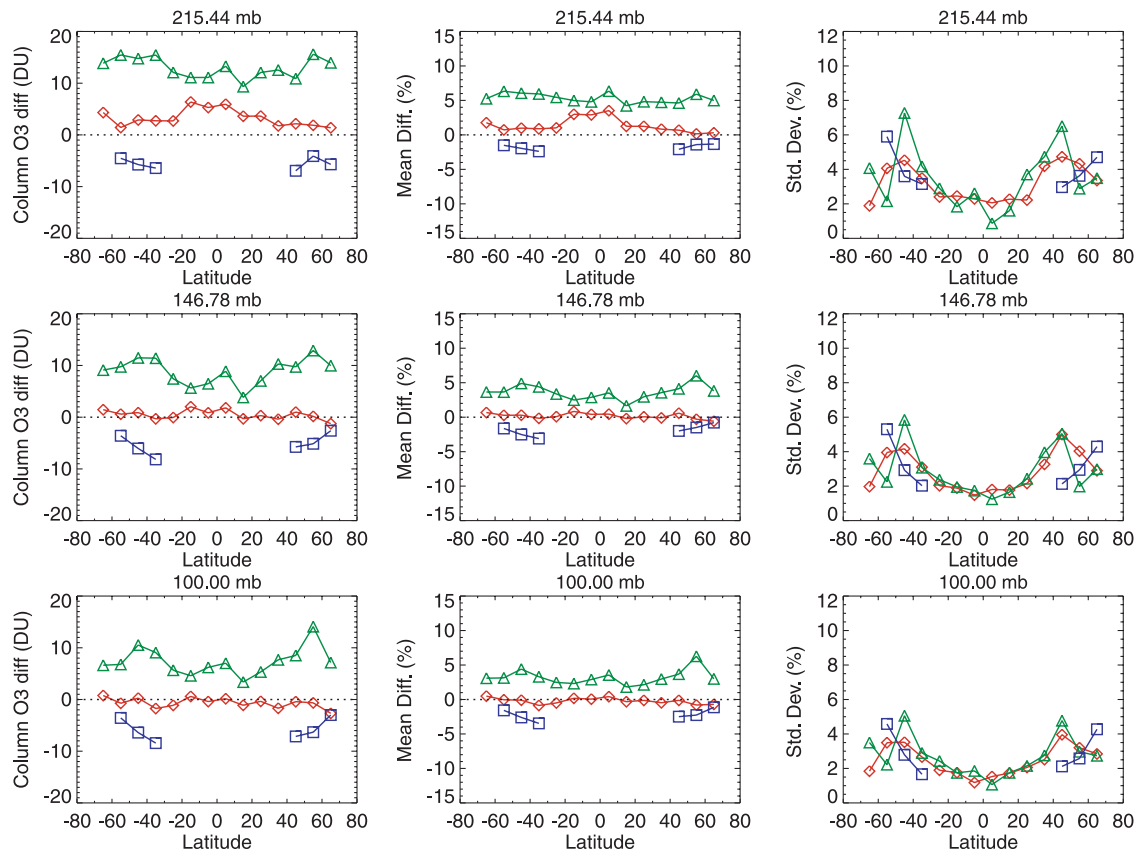


Figure 4. Mean differences (in Dobson units and %) and standard deviations of the differences (in %) of stratospheric ozone columns, in 10° latitude bands, calculated from MLS and coincident SAGE II (red), SAGE III (blue), and HALOE (green) measurements from August 2004 to July 2005. The lower boundaries of the columns are the bottoms of the MLS layers centered at 215.4, 146.8, and 100 mbar, respectively. As in Figure 3 the sign of the differences is MLS–other satellite measurements.

