

1           **Assessment and Applications of NASA Ozone Data Products**  
2           **Derived from Aura OMI/MLS Satellite Measurements in Context of**  
3           **the GMI Chemical Transport Model**

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19   **Abstract.** Measurements from the Ozone Monitoring Instrument (OMI) and Microwave Limb  
20   Sounder (MLS), both onboard the Aura spacecraft, have been used to produce daily global maps  
21   of column and profile ozone since August 2004. Here we compare and evaluate three strategies

to obtain daily maps of tropospheric and stratospheric ozone from OMI and MLS measurements: trajectory mapping, direct profile retrieval, and data assimilation. Evaluation is based upon an assessment that includes validation using ozonesondes and comparisons with the Global Modeling Initiative (GMI) chemical transport model (CTM). We investigate applications of the three ozone data products from near-decadal and inter-annual timescales to day-to-day case studies. Zonally averaged inter-annual changes in tropospheric ozone from all of the products in any latitude range are of the order 1-2 Dobson Units while changes (increases) over the 8-year Aura record investigated <http://eospsso.gsfc.nasa.gov/atbd-category/49> vary ~2-4 Dobson Units. It is demonstrated that all of the ozone products can measure and monitor exceptional tropospheric ozone events including major forest fire and pollution transport events. Stratospheric ozone during the Aura record has several anomalous inter-annual events including stratospheric warming split events in the Northern Hemisphere extra-tropics that are well captured using the data assimilation ozone profile product. Data assimilation with continuous daily global coverage and vertical ozone profile information is the best of the three strategies at generating a global tropospheric and stratospheric ozone product for science applications.

## **1. Introduction.**

Subtraction of stratospheric column ozone (SCO) from total ozone column, known as the tropospheric ozone residual method, yields a quantitative measure of tropospheric column ozone (TCO) from satellite remote sensing [*Fishman and Larsen*, 1987; 1990]. The original application of the method subtracted SCO obtained from Stratospheric Aerosol and Gas Experiment (SAGE) profiles from co-located Total Ozone Mapping Spectrometer (TOMS) total

column ozone. This residual concept has also been used with TOMS total ozone and SCO from Solar Backscatter Ultraviolet (SBUV) instruments [Fishman *et al.*, 2003] and from the Upper Atmosphere Research Satellite Microwave Limb Sounder (MLS) instrument [Chandra *et al.*, 2003]. Other methods to derive TCO from satellite measurements include a modified residual method incorporating TOMS-only total ozone [Hudson and Thompson, 1998], a UV radiance scan-angle method [Kim, *et al.*, 2001], a topography differencing method [Newchurch *et al.*, 2001], and cloud slicing using deep convective clouds [Ziemke *et al.*, 1998, 2009]. These techniques produce tropospheric ozone fields that have limited temporal or spatial coverage.

Our study focuses on ozone measurements from two Aura instruments, the Ozone Monitoring Instrument (OMI) and the Microwave Limb Sounder (MLS). These two instruments are currently operating and their measurements are being used in several new algorithms for deriving tropospheric and stratospheric ozone, both in column amounts and profiles. While the MLS instrument remains fully functional for measuring ozone and other trace gases, the OMI measurements are impacted by a “row anomaly” artifact which became large in January 2009 adversely affecting more than one third of the ozone measurements. Liu *et al.* [2010a, 2010b] uses an optimal estimation technique to determine ozone profiles from OMI radiances. This is a direct profile retrieval method using a single instrument and gives both stratospheric and tropospheric column ozone once the tropopause is specified. Although ozone profile retrievals are possible over the entire OMI record using this technique, the row anomaly adversely affected profiles after January 2009 (our study uses only ozone profiles through December 2008.) We also evaluate fields produced by data assimilation of OMI and MLS ozone measurements [Wargan *et al.*, 2013]. The third product investigated uses a trajectory mapping technique

[Schoeberl *et al.*, 2007] that estimates SCO fields by advecting MLS profile ozone from wind analyses and filling in subsequent SCO measurements between MLS orbits. TCO then follows from the trajectory mapping method by subtracting SCO from OMI total column ozone. Schoeberl *et al.* [2007] discuss improvements in daily TCO and SCO measurements from the OMI/MLS 2D Gaussian/linear interpolation method of Ziemke *et al.* [2006]. The assimilation and trajectory techniques both invoke MLS measurements that have better profile resolution near the tropopause for separating tropospheric from stratospheric column ozone compared to direct nadir profile retrieval.

The purpose of our study is to assess data quality and science application capability of these three gridded OMI/MLS ozone products. Assessment of the products includes comparisons with ozonesondes and ozone simulated using the Global Modeling Initiative (GMI) chemistry and transport model (CTM) as well as evaluation of the suitability of these products for science applications. The following sections 2 and 3 describe the datasets used and the GMI CTM. Section 4 provides a brief description of the three ozone data products evaluated in our study. Section 5 shows ozonesonde comparisons for validation of the products while section 6 discusses fundamental seasonal and spatial characteristics for these products. Section 7 describes applications for the products and finally section 8 provides a summary.

## **2. Description of relevant data sets.**

Ozone data from Aura OMI and MLS instruments are used to create daily maps of tropospheric column ozone (TCO) and profile ozone. The OMI and MLS ozone data have been archived as

level-2 (along-orbit swath) and level-3 (gridded for OMI) beginning in late August 2004. The Aura OMI is a nadir-scanning instrument. At visible and UV wavelengths OMI detects backscattered solar radiance to measure daytime total column ozone over the Earth with a resolution of  $13 \text{ km} \times 24 \text{ km}$  at absolute nadir. Documentation for the OMI instrument and data may be obtained from the webpage <http://eosps.gsfc.nasa.gov/atbd-category/49>. Our analyses use OMI version 8.5 ozone measurements which include several changes from version 8 including measured cloud pressures [Vasilkov *et al.*, 2008]. The sensitivity of OMTO3 to ozone varies from region to region (due to changes in surface reflectivity) and strongly with altitude, especially in the troposphere where retrieval efficiency on average varies from  $\sim 100\%$  in the stratosphere and upper troposphere to 40-70% at 3 km. The use of OMI spectra in the direct ozone profile retrieval slightly increases the retrieval efficiency in the lower troposphere [Liu *et al.*, 2010a].

The Aura MLS instrument measures vertical profiles of ozone tangent to the Earth's surface in the stratosphere and upper troposphere along the Aura orbital track beginning about 7 minutes before OMI for ascending orbit daytime measurements from MLS. This measurement time difference occurs because MLS looks forward along orbital path while OMI looks down. Because MLS detects microwave emission, this instrument measures ozone profiles during both daytime (ascending orbit) and nighttime (descending orbit). All of the ozone products which use MLS data include both day and night v3.3 measurements. Data quality and description of the MLS version 3.3 ozone profile product are discussed by Livesey *et al.* [2011].

Ozonesonde measurements are used for validation of the tropospheric ozone data products. This validation includes ozonesonde profile measurements from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC), Southern Hemisphere ADditional OZonesondes (SHADOZ), and Network for the Detection of Atmospheric Composition Change (NDACC). For consistency the tropopause pressure obtained from the NCEP 2K-km<sup>-1</sup> vertical lapse rate definition is applied for determining TCO and SCO for all of the products including ozonesondes.

### **3. Description of the GMI model.**

The GMI CTM includes a photochemical mechanism appropriate for the stratosphere and troposphere and is described by *Duncan et al.* [2007] and *Strahan et al.* [2007]. The emissions of trace gases and the aerosol fields used in the simulations are described by *Duncan et al.* [2008], except for the biomass burning emissions that are described by *van der Werf et al.* [2006]. The emission sources include industry/fossil fuel, biomass burning, biofuel combustions, and contributions from aircraft. The global emissions for year 2004 through year 2008 for the CTM are observationally derived daily emissions. Global emissions in the model for year 2008 are repeated for each of the following years 2009-2012. The meteorological input fields for the model are from Modern Era Retrospective analysis for Research and Applications (MERRA).

### **4. Description of the ozone data products.**

#### **4.1. Trajectory mapping.**

*Schoeberl et al.* [2007] describe a wind trajectory algorithm for producing high horizontal resolution maps of stratospheric and tropospheric column ozone. With this method the along-track profile v3.3 ozone from MLS each day is advected 2-days forward and 2-days backward to produce fields of stratospheric ozone profiles. These daily ozone profiles are then vertically integrated to produce high resolution ( $1^{\circ} \times 1.25^{\circ}$ ) gridded maps of SCO. The gridded SCO is then subtracted from coincident total column ozone from OMI to produce daily global maps of TCO. This method produces daily global maps that include regions associated with the dynamical wind jets. Vertical resolution from MLS is  $\sim 3$  km about the tropopause for separating ozone in the troposphere and stratosphere. The speed for the trajectory algorithm is fast when compared to direct profile retrieval (section 4.2) or data assimilation methods (section 4.3). The fields produced using this method incorporate MERRA assimilated winds, the same winds as used by the GMI CTM described in section 3. The current version of the trajectory mapping product includes an improved tropopause pressure definition, corrects for a time stamp error which affected daily time iteration steps, and flags out bad retrieval measurements for high resolution mode days (occurring one day out of every 32 days). We have improved the trajectory mapping product on several points from the original product of *Schoeberl et al.* [2007].

