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Summertime Tropospheric Ozone Columns from Aura OMI/MLS Measurements Versus Regional Model Results Over the United States

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Summertime tropospheric ozone columns from Aura OMI/MLS measurements versus regional model results over the United States

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[1] Ozone columns below 147 hPa are derived over the United States from September 2004 to August 2005 from the differences between clear-sky Aura OMI columns and coincident MLS columns. The mean difference from coincident ozonesonde measurements at four USA sites is 0.3 DU with an rms difference of 10.1 DU and a correlation coefficient of 0.67. Semimonthly patterns of the columns over the USA for the summer of 2005 have been produced. The observed columns, as well as Regional Air Quality Forecast (RAQAST) model columns, show high values over the southeastern USA and its surrounding oceans. Changes of these columns exceeding 6 DU in many places were observed between June 17–30 and July 1–16 and the changes reversed in the following two-week period. Comparisons against calculations from the RAQAST model, as well as correlations with geopotential height changes at 147 hPa, indicate that these changes were primarily related to dynamics. **Citation:** Jing, P., D. Cunnold, Y. Choi, and Y. Wang (2006), Summertime tropospheric ozone columns from Aura OMI/MLS measurements versus regional model results over the United States, *Geophys. Res. Lett.*, *33*, L17817, doi:10.1029/2006GL026473.

1. Introduction

[2] Studies of regional air quality over the United States have indicated an increasing trend of tropospheric ozone concentrations in the 1980s and 1990s [*Oltmans et al.*, 1998; *Lin et al.*, 2000]; studies have also shown the long-range transport of ozone precursors from Asia [e.g., *Jacob et al.*, 1999; *Wang et al.*, 2006]. As a primary source of HO_x radicals (OH and HO_2), which are the main oxidants of tropospheric photochemistry, ozone serves as one of the most critical agents that indirectly control the lifetimes of many chemical compounds. Monitoring of the tropospheric column ozone (TCO) can provide a useful way to assess the simulation and forecasting of ozone and related photochemistry in regional air quality models.

[3] Among the currently available techniques, satellite-borne remote sensing instruments are probably the most effective way to obtain the necessary global distribution of tropospheric ozone. Since *Fishman et al.* [1990], studies have used the tropospheric ozone residual (TOR) method to derive TCO by calculating the difference between the total column ozone from Total Ozone Mapping Spectrometer (TOMS) measurements and the stratospheric column ozone (SCO) from satellite-based instruments such as the Stratospheric Aerosol and Gas Experiment (SAGE) measurements [*Fishman et al.*, 1990], Microwave Limb Sounder (MLS) measurements [*Chandra et al.*, 2003, 2004], and Solar Backscattered Ultraviolet (SBUV) measurements [*Fishman et al.*, 1996]. However, SAGE stratospheric ozone profiles provide global coverage only about once per month, and the quality of SBUV ozone profiles deteriorates below 32 hPa. The Aura MLS instrument reliably measures ozone profiles from 0.46–147 hPa, although significant biases exist at 215 hPa and below, particularly at low latitudes [*Froidevaux et al.*, 2006; *Yang et al.*, 2005]. Moreover, the Aura Ozone Monitoring Instrument (OMI) is providing total ozone columns almost simultaneously.

[4] The objectives of this study are to derive tropospheric column ozone from the OMI and MLS measurements using the TOR method, and to interpret the variations in the derived TCO using comparisons against regional chemistry and transport model results over the United States. This study follows the methodology given by *Chandra et al.* [2003, 2004] to a significant extent (see also the OMI/MLS results in *Ziemke et al.* [2006]), with the emphasis here being on the midlatitude United States.

2. Data and Method

[5] The OMI and MLS on NASA's Aura Mission have been in full-up science operation since August 2004. The OMI Level 3 data products of total column ozone and reflectivity are used, which are produced by applying the TOMS Version 8 algorithm to OMI global mode measurements. They are provided on a $1^\circ \times 1.25^\circ$ (latitude by longitude) grid size globally each day. There are typically 3494 MLS Level 2 Version 1.5 along track profiles per day over the globe with horizontal resolution of ~150 km and vertical resolution of ~3 km in the upper troposphere and lower stratosphere.