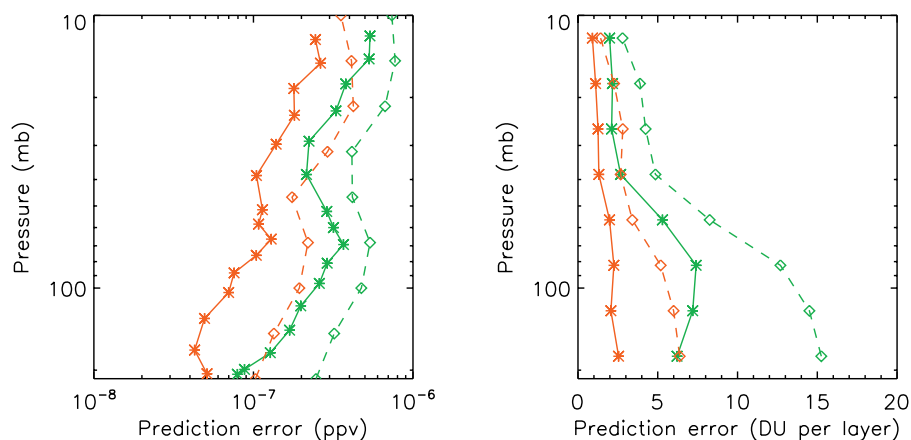


Figure 5. Mean prediction errors of stratospheric ozone mapping for 40–50°N in March (green lines) and September (red lines) 2005. The solid lines show standard deviations of the differences between measured MLS profiles and profiles which have been mapped to the locations of the measured profiles. The dashed lines (March in green and September in red) represent standard deviations of the differences between MLS measured profiles and the profiles averaged from measurements on the previous and succeeding orbits at the same latitude. (left) The solid lines were calculated on isentropic surfaces, and the average pressures of those surfaces have been used as the ordinate. (right) Prediction errors in Dobson units per layer between adjacent MLS pressure levels are shown.

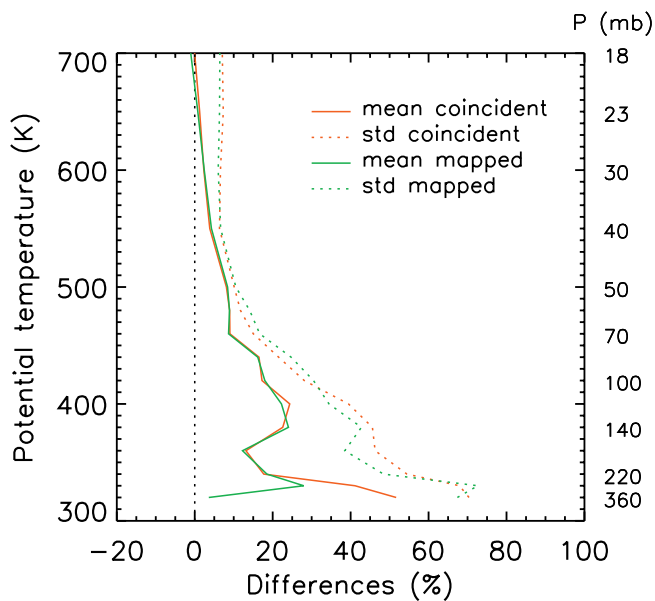


Figure 6. Mean and standard deviations of lower stratospheric differences (%) between mapped and coincident MLS and ozonesonde profiles at eight ozonesonde stations between 35 and 60°N over the period of December 2004 to November 2005. The coincidence criteria are ± 12 h in time, $\pm 8^\circ$ in longitude, and $\pm 1^\circ$ in latitude. The mapped ozone is predicted at the time and location of the ozonesonde measurements. The ozonesonde sites include Boulder, Huntsville, Narragansett, Trinidad Head, Uccle, Churchill, Hohenpeissenberg, and Payerne. The approximate pressures on selected isentropic surfaces corresponding to the potential temperatures are shown on the right-hand ordinate.

lines in Figure 5 represent, for March and September, respectively, the standard deviations of the differences between MLS measured profiles and the profiles obtained by averaging profiles measured on the previous and on the succeeding orbits at the same latitude. Because the solid line points are almost a factor of two less in standard deviation than the dashed line points, and because the MLS measurement errors are small (Figure 2), significant improvements are indicated by using mapping as opposed to using spatial linear interpolation to fill in for a missing orbit of MLS measurements. This indicates that mapping is clearly advantageous when interpolation is required over 24° of longitude.