#### 4.2. Nadir profile.

In the OMI ozone profile algorithm the profile of partial ozone columns in Dobson Unit (DU) is retrieved at 24 layers from the surface to  $\sim 60$  km ( $\sim 2.5$ -km thick per layer) from BUUV radiances in the spectral region 270-330 nm using the optimal estimation technique [*Liu et al.*, 2010a,

2010b]. This retrieval scheme approaches an optimal solution by simultaneously and iteratively minimizing the differences between measured and simulated radiances and between retrieved and *a priori* state vectors, constrained by measurement error and the monthly and zonally mean LLM ozone profile climatology [McPeters *et al.*, 2007]. The retrieved ozone profiles have 4 to 7 layers in the troposphere, depending on the latitude. The horizontal resolution of our retrievals used here is 52x48 km<sup>2</sup> at nadir by co-adding 4 UV1 (8 UV2) pixels. The vertical resolution, estimated by Liu *et al.* [2010a], varies from 7-11 km in the troposphere and 10-14 km in the stratosphere. The retrieval errors (defined as the root sum square of precisions and smoothing errors) range from 1-6 % in the stratosphere to 6-35 % in the troposphere. The retrieval errors in tropospheric ozone columns are typically within 2-4 DU. For this study we use the daily data gridded to 2.5° longitude × 2° latitude. As the retrieval quality degrades due to the occurrence of OMI row anomaly, only retrievals from October 2004 through the end of 2008 are used in this study.

#### 4.3. Data assimilation.

The GMAO assimilated ozone product is generated by ingesting OMI v8.5 total column ozone and MLS v3.3 ozone profiles into the Goddard Earth Observing System version 5.7.2 (GEOS-5.7.2) assimilation system with several modifications listed below. GEOS-5.7.2 is an updated version of the global data assimilation system used to produce NASA's Modern Era Retrospective Analysis (MERRA reanalysis) and described by Rienecker *et al.* [2011]. It consists of an Atmospheric General Circulation Model (AGCM) and a statistical analysis module that combines 6-hourly meteorological and chemical forecasts from the AGCM with



observational data from conventional measurements and satellite-borne sounders to produce an analysis state. This AGCM is a version of the Fortuna general circulation model described in *Molod et al.* [2012]. The statistical analysis is done using the Gridpoint Statistical Interpolation (GSI) approach [Wu et al., 2002; Purser et al., 2003a, 2003b]. The assimilated product was generated at a horizontal resolution of  $2^\circ$  latitude by  $2.5^\circ$  longitude and at 72 vertical layers between the surface and 0.01 hPa. Depending on the height of the tropopause, about 25 to 35 of those layers are in the troposphere.

Ozone profiles from the MLS retrievals are given on 38 levels between 261 hPa and 0.0215 hPa. Prior to assimilation each profile is averaged in the vertical so that the mid-level average values of the mixing ratios are assimilated. The averaging eliminates some of the negative ozone values reported by MLS while preserving the integrated columns. In order to account for variable sensitivity of total ozone measurements by the OMI instrument we use the efficiency factor information (averaging kernels) provided with the data to weight the impact of these observations in the vertical. The GSI convolves the forecast ozone profile at each OMI observation location and the climatological *a priori* with their respective efficiency factors and subtracts their sum from the observed value of the total ozone column. By doing this, the dependence on OMI's *a priori* is largely removed in the assimilation. Although the row anomaly has time dependence that affects a growing number of rows over the Aura record, the assimilation uses the same OMI level-2 rows #2-24 over the entire record for consistency. The GEOS-5 AGCM transports the assimilated ozone using the assimilated meteorology but complete ozone chemistry is not accounted for. There is no model chemistry in the troposphere and only a dry deposition mechanism at the surface is applied. This approach works well

because the ozone time scale is long compared to the 6 hours between analysis times when the background field is corrected by observations. In the stratosphere the model applies time dependent (monthly) zonally symmetric ozone production and loss rates derived from a two-dimensional model as in *Stajner et al.* [2008]. This approach also works well for the same reason of comparatively long ozone lifetimes in the lower stratosphere which contributes the bulk of the total ozone column.

The main differences between the operational configuration of GEOS-5.7.2 and the system used in this study are: (1) The use of MLS ozone data instead of Solar Backscatter Ultraviolet observations, (2) the use of efficiency factors for OMI, (3) state dependent background error covariances similar to those used in *Stajner et al.* [2008], and (4) the absence of an ozone chemistry parameterization below the tropopause. Tests showed that the previous tropospheric parameterization did not significantly impact the accuracy of the ozone in the troposphere compared to ozonesonde measurements. A complete description of this system is given by *Wargan et al.* [2013]. Ozonesonde validation of the assimilated ozone profiles is shown by *Wargan et al.* [2013] and primarily focuses on the variability and structure of ozone in the upper troposphere and lower stratosphere. Information that data assimilation has of tropospheric ozone comes from OMI total column ozone which has reduced sensitivity in the low troposphere as mentioned in section 2.

## **5. Ozonesonde validation of the TCO products.**

Ozonesonde measurements are used to evaluate the TCO gridded products with the sonde station locations shown in Figure 1. Ozonesonde profiles were integrated from ground to the tropopause to derive daily TCO measurements that were then plotted against daily TCO from the three global data products and the GMI CTM simulation. For the remainder of our study these are abbreviated in figures and text as TRAJ, PROF, ASSIM, and GMI (corresponding to trajectory mapping, nadir profile retrieval, GMAO data assimilation, and the GMI CTM, respectively).

Figure 2 shows TCO for the three data products and CTM plotted versus ozonesonde TCO where all days of the years (years indicated in figure caption) and all latitudes for the sondes are included. In Figure 2 the TRAJ, ASSIM, and GMI TCO measurements all use the same tropopause pressures which come from the TRAJ data product. These tropopause pressures include quality flagging which filters out highly dynamical conditions such as tropopause folds which can produce large uncertainties in the derived column amounts. For the TRAJ TCO product an additional filter is applied for clouds (i.e, only OMI scenes with reflectivity less than 0.3 are used) to nearly eliminate (down to ~1-2 DU) artificial ghost column ozone in TCO. For the PROF TCO product the NCEP tropopause pressures were not filtered for either clouds or highly dynamical conditions. For these reasons the values for  $N$  in Figure 2 for the three data products are different and why  $N$  is similar between GMI and ASSIM.

In Figure 2 the variability for both product and sonde comes largely from seasonal cycles as well as variations with latitude/region for the many station locations included for each scatter plot. Statistical quantities listed in each of the four scatterplots include the mean offset (product or model minus ozonesonde) and the difference RMS. Time averages were removed from all data

products/GMI and ozonesondes prior to calculating difference RMS numbers. Figure 2 is useful in making baseline comparisons, but nothing can be said regarding latitudinal dependences.

Figure 3 shows the ozonesonde TCO comparisons of Figure 2 for separate latitude bands of  $20^{\circ}$  to  $40^{\circ}$  intervals as described in the Figure 3 caption. The top, middle, and bottom panels in Figure 3 plot differences from sonde values, difference RMS, and correlation between sonde values with each product as well as the simulation. The total number of ozonesonde profiles included in these calculations (“N”) is listed in the bottom panel of Figure 3.

The differences in Figure 3 (top) show that the ASSIM TCO and TRAJ TCO have the largest offsets relative to ozonesondes. Despite near-zero offset for GMI in Figure 2, latitude dependence of the offset for GMI in Figure 3 shows  $\sim 5$  DU shift from the tropics to high latitudes in both hemispheres. The difference RMS values in Figure 3 (middle) show consistency between the three data products and model in the tropics and subtropics but in mid-high latitudes, particularly in the SH the RMS numbers show greater spread between them. The TRAJ TCO product has largest difference RMS numbers. Figure 3 (bottom) shows that all three products and model have correlations  $\sim +0.8$  in the tropics. Outside the tropics the smallest correlations correspond to the TRAJ product, particularly in the SH. Largest correlation in Figure 3 for any product is about  $+0.85$ .

The ozonesonde TCO comparisons are extended by examining individual seasons of the year rather than entire year as in Figure 2. Figures 4-7 show scatter plots similar to Figure 2 but for the four seasons December-January-February (DJF), March-April-May (MAM), June-July-

August (JJA), and September-October-November (SON) for each of the data products and GMI CTM. There are several obvious characteristics in Figures 4-7 for the three data products and the CTM model. The assimilated TCO in Figure 4 in each season is biased low by several DU compared to the ozonesondes but the difference RMS numbers each season are small at only about 5-7 DU. For PROF TCO in Figure 5 offsets are only a few DU but there are many outliers, especially for the DJF. (These outliers drive the calculated difference RMS to nearly 10 DU.) The TRAJ TCO product in Figure 6 has the largest difference RMS and largest offsets relative to sondes, however even the largest difference RMS in MAM is still about 9 DU or less and the RMS numbers are only about 6 DU for JJA and SON seasons. For the GMI CTM in Figure 7 the offsets are smaller in each season compared to the data products, yet the difference RMS numbers are slightly larger each season compared to assimilated TCO.

In summary, we find that the ASSIM data product compares best overall with the ozonesondes. Outside the tropics the trajectory mapping product has the largest bias and difference RMS values. Statistics for the direct profile retrieval method are favorable in terms of a small average bias relative to sondes, yet there is larger scatter and higher difference RMS with sondes than for data assimilation. The GMI model has smallest offsets with the ozonesondes but has slightly larger difference RMS values in each season when compared with the ASSIM product.

## **6. Spatial variability of the ozone products.**

We next examine spatial characteristics of TCO and SCO for the three data products and GMI CTM as functions of season to identify consistencies and differences between them. Seasonal

mean maps of TCO and SCO for the products and CTM are plotted in Figures 8-11 where TCO and SCO in each figure are shown as part (a) and part (b), respectively. These seasonal averages were obtained from eight-year time series (January 2005 – December 2012) with the exception that the PROF measurements extend about four years from October 2004 through December 2008.