[6] The tropopause, calculated according to the WMO 2 K/km lapse rate definition by the National Center for Environmental Prediction (NCEP) Climate Diagnostics Center, is generally located above the 150 hPa pressure surface between 40°N and 30°S in June–August 2005. This study focuses on the area between 25°N and 40°N over the USA, where the atmosphere below 150 hPa is almost entirely tropospheric. (MLS ozone retrievals have been shown to agree with corresponding ozonesondes and other satellite measurements within 10% between 0.46 and 147 hPa [*Livesey et al.*, 2005; *Yang et al.*, 2005]) This study therefore uses 147 hPa as an approximation to the tropopause level, and MLS ozone profiles are integrated from 147 hPa to 0.46 hPa to yield the equivalent SCOs. This approach avoids uncertainties introduced in defining the

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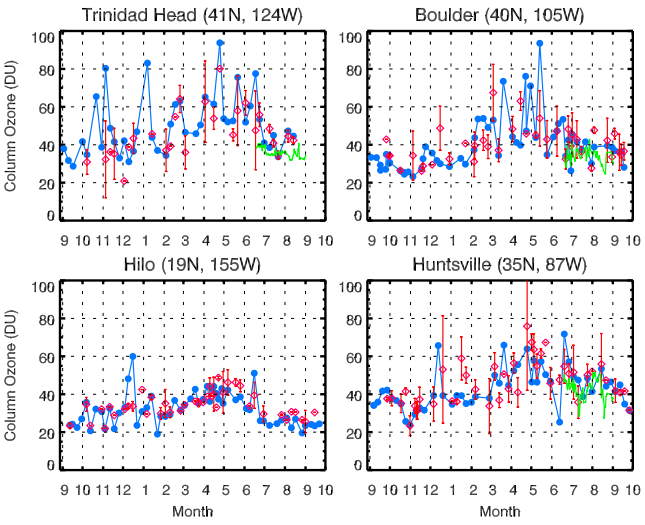

Figure 1. Column ozone below 147 hPa from ozonesonde measurements (blue dots) and from OMI/MLS residuals (red diamonds) over four USA stations from September 2004 to September 2005. Error bars, representing one standard deviation, are given for the OMI/MLS column ozone when more than three OMI/MLS residuals meet the coincidence criteria. The coincidence criteria are: ±24 hours in time, ±10° in longitude, ±5° in latitude around ozonesonde measurement locations. The green solid line represents the daily average RAQAST assimilations over the stations in summer 2005.

tropopause level, but it means that from this point on what we shall sometimes refer to as TCOs are in fact ozone columns between 147 hPa and the surface. This procedure has the additional advantage that it does not include the somewhat less reliable MLS measurements at 215 hPa.

[7] Corresponding to each MLS ozone column measurement above 147 hPa, a total ozone column is calculated by linear interpolation of the four nearest 1° × 1.25° grids of OMI level 3 ozone data. The time difference of the two measurements is approximately 7 minutes. Under clear-sky conditions, herein defined by OMI level 3 reflectivities less than 15%, the ozone column below 147 hPa is then obtained by subtracting the MLS 'stratospheric' column from the OMI total column. The choice of a 15% reflectivity cutoff is conservative, but increasing the cut-off to 30% yielded only approximately 20% more ozone columns over the USA. The derived OMI/MLS TCO is gridded with a 1° × 1.25° resolution using a Barnes interpolation scheme, which interpolates to a target grid point using a weighted (weighting exp(−d/D)) average of the TCOs surrounding the target point within an influence distance (D) of 250 km. Typically this includes eight TCOs with distances d to the target point.

[8] The Regional Air Quality Forecast (RAQAST) model predicts concentrations of ozone and its precursors over the USA and its surrounding oceans on 23 vertical layers from the surface to 10 hPa with a horizontal resolution of ~70 km. The present RAQAST model has been updated from that of *Choi et al.* [2005] to account for the stratospheric variability of ozone and its precursors. The new version of the RAQAST model extends to 10 hPa through the addition of three extra model layers. Also, biogenic emissions of hydrocarbons are no longer limited to the region south of 30°N and the boundary layer turbulence scheme has been changed from the Blackadar to the Eta scheme. The meteorological fields of the model are assimilated using the National Center for Atmospheric Research/Penn State MM5 based on the NCEP reanalysis data, plus surface and rawinsonde observations. Daily chemical boundary conditions are taken from global GEOS-CHEM simulations [*Bey et al.*, 2001]. Fossil fuel emissions of NO_x and CO over the USA are obtained from the 1999 US Environmental Protection Agency National Emission Inventory Version 2. Other emission inventories from combustion and industrial sources and emission algorithms for vegetation and soils are taken from the global GEOS-CHEM model [*Bey et al.*, 2001]. Forecasted chemical concentrations from the chemistry module are archived every hour and meteorological fields from the MM5 are archived every half hour. The RAQAST data, coincident with the OMI/MLS derived columns, are obtained by interpolation in time and space to the locations of the MLS profiles and are then binned onto the 1° × 1.25° grids also using the Barnes interpolation method.