[30] The mapped and the coincident ozone measurements from MLS are next compared with ozonesondes (Figure 6) between 35 and 60°N over the period of August 2004 to July 2005. MLS ozone is indicated to be unbiased relative to the sondes from 600 K to 700 K (approximately 29–18 mbar or 24–28 km), it is approximately 5% larger than the sondes from 460 K to 600 K (approximately 70–29 mbar or 19–24 km), and it is approximately 15% larger below this, down to the tropopause which is located at approximately 340 K (approximately 220 mbar or 11 km). These results are similar to the direct MLS/ozonesonde comparisons shown in Figure 3 for 30–70°N.

[31] The standard deviations of the differences shown in Figure 6 are approximately 5% above 600 K; this is consistent with the MLS measurement precisions shown in Figure 3 when combined with ozonesonde precisions of approximately 5%. Below 600 K the standard deviations increase to approximately 40% at the tropopause. This occurs not only because of the reduction in the precision of the MLS measurements but equally, and probably more, importantly because of small-scale (less than 500 km) atmospheric variability in the lower stratosphere. This affects the comparisons through the lack of complete coincidence between the profiles being measured. Similar standard deviations of the differences have been reported in SAGE II/ozonesonde comparisons in the lower stratosphere [Wang *et al.*, 2002].

[32] Figure 6 suggests that mapping produces no more than a small annually averaged reduction in the standard deviations of the differences (e.g., around 360 K (160 mbar)) compared to directly using MLS profiles within 8° of longitude. Mapping produces smaller mean differences only around 330 K (250 mbar). It may be that the nominal spatial resolution of the NCEP reanalysis data of approximately 250 km is not sufficient to allow mapping to provide more clearly superior results for this coincidence criterion.

5. Column Ozone Between the Tropopause and 215 mbar

[33] The small midlatitude lower stratospheric ozone column contributions from the layer between the lowest utilized level of the MLS data (215 mbar) and the tropopause are obtained using SAGE II ozone profile measurements. It has been shown that SAGE II version 6.2 ozone data and ozonesondes agreement in the mean is approximately 10% down to the tropopause and better than 5% between 15 (~120 mbar) and 20 km (~55 mbar) [Wang *et al.*, 2002, 2006]. Jing *et al.* [2004] have shown that mapped lower stratospheric SAGE II profiles have standard deviations of differences from Hohenpeissenberg ozonesondes of less than 30%. Estimated column amounts in this layer, when the tropopause is below 215 mbar, have been calculated four times daily, with the same spatial and temporal resolutions as the meteorological data set, using mapped SAGE II measurements. Figure 7 shows that the resulting seasonal mean ozone columns in this layer for December 2004 to November 2005 have a strong latitudinal and seasonal dependence. In midlatitude regions, the mean column decreases from ~9 DU at about 55° latitude to ~3 DU at about 35° latitude. The SAGE II column amounts in this layer differ from the corresponding columns measured by ozonesondes (at the Figure 6 sites) from August 2004 to November 2005 by -0.3 DU in the mean, with a standard deviation of the differences of 2.5 DU (32%). In all the calculations the tropopause pressure was obtained from the NCEP reanalysis data set.

[34] In 2005, for 21% of the tropospheric ozone columns derived using OMI/MLS coincidences the tropopause was located at a higher pressure than 215 mbar. The fraction of tropospheric ozone columns for which SAGE II mapping were used (still referred to as OMI/MLS tropospheric columns) had a strong latitude dependence, increasing from 1.5% for the 20–30° latitude band to approximately 45%

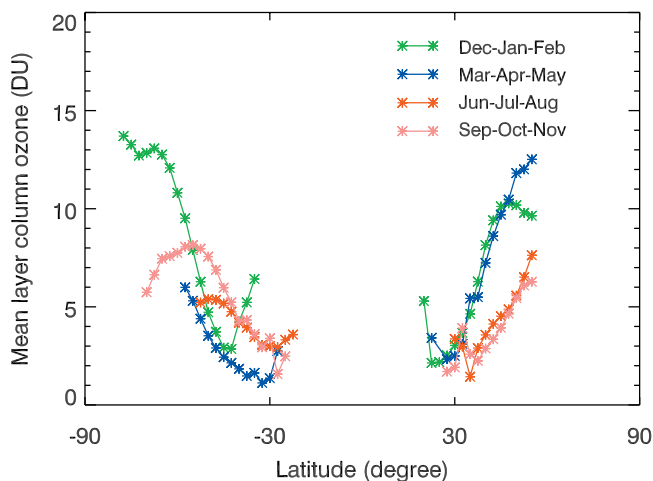


Figure 7. Seasonal mean ozone columns (DU) between the tropopause and 215 mbar over the time period from December 2004 to November 2005. These were calculated using equation (4) with the monthly mean coefficients at each isentropic level for each calendar month, having been derived using SAGE II measurements from January 1995 to August 2005. Only data when the tropopause was at a higher pressure than 215 mbar were included in the calculations of the seasonal means.