In the tropics, TCO for the data products and the GMI CTM in Figure 8 for DJF show a characteristic zonal wave-one variability with largest ozone in the Atlantic and smallest ozone over Indonesia. This wave-one zonal pattern occurs in all seasons with each product and the CTM as evident from Figures 9-11. For ASSIM TCO the Pacific values are larger and zonal amplitude of the wave-one pattern is smaller when compared to the other measurements and CTM. The year-round nature of the wave pattern in tropical TCO was first shown by *Fishman et al.* [1990] from combined TOMS and SAGE satellite measurements. Persistence of the wave-one is due to the year-round east-west tropical Walker circulation in the troposphere. TCO in the tropics maximizes in the Atlantic region during the SON season that corresponds to peak biomass burning in tropical South America and Africa. Despite intense biomass burning, *Sauvage et al.* [2007] suggests that the enhanced TCO in the Atlantic during SON is caused more by lightning rather than biomass burning.

All three data products and GMI CTM have smallest global mean TCO during DJF (Figure 8a). In MAM (Figure 9) TCO for all four have similar NH springtime buildup and horizontal structure. By JJA (Figure 10) all products and CTM have largest TCO in the NH extra-tropics with a corresponding buildup of TCO in the tropical/subtropical south Atlantic. The large TCO

in the NH extra-tropics during JJA is caused mostly by combination of stratosphere-troposphere exchange, anthropogenic pollution, and lightning [e.g., *Lelevield and Dentener*, 2000].

There are other basic characteristics of TCO and SCO in Figures 8-11. Spatial variability and seasonal dependence of TCO over the course of the year are almost identical in general sense between the three data products and GMI CTM. During DJF TCO in the SH polar region is as small as in the tropical western Pacific. SCO in Figures 8-11 for all three data products and CTM has small spatial variability throughout the tropics but large spatial variability and highest column amounts in the mid-high latitudes in both hemispheres during winter-spring. SCO during SON with all three products and CTM depict buildup of stratospheric ozone south of Australia lying northward along the edge of the SH stratospheric polar vortex.

Although TCO and SCO in Figures 8-11 have consistent geophysical features during all seasons there are also some inconsistencies. An example of this that we now examine more closely is the zonal variability of TCO and SCO in the tropics. It was noted for Figures 8-11 that TCO for all products in the tropics has a characteristic zonal wave-one pattern while SCO in the tropics exhibits small zonal variability. In Figure 12 we show TCO for the four products along the equator (Figure 12a) and corresponding SCO along the equator (Figure 12b) as time versus longitude. TCO in Figure 12a for all data products and CTM has a similar wave-one pattern that maximizes around September-October corresponding to the months of peak biomass burning over Africa and South America. The wave-one pattern of TCO for ASSIM is larger year-round in the Pacific by several DU when compared with the other data products and CTM.

There are some inconsistencies for SCO in Figure 12b between the three data products and GMI CTM. TRAJ SCO in Figure 12b during any month has very small zonal variability of only a few DU. This small zonal variability of SCO has been shown in previous studies using measurements from MLS, SAGE, and HALOE satellite instruments, as well as ozonesondes [e.g., *Fishman et al.*, 1990; *Ziemke et al.*, 1998; *Thompson, et al.*, 2003]. SCO in Figure 12b for September (i.e., when largest SCO occurs) has zonal variability of  $\sim 10$  DU for both the CTM and ASSIM, whereas zonal variability of SCO for PROF is  $\sim 5$  DU and only about 2-3 DU for TRAJ. We can conclude from Figure 12 that the TRAJ product may be slightly better (by several DU) at resolving zonal variability of tropical SCO compared to the other data products and CTM.

We include lastly a statistical evaluation of the spatial variability of the three TCO data products ASSIM, TRAJ, and PROF. This evaluation is listed in Tables 1-3 where a baseline reference TCO field for each of the four seasons was calculated by averaging corresponding seasonal means of the three data products and GMI CTM from Figures 8-11. Listed in Tables 1-3 for the three data products are spatial correlations, offsets, and difference RMS numbers, respectively. Positive correlations  $> 0.9$  in Table 1 suggest highly consistent spatial variability but there are also small correlations for the TRAJ and PROF products in the latitude bands  $30^{\circ}\text{N}$ - $60^{\circ}\text{N}$  and  $30^{\circ}\text{S}$ - $60^{\circ}\text{S}$  during winter. The mean offsets listed in Table 2 are for the most part self-explanatory with largest offsets for TRAJ followed secondly by ASSIM. The spatial difference RMS calculations for the data products in Table 3 are on average only about 1-2 DU. These low RMS numbers show that the three data products have consistent zonal and meridional variations of TCO during all seasons and in all latitude bands.



## 7. Applications using the ozone products.

The primary ozone product which we focus on for applications is ASSIM as this product compares well with ozonesondes, has daily global coverage of long record, and provides ozone profile information [e.g., *Wargan et al.*, 2013]. By comparison the TRAJ daily product does not provide ozone profile information, and the PROF daily ozone profile product extends only through year 2008 with coarse vertical resolution when compared with ASSIM. We discuss some applications in this section using these ozone products involving case studies and inter-annual variability.

### 7.1. Western Russia wildfires of year 2010 and pollution events

Record heat occurred over a broad region of western Russia in the summer of 2010 along with related intense uncontrolled wildfires. A prolonged anti-cyclonic circulation over western Russia caused sustained hot and dry conditions beginning in May 2010 and extending into early August 2010. This unusual meteorology created conditions favorable for wildfires. The breadth and intensity of the wildfires were the largest since Aura instruments began observations in the second half of 2004 [*Witte et al.*, 2011].

Figure 13 (top) plots monthly mean TCO for ASSIM, TRAJ, and GMI CTM for the burning region in western Russia over the eight-year record. Peak levels of ozone are observed in July 2010 for the two data products and the CTM. In the bottom panel of Figure 13 daily ozone profiles from ASSIM averaged over this region are plotted as log-pressure altitude versus day for

year 2010. The ozone profiles in Figure 13 are given as vertical column densities (in units DU-  
km<sup>-1</sup>) to indicate relative contribution to TCO as function of altitude. The ASSIM profile ozone  
suggests that the anomalous increase in TCO in July 2010 extended throughout the troposphere  
with most contribution coming from below ~6 km. This vertical distribution of tropospheric  
ozone in July coincided with the anti-cyclonic blocking high observed by *Witte et al.* [2011] that  
extended from the near surface through the free-troposphere.

Simulated increases in tropospheric ozone during the 2010 wildfire event suggest that anomalous  
meteorology is the main driver of these ozone enhancements. As described in section 3 the  
annual emissions for the GMI model for years 2009-2012 are a repeat of the 2008 emissions and  
therefore the July 2010 enhanced TCO for the model in Figure 13 cannot be attributed directly to  
a change in emissions. *Witte et al.* [2011] showed from satellite Outgoing Long-wave Radiation  
(OLR) anomalously high tropospheric warming during summer 2010 over western Russia  
compared to previous years 2003-2009. This suggests that meteorological change in summer  
2010 is related directly to these increases in tropospheric ozone. The anomalous warming in  
summer 2010 was associated with a persistent subsidence in the region and associated anti-  
cyclonic circulation.

*Kar et al.* [2010] showed that OMI/MLS tropospheric ozone has signatures of pollution in urban  
areas. However, while some urban regions showed local enhancement of tropospheric ozone,  
other urban regions did not. One important issue with detecting urban pollution ozone from OMI  
is that ozone sensitivity in the troposphere (which varies greatly with surface reflectivity  
conditions) decreases on average from about 100% in the mid-upper troposphere down to about

40%-70% at 3 km altitude, and to only a few % (if not zero %) at the surface. Unless pollution related ozone in the boundary layer is dynamically lifted upward it will not be readily detected by OMI. It is well known that photochemical production of ozone in the troposphere requires sufficient concentrations of NO<sub>x</sub> and volatile organic compounds, sufficient UV radiation and favorable meteorological conditions. Even drastic changes in pollution and ozone precursors may not significantly change tropospheric ozone. *Witte et al.* [2009] showed from OMI measurements that large reductions in tropospheric NO<sub>2</sub> and SO<sub>2</sub> occurred during the 2008 Olympics event compared to previous Aura years 2004-2007. These sharp reductions were indicative of the rigorous pollution control measures implemented over eastern China prior to and during the Olympics. Despite the large reductions in certain target pollutants we have found no significant reductions in TCO over eastern China for either the TRAJ and ASSIM data products or CTM (figures not shown).

We conclude that the daily ozone products illustrate that they can contribute to case studies involving events such as the Russian fires and the 2008 Olympics. However, much of tropospheric ozone variability occurs over longer timescales including annual cycles and inter-annual periods. In the following we use the important property of long decadal records with the OMI/MLS products to show applications involving inter-annual variability.

## 7.2. The tropical El Niño Southern Oscillation.

The tropical El Niño Southern Oscillation (ENSO) is the dominant source of inter-annual changes of the tropical lower atmosphere and ocean. A description and historical account of

ENSO is given by *Trenberth* [1997; and references therein]. “ENSO” refers to either El Niño (anomalously warm ocean temperatures in the tropical eastern Pacific) or La Niña (anomalously cool ocean temperatures in the tropical eastern Pacific). Consecutive El Niño or La Niña events have a varying periodicity of about 2-7 years and maximize during months around NH late autumn to late winter.

A multi-decadal monthly tropospheric ozone ENSO index (OEI) was derived by *Ziemke et al.* [2010] to monitor intensity of ENSO events in tropospheric ozone and to use as a diagnostic test for models of atmospheric chemistry and transport. To calculate the OEI, TCO for each month is first averaged over the eastern Pacific (15°S-15°N, 110°W-180°) and western Pacific (15°S-15°N, 70°E-140°E). (These two regions were chosen to maximize correlation between TCO and the Niño 3.4 temperature anomaly ENSO index.) A monthly time series is then generated by taking the difference of western minus eastern Pacific tropospheric ozone columns. This time series is then deseasonalized, followed by a 3-month running average (to be consistent with calculation of the monthly Niño 3.4 ENSO index).