3. Column Ozone Below 147 hPa From OMI/MLS Residual

[9] The clear-sky TCOs (below 147 hPa) have been derived from September 2004 to August 2005. Compared to the ozone columns below 147 hPa from ozonesonde observations over four USA stations, for example, Hilo (19°N, 155°W), Huntsville (35°N, 87°W), Boulder (40°N, 105°W), and Trinidad Head (41°N, 124°W), the derived OMI/MLS TCOs reveal similar seasonal trends. Both the sondes and OMI/MLS show the highest column ozone values in April and May (Figure 1). For individual stations from Hilo at lower latitude to Trinidad at higher latitude, the mean bias of the OMI/MLS TCO compared to the sondes ranges from 1.3 to −3.8 DU (or from 4% to −9%) and the rms differences range from 7.0 to 12.8 DU (or from 18% to 23%). Combining the four stations, the relative bias is +0.3 DU (or +1%), the rms is 10.1 DU (or 22%), and the correlation coefficient is 0.67. The OMI/MLS TCO generally agrees well with the sonde TCO except for springtime over Trinidad Head and Boulder. These two stations are under greater influence than the other two lower latitude stations from the springtime tropopause variability, which, often of smaller scale, may not be 'captured' by the coincidence criteria. Our study focuses on the summer months in 2005, when the stratospheric intrusion of ozone is relatively weak and the tropopause is close to the 147 hPa pressure level. For the summertime comparisons between OMI/MLS TCO and ozonesondes in Figure 1, the mean bias is +0.8 DU (+2%), RMS is 8.9 DU (23%), and correlation coefficient is 0.73. The RAQAST assimilated TCOs are also shown to be in good agreement with the ozonesondes for summer 2005 (Figure 1), with a bias of −6.2 ± 6.5 DU (−12.2 ± 12.8%) and correlation coefficient of 0.68.

[10] Uncertainty in the estimated TCOs due to differences between the thermal tropopause and the 147 hPa pressure level is investigated using the ozonesonde measurements (i.e., air pressure, temperature, and ozone profiles) for summer 2005. For the two stations at latitudes lower than 35°N (Hilo and Huntsville), the tropopause is usually located at 123 ± 18 hPa, and the actual tropospheric ozone columns are 1.5 ± 2.5 DU more than the columns below 147 hPa. For

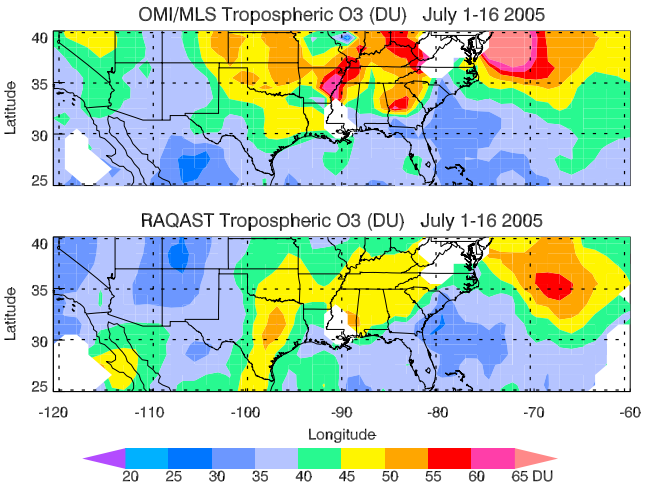

Figure 2. Time-averaged 'tropospheric' ozone columns (in DU) below 147 hPa for July 1–16, 2005 derived from (top) OMI/MLS residuals and (bottom) RAQAST modeling results.

Boulder and Trinidad Head, the tropopause is located at 160 ± 35 hPa, and the true tropospheric ozone columns are 5.0 ± 7.0 DU less than the columns below 147 hPa.