for the 40–50° latitude band and to 95% for the 70–80° latitude band.

6. MLS Lower Stratospheric Columns and Derived Tropospheric Columns

[35] The MLS lower stratospheric ozone columns between 700 K (~ 18 mbar) and 215 mbar (or the tropopause if this is above the 215 mbar level) and the derived tropospheric ozone columns have been compared separately against corresponding columns measured by ozonesondes at eight Northern Hemisphere midlatitude sites (the same sites as those used to make Figure 6) for the period from August 2004 to November 2005. The comparisons against the time series of lower stratospheric columns and tropospheric ozone columns measured by Hohenpeissenberg ozonesondes are presented in Figure 8. It is clear that the lower stratospheric ozone columns from the satellites have captured most of the variability seen by the ozonesondes. It is also clear that the most divergent comparisons in lower stratospheric columns occur in winter months (November to March) and that mapping produces improvements in the comparisons on several occasions (e.g., 31 January and 4 March). On the basis of the data from all eight stations, the lower stratospheric ozone columns have approximately 0.8 correlation coefficients with coincident total ozone column data, but the tropospheric ozone columns from ozonesondes and the satellite products have correlation coefficients of approximately 0.14 and 0.4 with total ozone. The high correlations between lower stratospheric ozone and total ozone are not surprising since the lower stratosphere contributes most of the ozone in the total column, but the variations in the total column explain a relatively small

proportion of the variations in tropospheric ozone columns at midlatitudes.

[36] Summaries of the statistics based on the eight ozonesonde station comparisons are presented in Tables 1 and 2. They indicate that both mapped and coincident MLS lower stratospheric ozone columns are larger by about 11 DU than the sonde measured columns. This is consistent with the mean differences shown in Figure 6, which also indicates that most of the offset arises from the region between 50 mbar (500 K) and the tropopause. Only during the spring are the mean differences, with respect to the sondes, between the mapped (8.7 DU) and the coincident (14.5 DU) MLS lower stratospheric columns significant at the 95% confidence level. Note, however, that the standard deviations of the differences between sondes and the mapped and coincident MLS columns in spring are similar. Mapping results in somewhat smaller standard deviations in winter and a more consistent bias in the tropospheric columns relative to the other seasons. Constraining the coincidence criteria to $\pm 4^\circ$ of longitude instead of $\pm 8^\circ$ reduces the mean tropospheric column offset in winter to -4.6 DU, but there are no other improvements in the comparisons.

[37] Linear interpolation in longitude and latitude produces annually averaged differences versus the ozonesondes over the August 2004 to November 2005 period of 12.0 ± 11.3 (standard deviation) for the lower stratosphere and -5.0 ± 10.1 in the troposphere. This is slightly worse than the mapped results, but it is in winter that the mapped results are definitely superior because the linear interpolation approach yields differences versus sondes of 17.8 ± 16.1 (versus 10.7 ± 13.0 using mapping) in the lower stratosphere and -9.5 ± 16.0 (versus -3.9 ± 12.8 using mapping) in the troposphere. A particular strength of the mapping technique compared to using linear interpolation or coincidence is that mapping is much less affected by missing MLS measurements. For example, in the comparisons with the sondes there were often twice as many comparisons that could be made using mapping than by using linear interpolation.

[38] The derived tropospheric columns are, in the mean, 4 DU smaller than the ozonesonde column measurements. Figure 4 showed that the MLS stratospheric columns are larger than SAGE II columns by approximately 3 DU. In the study by Ziemke *et al.* [2006], MLS columns were also shown to be ~ 4 DU high relative to OMI columns above convective clouds. Jing *et al.* [2006], however, reported an OMI/MLS tropospheric ozone column versus sonde column difference of 1 ± 9 DU (1 standard deviation), but this is because the MLS columns in their work were calculated using equation (2) for the column integration which, as was pointed out earlier, produces smaller stratospheric ozone column amounts by approximately 1%.