Figure 14a plots the TCO OEI derived separately from ASSIM (dotted purple curve), PROF (dashed blue curve), TRAJ (dotted-dashed green curve), and GMI (solid red curve). Also included in Figure 14 is the Niño 3.4 index (solid black curve) and correlation “r” between Niño 3.4 and the four OEI’s. All four OEI’s extend from January 2005 through December 2012 except for PROF OEI which ends December 2008.

Each of the four OEI time series in Figure 14a are nearly identical and within about 1 DU from each other each month. However, all four OEI's differ from Niño 3.4 at certain times of the record. It can be shown from daily values that these discrepancies between the OEI's and Niño 3.4 coincide with intra-seasonal/Madden-Julian Oscillation (MJO) variability [Madden and Julian, 1971, 1994, and references therein] in tropospheric ozone during NH winter-spring months. Large discrepancy occurs around January 2008 (i.e., during La Niña) and shows all four OEI's with relative maxima while Niño 3.4 has a relative minimum. Understanding the tropical intra-seasonal variability/MJO and its coupling effect with ENSO and monthly OEI is beyond the scope of our current study.

We show that the profile ozone from ASSIM may provide some further insight regarding the vertical distribution of the OEI in tropospheric ozone. In Figure 14b ASSIM upper tropospheric column ozone (dotted blue curve) is plotted with lower tropospheric column ozone (dashed red curve) and Niño 3.4 (solid black curve). Upper and lower tropospheric column ozone in Figure 14b are defined as the ozone column between 500 hPa to tropopause and 500 hPa to ground, respectively. The ASSIM ozone in Figure 14b indicates that most contribution to the OEI originates in the upper troposphere above 500 hPa. The month-by-month changes in upper and lower tropospheric ozone generally track each other over the long record, but two exceptions occur. One exception is centered around January 2010 (El Niño) and another around January 2011 (La Niña). The ASSIM tropospheric profile ozone for these two cases suggests that upper and lower tropospheric ozone in the tropical Pacific are not always coherent. Conclusions regarding ASSIM tropospheric ozone profile information are not certain due to lack of a tropospheric chemistry scheme and reduced OMI efficiency in the lower troposphere.

### 7.3. A global analysis of inter-annual variability of ozone.

General characteristics of inter-annual variability in global tropospheric and stratospheric ozone are examined using the eight years of column ozone and profile ozone products. Figure 15 shows monthly zonal mean SCO (top panel) and TCO (bottom panel) from data assimilation. Both SCO and TCO in Figure 15 have recurring seasonal patterns each year but with some years indicating inter-annual change. For SCO there is a 2-3 year QBO cycle in the tropics and extratropics and also anomalous enhanced SCO in the tropics and subtropics around the months October 2010 – January 2011. Equatorial TCO for the final 2-3 years is slightly larger compared to the previous years.

The inter-annual features in SCO and TCO are better identified by removing seasonal cycles in the data. The SCO and TCO from ASSIM in Figure 15 were deseasonalized and re-plotted in Figure 16a to better identify inter-annual differences and near-decadal changes. For comparison with ASSIM, deseasonalized SCO and TCO from TRAJ measurements are shown in Figure 16b. The SCO and TCO products from ASSIM and TRAJ in Figure 16 are equivalent everywhere to within a few Dobson Units. Figure 16 shows that both SCO products have a clear QBO in the tropics and throughout the SH, but in the NH the QBO signal for SCO is mixed with other inter-annual changes occurring in mid-high latitudes (discussed later).

It can be shown for all data products including the GMI CTM that even though inter-annual changes of TCO on a regional basis may constitute many DU, inter-annual changes for zonal mean TCO are exceedingly small. Figure 16 shows that inter-annual variability of zonal mean

TCO is only about 1-2 DU on average for ASSIM and TRAJ. This shows that most all inter-annual change present in zonal mean total column ozone is due to inter-annual change in SCO. Both zonal mean TCO products in Figure 16 also show small increases in the latter years of the record. This is currently being investigated using the GMI model (figures not shown) which also shows small net increase in TCO in the latter years during predominantly La Nina conditions (particularly the strong La Nina event centered around January 2011). *Doherty et al.* [2006] indicated from a coupled chemistry climate model that the shift from El Niño to La Niña conditions in the troposphere may produce several percent increase in the total ozone burden. This is not inconsistent with the small net increases we find in tropical tropospheric ozone in Figure 16.

In Figures 17-19 we show inter-annual variability of zonal mean ozone profile densities (in DU- $\text{km}^{-1}$ ) for selected latitude bands in correspondence to the 8-year record of column ozone in Figure 16. In Figure 17 deseasonalized ozone profiles from ASSIM (top) are plotted with ozone profiles from MLS (bottom) for general one-to-one comparison. Largest contribution to the equatorial QBO shown earlier in Figure 16 lies within the vertical band of about 20-25 km in log-pressure altitude in Figure 17. Figure 18 compares ASSIM ozone profile densities at 40°N and 40°S. Ozone profiles in the SH at 40°S in Figure 18 have a clear 2-3 year QBO signal (which is mostly out-of-phase with the equatorial QBO signal), but not so for the NH because of other additional inter-annual changes. Most stratospheric ozone inter-annual variability in both the NH and SH mid-latitudes lies around 20 km.

ASSIM deseasonalized ozone profiles are plotted again in Figure 19 as in the previous figure but instead for high polar latitudes 80°N and 80°S. Largest inter-annual variability at 80°N in Figure

19 occurs during winter when OMI has no ozone measurements because of polar night. Several enhanced inter-annual changes in ozone in Figure 19 associated with sudden stratospheric warming events occur in the northern polar region. These NH warming events are identified in Figure 19 as large inter-annual changes during winter-spring months. Two intense stratospheric warming events in the NH during winter months are identified in 2006 and 2009. At least three stratospheric warmings in Figure 19 coincide with Arctic split events [e.g., *Ripese et al.*, 2012] in spring 2005, winter 2009, and winter 2010. For each split event the ASSIM ozone has a relative minimum in the vertical centered around 15 km. These relative minima in the lower stratosphere are not present for other warming events such as 2006 and 2011. As a note, winter-spring 2011 in Figure 19 indicates reductions of ozone rather than increases in the lower stratosphere. The anomalous reduction of stratospheric ozone in March 2011 was largely explained in recent studies by *Manney et al.* [2011], *Isaksen et al.* [2012], and *Strahan et al.* [2013] as an unusually strong polar vortex that year which suppressed and delayed the transport of stratospheric ozone from lower latitudes. Inter-annual variability in the SH polar region in Figure 19 (bottom) is small when compared to the NH polar region. For the SH two anomalous events (exceeding +5 DU-km<sup>-1</sup>) occurred in the range 20-25 km during October-November in 2005 and 2012. These two events signify anomalously high stratospheric ozone levels in the immediate lower stratosphere polar region during Antarctic ozone depletion conditions.

## 8. Summary.

Three different strategies to obtain tropospheric and stratospheric ozone from Aura OMI/MLS measurements beginning late 2004 have been evaluated for their overall data quality and



usefulness for science applications. These three strategies are trajectory mapping (denoted TRAJ), direct profile retrieval (denoted PROF), and data assimilation (denoted ASSIM). The evaluation of the three products includes validation using ozonesondes and comparisons with the Global Modeling Initiative (GMI) chemical transport model (CTM).

The three OMI/MLS ozone data products were compared with ozonesonde measurements from WOUDC, SHADOZ, and NDACC. The TRAJ tropospheric column ozone (TCO) product was shown to have the largest bias (i.e., a low bias exceeding 5 DU on average) relative to the sondes. This low bias is mostly due to MLS ozone being high biased in the low stratosphere [e.g. *Livesey et al.*, 2011] and a calibration offset between OMI v8.5 and MLS v3.3 measurements. The ASSIM TCO also has a low bias but it is reduced by better accounting for measurement uncertainties/noise that also reduces RMS differences with sondes. RMS differences are smallest and correlations are closest to one for the ASSIM ozone.

The daily ozone products are demonstrated to be useful for evaluating regional case studies such as the 2010 Russian wildfires and the 2008 Beijing Olympics. In summer 2010 intense wildfires persisted in western Russia caused by anomalously dry conditions. Tropospheric ozone was largest in all of the products and the GMI CTM during July 2010 compared to any other month of the 2005-2012 record. The anomalous increase in tropospheric ozone in western Russia was about 5 DU compared to an average background of around 35 DU. Tropospheric ozone profiles from data assimilation indicate that the anomalous increases in tropospheric ozone in July 2010 extended deep throughout the troposphere with most contribution coming from the lower troposphere below 500 hPa. The ozone increases coincided with anomalous meteorological

conditions that year involving an unusual stationary high pressure blocking event and warm temperatures.

The photochemical production of tropospheric ozone is sensitive to the concentrations of  $\text{NO}_x$  and volatile organic compounds (VOC's), UV radiation, and meteorological conditions. Drastic changes in ozone precursors may not necessarily coincide with measureable changes in tropospheric ozone. A case study example of this effect was the 2008 Beijing Olympics whereby there were large reductions in regional pollution including ozone precursors, yet OMI/MLS data products and the CTM all showed no reduction in tropospheric ozone.