4. Comparison With the RAQAST Regional Modeling Results

[11] Monthly average clear-sky tropospheric ozone columns for mid-latitudes by TOMS and MLS measurements on the Upper Atmosphere Research Satellite (UARS) have been reported by *Chandra et al.* [2003, 2004]. The OMI and MLS instruments on Aura enable us to generate semimonthly TCO maps for the summer months in 2005 with smaller reflectivity (<15%) and resolution (250 km) than those (with reflectivity <20% and resolution ~500 km) given by *Chandra et al.* [2003, 2004].

[12] The semimonthly clear-sky OMI/MLS TCO maps over the USA in summer 2005, for example in July 1–16, 2005 (Figure 2, top), show maximum ozone regions over the northwestern Atlantic and over most of the eastern and mid-western states. The modeling results of TCO from RAQAST also show maximum ozone over these two regions (Figure 2, bottom). The correlation coefficient R between the two TCO maps is 0.55 (Figure 3). The high TCO levels over the southeastern states are associated with high RAQAST near-

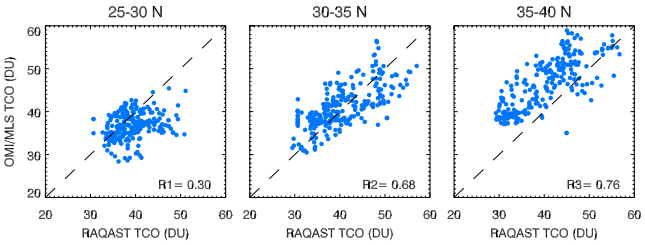

Figure 3. Scatter plots of the OMI/MLS versus RAQAST ozone columns below 147 hPa for July 1–16, 2005 in three latitude bands over the USA: 25°~30°, 30°~35°, 35°~40°. The dashed line is the perfect fit line.

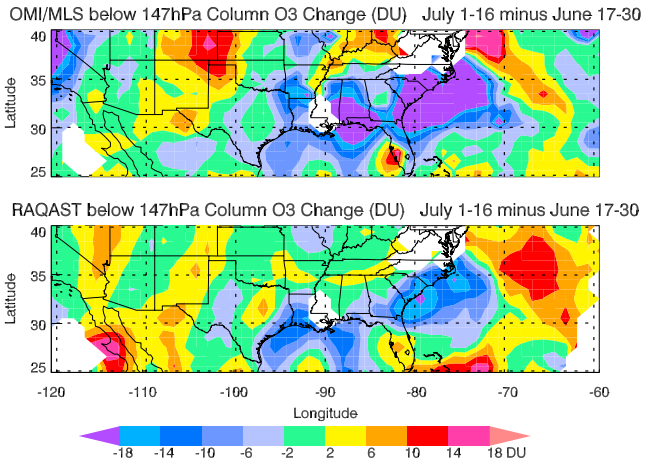

Figure 4. 'Tropospheric' column ozone differences (in DU) between July 1–17 and June 17–30, 2005 from the (top) OMI/MLS residual method and (bottom) RAQAST model.

surface (herein defined as the column below 750 hPa or ~2 km altitude) ozone levels (R~0.6) and high NOx concentrations (not shown). At latitudes greater than 35°N, the OMI/MLS TCOs are correlated with the RAQAST TCOs (R = 0.78), but they are ~+10 DU higher. This difference is not inconsistent with the −6 DU bias found in the limited number of RAQAST ozonesonde comparisons.

[13] The changes from June 17–30 to July 1–16 in the OMI/MLS and RAQAST TCOs show significant decreases (>6 DU) along the southeast coast of the USA and the Gulf of Mexico. Significant increases (>6 DU) are found over the northwestern Atlantic and over the western states (Figure 4). Opposite increasing and decreasing tendencies are found in GMAO (Global Modeling and Assimilation Office) geopotential height changes at 147 hPa (Figure 5). For the two lower mid-latitude bands (i.e., 25~30°N and 30~35°N), the correlation coefficient of the OMI/MLS TCO changes is