[39] The lower stratospheric mean offset of +11 DU relative to the sondes, combined with the -4 DU offset of the derived tropospheric columns, implies that the upper portion of the MLS stratospheric columns is low by approximately 7 DU. Relative to SAGE II (and SAGE III and HALOE), MLS indeed has shown smaller values of ozone above approximately 10 mbar (Figure 3) [see also Froidevaux *et al.*, 2006].

[40] The standard deviations of the derived tropospheric ozone column differences are significantly larger in the

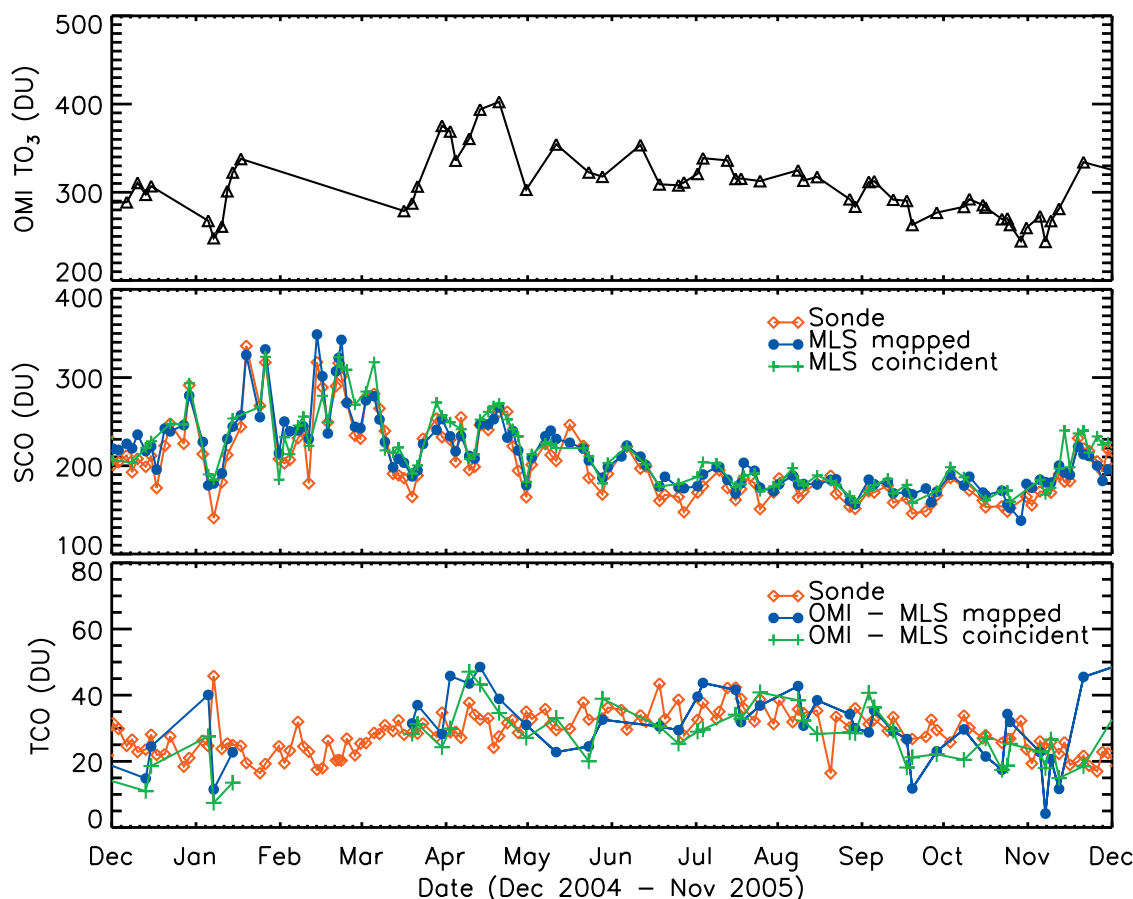


Figure 8. (middle) Time series comparisons of MLS stratospheric ozone columns (SCOs) between 215 mbar (or the tropopause if it is located above the 215 mbar level) and 700 K (~18 mbar) and (bottom) resulting derived tropospheric ozone columns (TCOs) against corresponding columns measured by Hohenpeissenberg (47.80°N, 11.02°E) ozonesondes from December 2004 to November 2005. Both coincident columns and columns mapped to the times and the location of the ozonesondes are shown. (top) For reference, clear sky OMI total ozone columns (TO₃) coincident with the ozonesonde measurements.