With the Aura OMI and MLS measurements beginning late 2004, the ozone product records are now long enough to study and characterize inter-annual to near-decadal variability of stratospheric and tropospheric ozone. The OMI/MLS ozone products show that while the quasi-biennial oscillation (QBO) is the dominant source of inter-annual variability of stratospheric ozone in the tropics and SH, variability of stratospheric ozone in the NH includes an additional mix of anomalous inter-annual increases and decreases. During the Aura record there were several events of stratospheric sudden warmings in the NH including split events. These sudden warmings occur in polar latitudes around winter months when there are no OMI ozone measurements due to polar night. MLS stratospheric ozone profiles also do not fully cover the polar region and do not “fill in” the ozone profiles missing between orbit paths as assimilation does.

It is well known that the El Niño Southern Oscillation (ENSO) is the dominant source of inter-annual change in tropospheric ozone in the tropical Pacific. The tropospheric ozone ENSO index (OEI) of *Ziemke et al.* [2010] was calculated for the three data products and the CTM for the 8-year Aura record. OEI derived for the three products and CTM are all remarkably consistent to within about 1 DU from each other every month. Ozone profiles from data assimilation suggest further that the OEI is driven mostly by ENSO forced ozone variations in the upper troposphere above 500 hPa.

The three ozone data products and the CTM show that zonal mean tropospheric column ozone has small inter-annual change of about 1-2 DU in any latitude range. The three data products and CTM also show slight increase of 2-4 DU in tropospheric ozone toward the end of the Aura record (i.e., years 2010-2012); the CTM suggests that this may be due in part to La Nina conditions persisting in the tropics toward the end of the Aura record (this is ongoing work beyond our current study).

When based upon ozonesonde validation and the broad set of science applications investigated, the ASSIM ozone is overall the most useful of the three OMI/MLS data products. The assimilated ozone provides a continuous global record of ozone which includes profile information. Profile information for ozone is not provided by the TRAJ product and the PROF profile product has a shorter record (through 2008) and comparatively coarse vertical resolution for ozone profiles. Despite lack of profile information there are benefits of using the TRAJ daily ozone product. The TRAJ product provides a long record of ozone and includes additional geophysical parameters within the product files such as in situ cloud fractions and cloud

pressures, tropopause pressures (various definitions), and tropospheric and stratospheric column amounts (using various tropopause pressure definitions), etc.

The GMAO assimilated ozone in our study is currently a preliminary product that will at later time be made available to the public following further validation by *Wargan et al.* [2013]. Information about using the TRAJ product can be obtained from the NASA Goddard ozone air quality website <http://ozoneaq.gsfc.nasa.gov/>.

## References

Doherty, R. M., D. S. Stevenson, C. E. Johnson, W. J. Collins, and M. G. Sanderson, Tropospheric ozone and El Niño–Southern Oscillation: Influence of atmospheric dynamics, biomass burning emissions, and future climate change, *J. Geophys. Res.*, *111*, D19304, doi:10.1029/2005JD006849, 2006.

Duncan, B.N., S.E. Strahan, Y. Yoshida, S.D. Steenrod, and N. Livesey, Model Study of the Cross-Tropopause Transport of Biomass Burning Pollution, *Atmos. Chem. Phys.*, *7*, 3713-3736, 2007.

Duncan, B. N., J. J. West, Y. Yoshida, et al., The influence of European pollution on ozone in the Near East and northern Africa, *Atmos. Chem. Phys.*, *8*, 22-2283, 2008.

Fishman, J., and J. C. Larsen, Distribution of total ozone and stratospheric ozone in the tropics: Implications for the distribution of tropospheric ozone, *J. Geophys. Res.*, *92*, 6627-6634, doi:10.1029/JD092iD06p06627, 1987.

636

637 Fishman, J., C. E. Watson, J. C. Larsen, and J. A. Logan, Distribution of tropospheric ozone  
 638 determined from satellite data, *J. Geophys. Res.*, 95(D4), 3599-3617,  
 639 doi:10.1029/JD095iD04p03599, 1990.

640

641 Fishman, J., A. E. Wozniak, and J. K. Creilson, Global distribution of tropospheric ozone from  
 642 satellite measurements using the empirically corrected tropospheric ozone residual technique:  
 643 Identification of the regional aspects of air pollution, *Atmos. Chem. Phys.*, 3, 893-907,  
 644 doi:10.5194/acp-3-893-2003, 2003.

645

646 Foltz, G. R., and M. J. McPhaden, The 30-70 day oscillations in the tropical Atlantic, *Geophys.*  
 647 *Res. Lett.*, 31(15), L15205, 2004.

648

649 Foltz, G. R., and M. J. McPhaden, Mixed layer heat balance on intraseasonal time scales in the  
 650 northwestern tropical Atlantic Ocean, *J. Clim.*, 18(20), 4168-4187, 2005.

651

652 Hudson, R. D., and A. M. Thompson, Tropical tropospheric ozone from total ozone mapping  
 653 spectrometer by a modified residual method, *J. Geophys. Res.*, 103, D17, 22,129-22,145,  
 654 doi:10.1029/98JD00729, 1998.

655

656 Isaksen, I. S. A., C. Zerefos, W. C. Wang, D. Balis, K. Eleftheratos, B. Rognerud, F. Stordal, T.  
 657 K. Bernsten, J. H. LaCasce, O. A. Sovde, D. Olivie, Y. J. Orsolini, I. Zyrichidou, M. Prather, and

O. N. E. Tuinder, Attribution of the arctic ozone column deficit in March 2011, *Geophys. Res. Lett.*, *39*, L24810, doi:10.1029/2012GL053876, 2012.

Kiladis, G. N., K. H. Straub, G. C. Reid, and K. S. Gage, Aspects of interannual and intraseasonal variability of the tropopause and lower stratosphere, *Q. J. R. Meteorol. Soc.*, *127*, 576, 1961-1983, doi:10.1256/smsqj.57605, 2001.

Kar, J., J. Fishman, J. K. Creilson, A. Richter, J. R. Ziemke, and S. Chandra, Are there urban signatures in the tropospheric ozone column products derived from satellite measurements?, *Atmos. Chem. Phys.*, *10*, 5213-5222, doi:10.5194/acp-10-5213-2010, 2010.

Kim, J. H., M. J. Newchurch, and K. Han, Distribution of tropical tropospheric ozone determined by the scan-angle method applied to TOMS measurements, *J. Atmos. Sci.*, *58*, 18, 2699-2708, doi:10.1175/1520-0469(2001)058<2699:DOTTOD>2.0.CO;2, 2001.

Liu, X., P. K. Bhartia, K. Chance, R. J. D. Spurr, and T. P. Kurosu, Ozone profile retrievals from the Ozone Monitoring Instrument, *Atmos. Chem. Phys.*, *10*, 2521-2537, 2010a.

Liu, X., P. K. Bhartia, K. Chance, L. Froidevaux, R. J. D. Spurr, and T. P. Kurosu, Validation of Ozone Monitoring Instrument (OMI) ozone profiles and stratospheric ozone columns with Microwave Limb Sounder (MLS) measurements, *Atmos. Chem. Phys.*, *10*, 2539-2549, 2010b.

680 Livesey, N. J., W. G. Read, L. Froidevaux, A. Lambert, G. L. Manney, H. C. Pumphrey, M. L.  
 681 Santee, M. J. Schwartz, S. Wang, R. E. Cofield, D. T. Cuddy, R. A. Fuller, R. F. Jarnot, J. H.  
 682 Jiang, B. W. Knosp, P. C. Stek, P. A. Wagner, and D. L. Wu, EOS MLS Version 3.3 Level 2  
 683 data quality and description document, Tech. rep., Jet Propulsion Laboratory, available from  
 684 <http://mls.jpl.nasa.gov/>, 2011.  
 685  
 686 Madden, R. A., and P. R. Julian, Description of the 40-50 day oscillation in the zonal wind in the  
 687 tropical Pacific, *J. Atmos., Sci.*, 28, 702-708, 1971.  
 688  
 689 Madden, R. A., and P. R. Julian, Observations of the 40-50 day tropical oscillation – a review,  
 690 *Mon. Wea. Rev.*, 122, 814-837, 1994.  
 691  
 692 Manney, G. L., Unprecedented Arctic ozone loss in 2011, *Nature*, 478, 469-475,  
 693 doi:10.1038/nature10556, 2011.  
 694  
 695 McPeters, R. D., G. J. Labow, and J. A. Logan, Ozone climatological profiles for satellite  
 696 retrieval algorithms, *J. Geophys. Res.*, 112, D5, D05308, doi:10.1029/2005JD006823, 2007.  
 697  
 698 Molod, A., L. Takacs, M. Suarez, J. Bacmeister, I.-S. Song, and A. Eichmann, The GEOS-5  
 699 Atmospheric General Circulation Model: Mean Climate and Development from MERRA to  
 700 Fortuna. NASA Technical Report Series on Global Modeling and Data Assimilation, NASA  
 701 TM-2012-104606, Vol. 28, 117 pp. [http://gmao.gsfc.nasa.gov/pubs/tm/archive/tm\\_2012.php](http://gmao.gsfc.nasa.gov/pubs/tm/archive/tm_2012.php),  
 702 2012.

703

704 Newchurch, M. J., X. Liu, J. H. Kim, Lower-tropospheric ozone (LTO) derived from TOMS  
 705 near mountainous regions, *J. Geophys. Res.*, *106*, D17, 20,403-20,412,  
 706 doi:10.1029/2000JD000162, 2001.

707

708 Park, C.-K., and S. D. Schubert, Remotely forced intraseasonal oscillations over the tropical  
 709 Atlantic, *J. Atmos. Sci.*, *50*, 89-103, 1993.