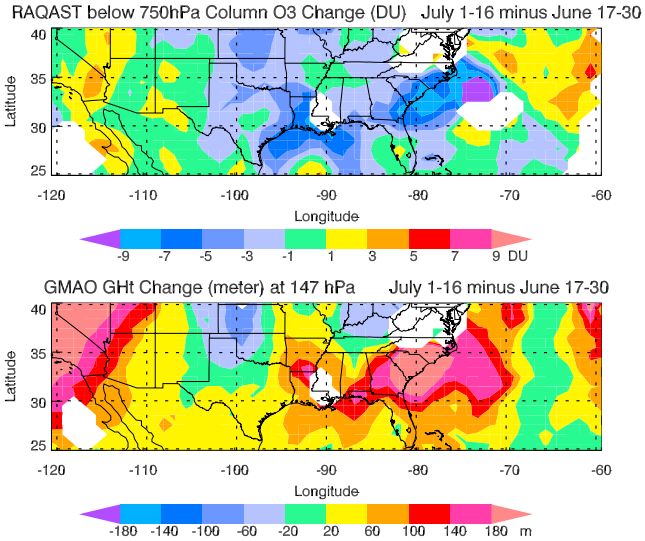

Figure 5. (top) RAQAST ground-to-750 hPa column ozone changes (in DU) and (bottom) GMAO geopotential height changes (in meters) at 147 hPa (d) between July 1–17 and June 17–30, 2005.

~0.62 with the RAQAST TCO changes, ~0.60 with RAQAST near-surface ozone changes, and ~0.57 with the geopotential height changes at 147 hPa. The corresponding correlation coefficients are 0.32, 0.02, and −0.50, respectively, for 35~40°N. The small correlation coefficient 0.02 indicates that the TCO changes at 35~40°N are not associated with the near-surface ozone changes but are related to dynamical processes. It has been noted that total column ozone is anti-correlated with geopotential height in the stratosphere [*Barsby and Diab*, 1995]. In this study, the TCO changes are anti-correlated with geopotential height changes at 147 hPa. A strong trough reaching into the deep south was noted on the geopotential height map at 147 hPa over the southeastern states in late June, but it was not found there in early July. The trough could bring ozone-rich air into the tropospheric column most likely from higher latitudes through quasi-horizontal advection or from higher altitudes through downward diabatic motions.

[14] Two weeks later, from July 1–16 to July 17–31, both the OML/MLS and the RAQAST simulations show that the increases and decreases have been mostly reversed, with TCO decreasing over the northwestern Atlantic and over the western states and increasing over the south and east coast (not shown), and they are associated with relevant reversed changes in the geopotential height at 147 hPa. The patterns of NOx concentrations near the surface (i.e., below 750 hPa) in the two periods are similar to the OMI/MLS and RAQAST TCO patterns, but NOx changes little between the two periods. This adds to the evidence that the observed semimonthly changes in the TCOs are mostly controlled by meteorological processes. Examination of the ground-to-316 hPa column ozone calculated by RAQAST shows similar semimonthly changes to those in the TCOs, indicating that the changes are unlikely to be dominated by changes in the small stratospheric portions of the columns.

5. Conclusions

[15] Clear-sky ozone columns below 147 hPa have been derived from Aura OMI and MLS measurements for September 2004 to August 2005. These ozone columns show good agreement with coincident ozone columns measured between June and August 2005 at four ozonesonde sites in the USA. The overall correlation coefficient is 0.73 and the mean difference is 1 ± 9(rms) DU. RAQAST regional chemical and transport model ozone columns compared with the three continental ozonesonde sites measurements show a low bias of 6 ± 7 DU and a correlation coefficient of 0.68.

[16] In the summertime USA, high OMI/MLS ozone columns below 147 hPa are found over the southeastern states and the northwestern Atlantic, and RAQAST modeling results indicate they are associated with high near-surface ozone levels. Both the OMI/MLS and RAQAST simulations show that the columns over the southeastern states decreased from late June to early July [see *Ziemke et al.*, 2006]. The semimonthly changes in the columns are correlated with ozone changes below 750 hPa (R ~ 0.6) at 25~35°, but the correlation coefficient, R, is nearly zero at 35~40° latitude. The column changes are shown to be anti-correlated with geopotential height changes (R ~ −0.50). The results demonstrate that meteorological conditions play an important role in the semimonthly changes in tropospheric ozone columns at midlatitudes in the USA in summer and that the changes have been captured by the OMI/MLS observations. Future research would be directed at deriving and validating tropospheric ozone column maps, based on accounting as accurately as possible for the location of the tropopause on shorter timescales (<two weeks). This should facilitate the employment of OMI/MLS TCO in tropospheric regional chemistry models.

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