winter/spring seasons because of greater dynamical and hence stratospheric column variability at that time of year. Note that the tropospheric ozone column statistics in Table 2 are based upon a subset of the columns that are coincident with the ozonesonde measurements because of the OMI

total ozone measurement requirement of clear skies. For direct comparison purposes, essentially the same subset has been used for the mapped samples. As an additional check on the statistics, differences between the coincident and the mapped column results were directly calculated. These

Table 1. Means and Standard Deviations of Differences Between Mapped and Coincident MLS Lower Stratospheric Ozone Column Measurements (From 215 mbar, or the Tropopause if It Is Above the 215 mbar Level, to 700 K (Approximately 18 mbar)) and Similar Columns at Eight Ozonesonde Stations Located Between 35 and 60°N^a

Months	Sonde, DU	Difference (MLS Mapped–Sonde)			Difference (MLS Coinc.–Sonde)		
		Data	DU, %	Std (DU, %)	Data	DU, %	std (DU, %)
DJF	215.5	89	10.7 DU (5.6%)	13.0 DU (7.0%)	62	9.5 DU (5.1%)	17.7 DU (8.4%)
MAM	210.2	90	8.7 DU (4.9%)	13.8 DU (7.1%)	72	14.5 DU (7.2%)	13.4 DU (6.7%)
JJA	167.0	131	11.0 DU (6.9%)	7.7 DU (4.9%)	109	9.4 DU (5.9%)	8.2 DU (5.1%)
SON	164.8	242	10.9 DU (7.1%)	9.5 DU (5.9%)	73	11.3 DU (7.1%)	11.7 DU (6.8%)
All NH	180.9	553	10.5 DU (6.4%)	10.6 DU (6.1%)	427	11.1 DU (6.5%)	12.4 DU (6.7%)
Lauder	196.3	46	9.5 DU (5.2%)	9.6 DU (4.8%)	35	8.5 DU (4.6%)	9.4 DU (4.8%)

^aIn these calculations the differences were first expressed separately in percentages of the sonde columns and in Dobson units (DU), respectively. Annually averaged differences are also shown for Lauder (−45.04°S, 169.68°E) ozonesondes. The period of comparison was August 2004 to November 2005.

Table 2. Means and Standard Deviations of the Differences Between Tropospheric Column Ozone Values Calculated From Clear Sky OMI Minus Mapped or Coincident MLS Measurements (With the Addition of Mapped SAGE II Measurements When the Tropopause Was Below the 215 mbar Level) and Similar Columns at Eight Ozonesonde Stations Between 35 and 60°N^a

Months	Sonde, DU	Difference (MLS Mapped–Sonde)			Difference (MLS Coinc.–Sonde)		
		Data	DU, %	std (DU, %)	Data	DU, %	std (DU, %)
DJF	29.1	27	−3.9 DU (−12.0%)	12.8 DU (37.9%)	27	−7.5 DU (−24.3%)	11.3 DU (35.0%)
MAM	36.3	40	−1.9 DU (−4.8%)	11.5 DU (33.4%)	40	−5.1 DU (−15.7%)	11.8 DU (35.9%)
JJA	39.9	48	−3.8 DU (−9.0%)	5.4 DU (13.6%)	48	−4.2 DU (−10.4%)	5.9 DU (15.1%)
SON	30.6	73	−4.1 DU (−12.8%)	8.0 DU (29.1%)	73	−3.1 DU (−10.0%)	8.2 DU (30.3%)
ALL NH	34.0	188	−3.5 DU (−10.0%)	9.1 DU (28.5%)	188	−4.4 DU (−13.3%)	9.2 DU (29.6%)
Lauder	23.9	20	−6.1 DU (−24.9%)	5.6 DU (22.6%)	20	−2.8 DU (−12.5%)	6.8 DU (30.8%)

^aSeparate differences are shown for mapped and for coincident MLS stratospheric ozone columns. As for Table 1, separate calculations were made using DU and percentage differences. Annually averaged differences are also shown for Lauder ozonesondes.

mean differences were exactly equal to the differences to be expected from Tables 1 and 2, and the standard deviations were somewhat larger than those shown in Tables 1 and 2. The latter result indicates that there is considerable variability in the differences between the individual coincident and the mapped columns.