710

711 Purser, R. J., W.-S. Wu, D. F. Parrish, and N. M. Roberts, Numerical aspects of the application  
 712 of recursive filters to variational statistical analysis. Part I: spatially homogeneous and isotropic  
 713 Gaussian covariances, *Mon. Wea. Rev.*, *131*, 1524-1535, 2003a.

714

715 Purser, R. J., W.-S. Wu, D. F. Parrish, and N. M. Roberts, Numerical aspects of the application  
 716 of recursive filters to variational statistical analysis. Part II: spatially inhomogeneous and  
 717 anisotropic general covariances, *Mon. Wea. Rev.*, *131*, pp. 1536-1548, 2003b.

718

719 Randel, W. J., and F. Wu, A stratospheric ozone profile data set for 1979-2005: Variability,  
 720 trends, and comparisons with column ozone data, *J. Geophys. Res.*, *112*, D06313,  
 721 doi:10.1029/2006JD007339, 2007.

722

723 Rienecker, M.M., M.J. Suarez, R. Gelaro, R. Todling, J. Bacmeister, E. Liu, M.G. Bosilovich,  
 724 S.D. Schubert, L. Takacs, G.-K. Kim, S.E. Bloom, J. Chen, D. Collins, A. Conaty, A. da Silva,  
 725 W. Gu, J. Joiner, R.D. Koster, R. Lucchesi, A. Molod, T. Owens, S. Pawson, P. Pegion, C.R.



726 Redder, R. Reichle, F.R. Robertson, A.G. Ruddick, M. Sienkiewicz, J. Woollen, MERRA -  
 727 NASA's Modern-Era Retrospective Analysis for Research and Applications, *J. Climate*, 24,  
 728 doi:10.1175/JCLI-D-11-00015.1, 2011.  
 729  
 730 Ripesi, P., F. Ciciulla, F. Maimone, and V. Pelino, The February 2010 Arctic Oscillation Index  
 731 and its stratospheric connection, *Q. J. R. Meteorol. Soc.*, 138, 1961-1969, doi:10.1002/qj.1935,  
 732 2012.  
 733  
 734 Rui, H., and B. Wang, Development characteristics and dynamic structure of tropical  
 735 intraseasonal convection anomalies. *J. Atmos. Sci.*, 47, 357,379, 1990.  
 736  
 737 Sassi, F., M. Salby, H. C. Pumphrey, and W. G. Read, Influence of the Madden-Julian  
 738 Oscillation on upper tropospheric humidity, *J. Geophys. Res.*, 107, D23, 4681,  
 739 doi:10.1029/2001JD001331, 2002.  
 740  
 741 Sauvage, B., R. V. Martin, A. van Donkelaar, and J. R. Ziemke, Quantification of the factors  
 742 controlling tropical tropospheric ozone and the South Atlantic maximum, *J. Geophys. Res.*,  
 743 112(D11) D11309, doi:1029/2006JD008008, 2007.  
 744  
 745 Schoeberl, M. R., J. R. Ziemke, B. Bojkov, N. Livesey, B. Duncan, S. Strahan, L. Froidevaux, S.  
 746 Kulawik, P. K. Bhartia, S. Chandra, P. F. Levelt, J. C. Witte, A. M. Thompson, E. Cuevas, A.  
 747 Redondas, D. W. Tarasick, J. Davies, G. Bodeker, G. Hansen, B. J. Johnson, S. J. Oltmans, H.  
 748 Vomel, M. Allaart, H. Kelder, M. Newchurch, S. Godin-Beekmann, G. Ancellet, H. Claude, S.

B. Andersen, E. Kyro, M. Parrondos, M. Yela, G. Zablocki, D. Moore, H. Dier, P. von der  
 Gathen, P. Viatte, R. Stubi, B. Calpini, P. Skrivankova, V. Dorokhov, H. de Backer, F. J.  
 Schmidlin, G. Coetzee, M. Fujiwara, V. Thouret, F. Posny, G. Morris, J. Merrill, C. P. Leong, G.  
 Koenig-Langlo, and E. Joseph, A trajectory-based estimate of the tropospheric ozone column  
 using the residual method, *J. Geophys. Res.*, *112*, D24S49, doi:10.1029/2007JD008773, 2007.

Stajner, I., K. Wargan, S. Pawson, H. Hayashi, L-P. Chang, R.C. Hudman, L. Froidevaux, N.  
 Livesey, P.F. Levelt, A.M. Thompson, D.W. Tarasick, R. Stubi, S.B. Andersen, M. Yela, G.  
 Konig-Langlo, F.J. Schmidlin, and J. C. Witte, Assimilated ozone from EOS-Aura: evaluation of  
 the tropopause region and tropospheric columns, *J. Geophys. Res.* *113*, D16S32,  
 doi:10.1029/2007JD008863, 2008.

Strahan, S. E., B. N. Duncan, and P. Hoor, Observationally-derived diagnostics of transport in  
 the lowermost stratosphere and their application to the GMI chemistry transport model, *Atmos.*  
*Chem. Phys.*, *7*, 2435-2445, 2007.

Strahan, S. E., A. R. Douglass, and P. A. Newman, the contributions of chemistry and transport  
 to low arctic ozone in March 2011 derived from Aura MLS observations, *J. Geophys. Res.*, *118*,  
 1563-1576, doi:10.1002/jgrd.50181, 2013.

Thompson, A. M., J. C. Witte, S. J. Oltmans, F. J. Schmidlin, J. A. Logan, M. Fujiwara, W. Volker, W. J.  
 H. Kirchhoff, F. Posny, G. J. R. Coetzee, B. Hoegger, S. Kawakami, T. Ogawa, J. P. F. Fortuin, and H.  
 M. Kelder, Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998-2000 tropical ozone

771 climatology 2. Tropospheric variability and the zonal wave-one, *J. Geophys. Res.*, *108*, D2, 8241,  
772 doi:10.1029/2002JD002241, 2003.

773

774 van der Werf, G. R., J. T. Randerson, L. Giglio, G. J. Collatz, P. S. Kasibhatla, and A. F.  
775 Arellano, Interannual variability in global biomass burning emissions from 1997 to 2004, *Atmos.*  
776 *Chem. Phys.*, *6*, 3423-3441, 2006.

777

778 Wargan, K., S. Pawson, M. Olsen, J. Witte, A. Douglass, J. Ziemke, S. Strahan, The Global  
779 Structure of Ozone in the UTLS Region in an Assimilation of EOS-Aura Data, in preparation,  
780 2013.

781

782 Witte, J. C., M. R. Schoeberl, A. R. Douglass, J. F. Gleason, N. A. Krotkov, J. C. Gille, K. E.  
783 Pickering, and N. Livesey, Satellite observations of changes in air quality during the 2008  
784 Beijing Olympics and Paralympics, *Geophys. Res. Lett.*, *36*, L17803,  
785 doi:10.1029/2009GL039236, 2009.

786

787 Witte, J. C., A. R. Douglass, A. da Silva, O. Torrs, R. Levy, and B. N. Duncan, NASA A-Train  
788 and Terra observations of the 2010 Russian wildfires, *Atmos. Chem. Phys.*, *11*, 9287-9301,  
789 doi:10.5194/acp-11-9287-2011, 2011.

790

791 Wu, W.-S., R. J. Purser, and D. F. Parrish, Three-dimensional variational analysis with spatially  
792 inhomogeneous covariances, *Mon. Wea. Rev.*, *130*, 2905-2916, 2002.

793

Ziemke, J. R., S. Chandra, and P. K. Bhartia, Two new methods for deriving tropospheric column ozone from TOMS measurements: The assimilated UARS MLS/HALOE and convective-cloud differential techniques, *J. Geophys. Res.*, *103*, 22,115-22,127, 1998.

Ziemke, J. R., S. Chandra, B. N. Duncan, L. Froidevaux, P. K. Bhartia, P. F. Levelt, and J. W. Waters, Tropospheric ozone determined from Aura OMI and MLS: Evaluation of measurements and comparison with the Global Modeling Initiative's Chemical Transport Model, *J. Geophys. Res.*, *111*, D19303, doi:10.1029/2006JD007089, 2006.

Ziemke, J. R., S. Chandra, B. N. Duncan, M. R. Schoeberl, M. R. Damon, O. Torres, and P. K. Bhartia, Recent biomass burning events in the tropics and elevated concentrations of tropospheric ozone, *Geophys. Res. Lett.*, *36*, L15819, doi:10.1029/2009GL039303, 2009.

Ziemke, J. R., S. Chandra, L. D. Oman, and P. K. Bhartia, A new ENSO index derived from satellite measurements of column ozone, *Atmos. Chem. Phys.*, *10*, 3711-3721, 2010.

**Table 1.** Spatial correlation between product TCO and mean field TCO within four latitude bands beginning with northern-most at the top. Calculations are done for three-month seasons of December-February, March-May, June-August, and September-November (DJF, MAM, JJA, SON, respectively). Each latitude band is comprised of 432 latitude-longitude grid points at  $5^{\circ} \times 5^{\circ}$  resolution.

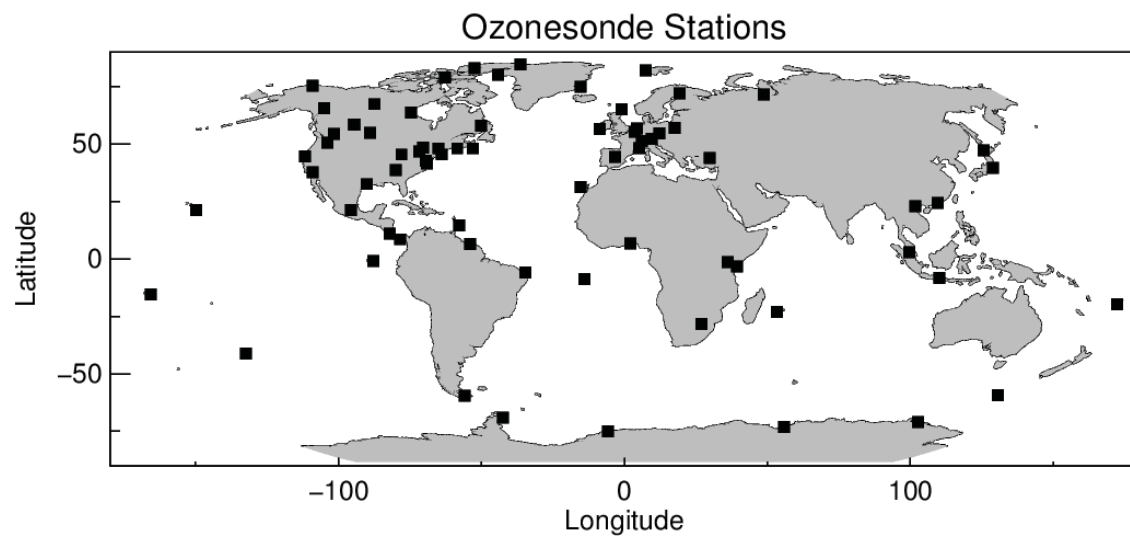
Product	Latitudes	DJF	MAM	JJA	SON
ASSIM	30N-60N	0.92	0.93	0.91	0.95
TRAJ	30N-60N	0.77	0.94	0.97	0.93
PROF	30N-60N	0.81	0.95	0.96	0.96
ASSIM	0-30N	0.94	0.97	0.97	0.93
TRAJ	0-30N	0.96	0.99	0.98	0.97
PROF	0-30N	0.96	0.99	0.97	0.97
ASSIM	0-30S	0.94	0.93	0.97	0.97
TRAJ	0-30S	0.98	0.98	0.93	0.97
PROF	0-30S	0.97	0.94	0.93	0.95
ASSIM	30S-60S	0.99	0.98	0.93	0.99
TRAJ	30S-60S	0.99	0.92	0.65	0.93
PROF	30S-60S	0.99	0.98	0.92	0.98

**Table 2.** Similar to Table 1 but instead for product TCO minus mean field TCO difference in Dobson Units.

Product	Latitudes	DJF	MAM	JJA	SON
ASSIM	30N-60N	-3.10	-2.74	-2.85	-5.15
TRAJ	30N-60N	-8.53	-7.32	-6.68	-8.33
PROF	30N-60N	1.73	3.19	1.63	1.72
ASSIM	0-30N	-2.48	-1.48	-0.37	-1.66
TRAJ	0-30N	-7.93	-7.04	-6.52	-6.66
PROF	0-30N	-1.96	-1.92	-1.63	-1.83
ASSIM	0-30S	-0.58	-0.88	-2.15	-1.92
TRAJ	0-30S	-6.88	-7.13	-8.22	-6.92
PROF	0-30S	-1.58	-2.15	-1.27	0.22
ASSIM	30S-60S	-0.17	-1.25	0.00	-0.81
TRAJ	30S-60S	-6.63	-7.15	-7.03	-6.77
PROF	30S-60S	-0.46	0.19	0.47	1.83

**Table 3.** Similar to Table 1 but instead for the difference RMS of Product TCO minus mean field TCO in Dobson Units. All TCO averages were removed prior to calculation of these difference RMS numbers.

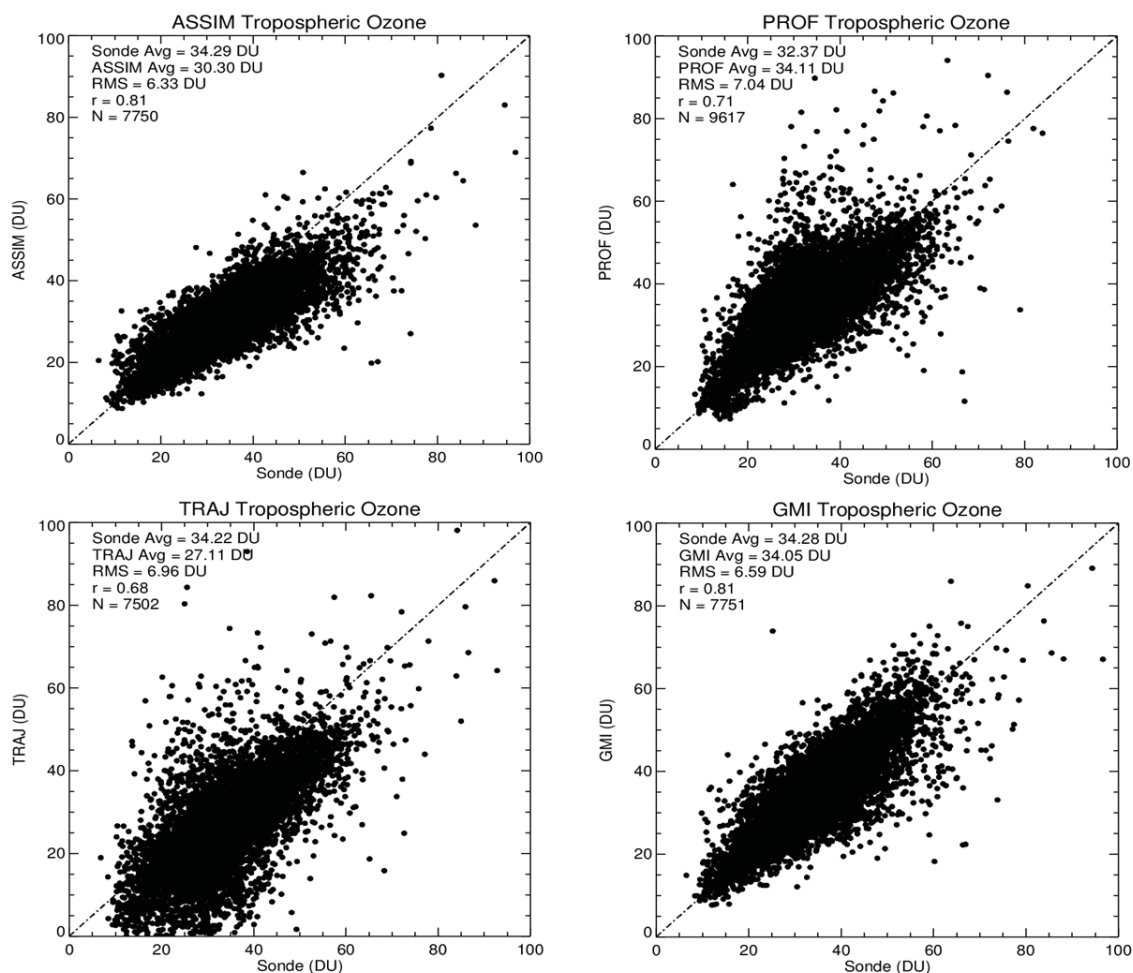
Product	Latitudes	DJF	MAM	JJA	SON
ASSIM	30N-60N	1.23	1.52	2.00	1.26
TRAJ	30N-60N	2.22	1.52	1.13	1.35
PROF	30N-60N	3.04	1.46	1.38	1.08
ASSIM	0-30N	1.82	2.19	1.70	2.07
TRAJ	0-30N	1.32	1.10	1.16	1.24
PROF	0-30N	1.54	1.77	1.56	1.33
ASSIM	0-30S	1.73	1.53	1.18	1.74
TRAJ	0-30S	0.97	0.95	1.84	1.74
PROF	0-30S	1.34	1.58	2.48	3.01
ASSIM	30S-60S	0.77	0.62	0.93	0.79
TRAJ	30S-60S	0.86	1.18	2.11	1.75
PROF	30S-60S	0.98	0.64	1.72	2.05



**Figure 1.** Geo-location sites for ozonesondes used in our study for validation of the ozone products. The ozonesonde data are daily ozone profile measurements from SHADOZ, WOUDC, and NDACC.