[41] Tables 1 and 2 also show mean annual differences against a relatively small number of Lauder ozonesondes. The results are not significantly different from the combined annual results from the eight northern hemisphere sites (labeled ALL). It is concluded that the overall conclusions of this paper based on the mostly northern hemisphere analysis are probably also applicable to the southern hemisphere.

[42] The clear sky condition is defined in this paper by a reflectivity less than 10%. Results have also been calculated for reflectivity values of up to 30%. Changing reflectivity thresholds from 10% to 30% produced 40% more tropospheric ozone column data in the NH, but the overall mean and standard deviation of the differences with ozonesondes remained about the same for the eight mid-latitude NH ozonesonde stations. Specifically, there was an about 0.7 DU (~3%) increase in the magnitude of the mean differences and an about 0.7 DU (~4%) increase in the standard deviation of the differences.

[43] Comparisons between OMI total columns and ground based total column measurements (e.g., by the Dobson instruments) show OMI values are higher by $0.4 \pm 0.5\%$ (G. Labow, private communication, 2007). In the work by *Visconti et al.* [2007] the accuracy and precision of OMI columns are given as 2% and 1%, respectively. Therefore the low bias (with respect to tropospheric ozonesonde measurements) in the derived tropospheric ozone columns almost certainly is related to the high bias in the MLS v1.5 stratospheric columns. In addition, the indicated precision of the OMI columns of approximately 1% suggests that the relatively large standard deviations (~10 DU) in the derived tropospheric ozone columns relative to the ozonesondes are associated with variations in the differences between the interpolated MLS lower stratospheric columns and the sonde columns. This is consistent with the standard deviations in the SAGE II/MLS column comparisons shown in Figure 4. However, there also may be some contributions to the differences produced by differing spatial resolutions of the satellite and sonde

measurements and by nonexact coincidence between the MLS and the ozonesonde measurements.

7. Use of Total Column Ozone as a Mapping Constraint

[44] It is well known that there is a close relationship between total ozone and synoptic conditions in the troposphere and lower stratosphere; troughs and crests of the upper troposphere are often temporally associated with high or low ozone concentration [*Shalamyanskiy and Romanshkina*, 1980]. In addition, the locations of arctic air, tropical air, and the midlatitude air are found to be, respectively, coincident with high, low, and intermediate total column ozone values [*Hudson et al.*, 2003, and references therein]. It has been noted that the passage of the upper tropospheric fronts is associated with a sharp increase or decrease of the total ozone value [*Hudson et al.*, 2003, and references therein].

[45] On the basis of these well established relationships, we have tested the use of total ozone columns as a third predictor in the relationship (equation (4)). So far this has only been tested in the mapping of SAGE II stratospheric ozone measurements. Using TOMS total columns as the third predictor is found to reduce the standard deviations of the stratospheric ozone column and tropospheric ozone column differences during winter relative to the ozone columns at Hohenpeissenberg in 1998 by about 4 DU. The addition of total ozone as a third predictor is expected to improve the winter period prediction for Aura MLS mapping as well, but such an approach needs further testing and careful application.

8. A Preliminary Estimate of MLS Version 2.2 Retrieval Results

[46] MLS version 2.2 provisional profiles were used for 15 d in 2004 and 2005 for comparisons versus midlatitude ozonesonde measurements. Using $\pm 1.25^\circ \times \pm 1.25^\circ$ longitude by latitude and the same day as coincidence criteria, 17 coincident profiles were found at 6 (out of 8) ozonesonde stations: Boulder, Narragansett, Trinidad Head, Uccle, Hohenpeissenberg, and Payerne. The comparison based on the 17 coincident profiles indicates that the MLS lower stratospheric columns (tropopause, or 215 mbar, to 700 K) produced from version 2.2 data have a mean offset of 9.4 DU

relative to the ozonesonde columns. This offset is about 2 DU less than the mean offset (11.5 DU) calculated for the version 1.5 data using the same ozonesonde profiles.

[47] A more extensive evaluation of stratospheric ozone columns from MLS v2.2 retrievals has been made by directly comparing the columns obtained using the two versions of MLS retrievals from the 15 d of measurements. The v2.2 total stratospheric columns are calculated to be a few DU less on average than the v1.5 columns. The standard deviations of the differences in these stratospheric columns between the two versions are approximately 4–5% (L. Froidevaux et al., Validation of Aura Microwave Limb Sounder stratospheric ozone measurements, submitted to *Journal of Geophysical Research*, 2007). Therefore use of the new version 2.2 MLS retrievals will probably result in OMI/MLS tropospheric ozone columns (which for MLS v1.5 were biased by 4 DU relative to the sondes) which are essentially unbiased with respect to tropospheric columns measured by ozonesondes, but the standard deviations of the differences are unlikely to change significantly except perhaps in summer.

9. Summary

[48] Procedures for combining Microwave Limb Sounder (MLS) stratospheric ozone column (above 215 mbar) measurements with Dutch-Finnish Ozone Monitoring Instrument (OMI) total atmospheric column ozone measurements to produce tropospheric ozone columns have been discussed. The mapping (referred to here as PV mapping) of the MLS stratospheric columns to the locations and times of the OMI measurements using relationships between ozone, potential vorticities (PV), and geopotential heights on isentropic surfaces over 3 d periods has been emphasized. Using this procedure with the NCEP/NCAR reanalysis meteorological data set, we find that the resulting tropospheric ozone columns are 4 DU low on average from August 2004 to November 2005 relative to measurements from eight northern hemisphere midlatitude ozonesonde sites. This has been shown to be consistent with the MLS (version 1.5) stratospheric columns above 215 mbar being high relative to Stratospheric Aerosol and Gas Experiment (SAGE II) columns by approximately 3 DU.

[49] In the lower stratosphere mapped MLS ozone columns between approximately 18 and 215 mbar (or the tropopause if it is above the 215 mbar level) have been calculated to have a standard deviation in the differences from the sonde measurements over the entire period of approximately 11 DU. On the basis of comparisons against other satellite measurements, as well as differences between mapped and individual coincident MLS profiles, uncertainties in the interpolated MLS measurements are most likely the principal contributors to these standard deviations. The standard deviations in the lower stratospheric column differences are mirrored in standard deviations in the differences between ozonesondes and OMI/MLS derived tropospheric columns of, for example, 12 DU in winter/spring and 6 DU in summer.

[50] Preliminary indications are that the 4 DU offset of the MLS/OMI derived tropospheric columns relative to ozonesondes will essentially disappear when version 2.2

MLS retrievals become generally available. The vertical integration procedure supplied by the MLS team should be used for obtaining stratospheric ozone columns from the MLS measurements. Other integration algorithms which are often used for the vertical integration result in column differences of approximately $\pm 1\%$ because of the MLS layer thickness of approximately 2.7 km.

[51] PV mapping of MLS columns has been shown to be especially effective for spatially interpolating over 24° of longitude, and it reduces the standard deviations of the lower stratospheric differences from ozonesonde measurements somewhat during winter compared to linear interpolation between MLS measurements or just using the nearest MLS measurement to the sonde location.

[52] The uncertainties (one sigma) in the individual OMI/MLS derived tropospheric columns, based on the ozonesonde comparisons, are approximately 35% and 15% in winter and summer, respectively. Therefore, for many applications, it may be best to use spatially or temporally averaged columns [e.g., *Jing et al.*, 2006]. A significant advantage of the mapping procedures is that it produces an increased number of averageable observations especially when there is missing MLS data. In addition, using a 30%, instead of a 10%, reflectivity as the condition for clear sky OMI measurements produced approximately 40% more tropospheric ozone column measurements without any significant increase in their mean differences and their standard deviations relative to the sondes.

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