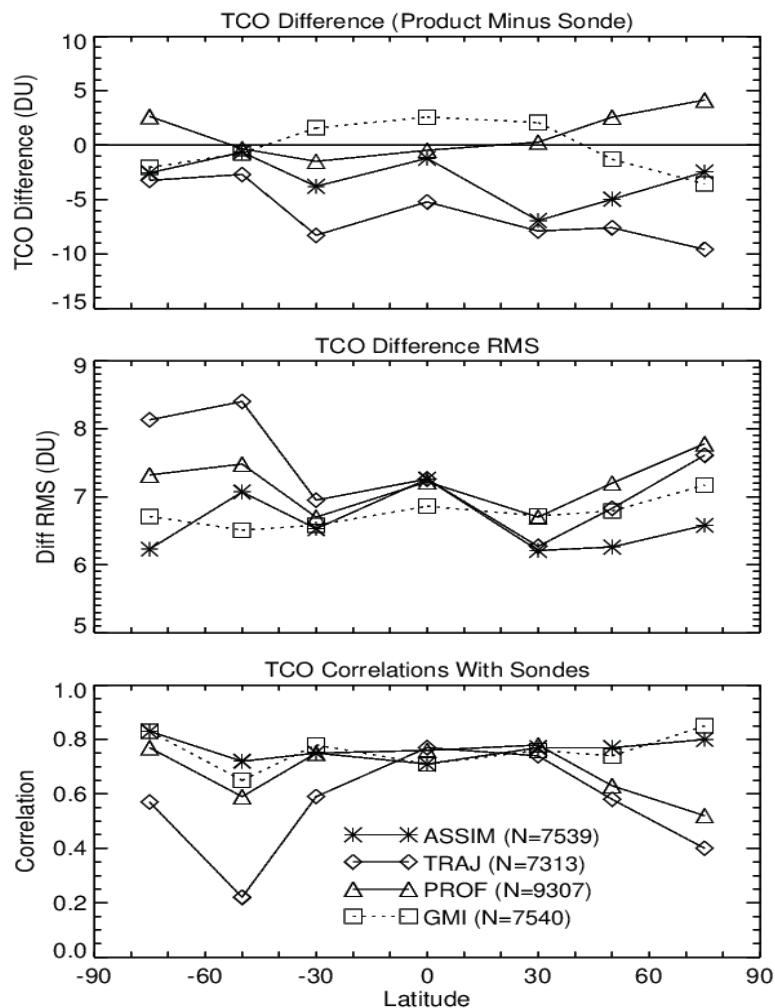


## Ozonesonde TOR Scatter Plots for Global Gridded Products: All Ground Stations Between 90°S and 90°N



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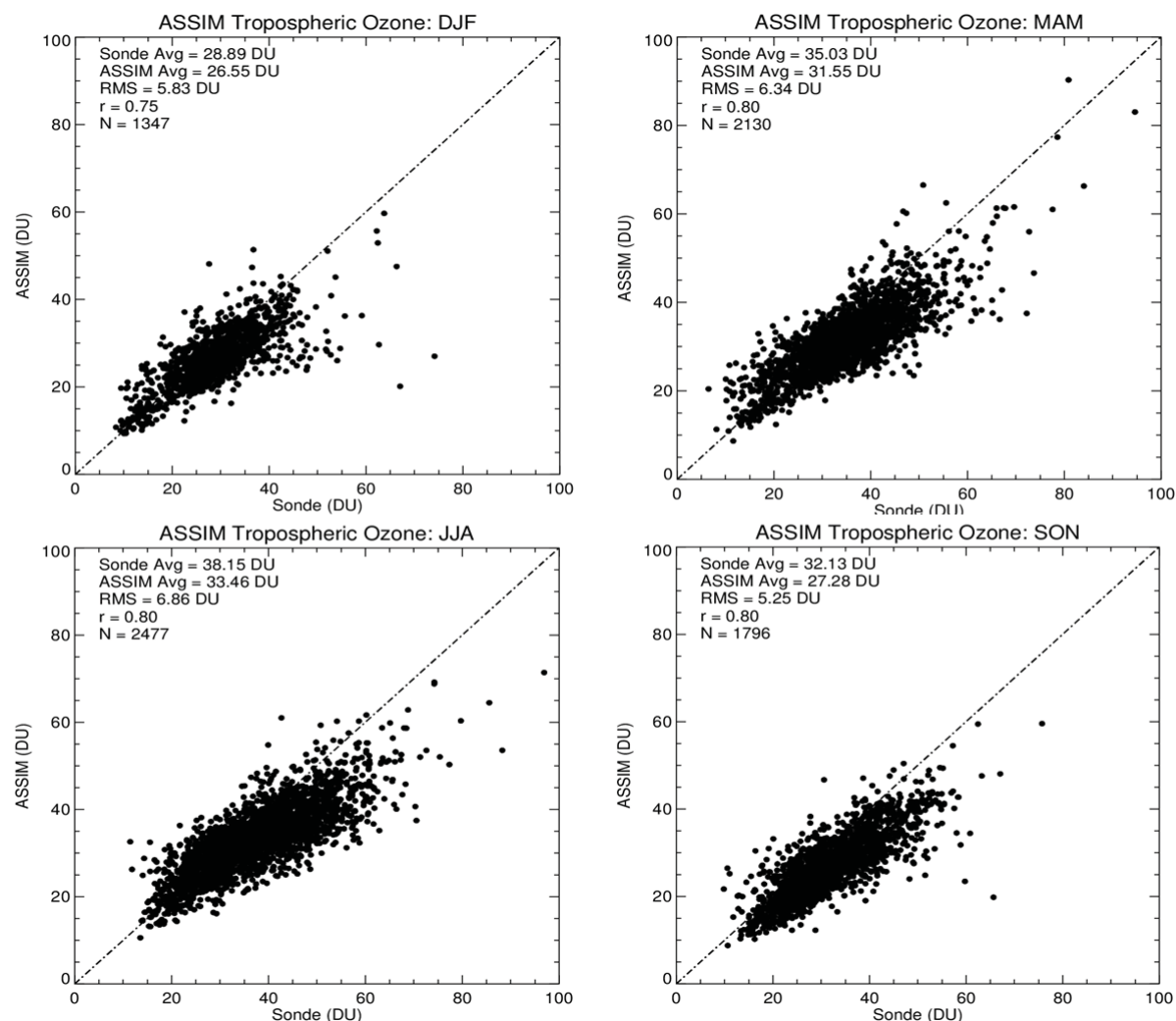
891 **Figure 2.** Daily TCO for the four products (data assimilation, direct profile retrieval, trajectory  
892 mapping, and GMI model) plotted versus coincident ozonesonde TCO. Included for each  
893 product in the scatter plots are their averages, difference RMS value, correlation, and total  
894 number of data pairs. The 1-1 line is also shown. The years included for ozonesonde  
895 comparisons for PROF are 2004-2008 and 2005-2010 for GMI, TRAJ, and ASSIM.



**Figure 3.** Expansion of the product/ozonesonde TCO comparisons of Figure 2 as function of latitude band. These latitude bands vary from 20° to 40° intervals as follows: 60°S-90°S, 40°S-60°S, 20°S-40°S, 20°S-20°N, 20°N-40°N, 40°N-60°N, and 60°N-90°N. The top, middle, and bottom panels show data product (or model) minus sonde difference, product/model minus ozonesonde difference RMS, and correlation between product/model and ozonesonde, respectively. Also listed in the bottom panel is “N” which represents the total number of ozonesonde profiles included in these calculations.

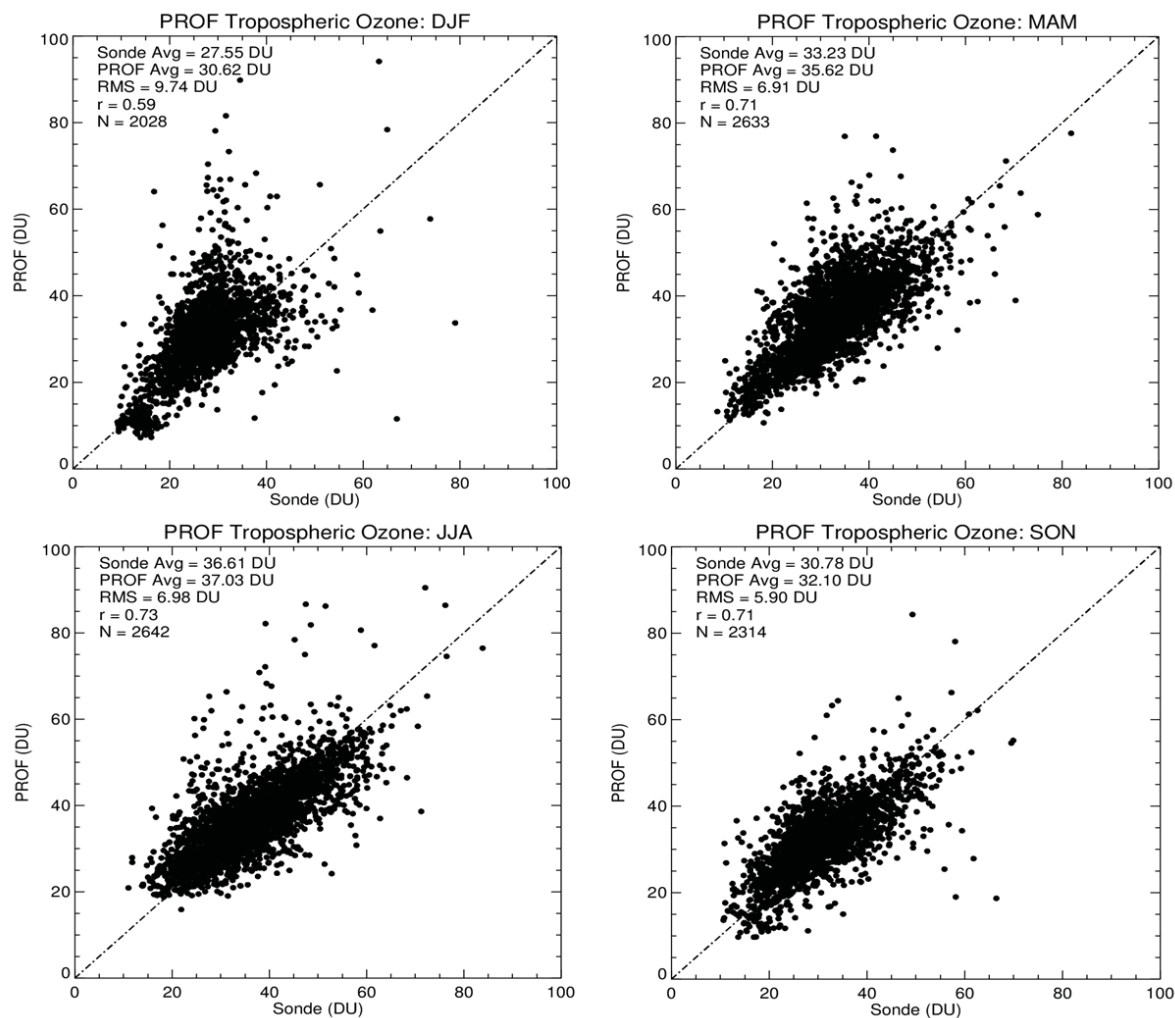
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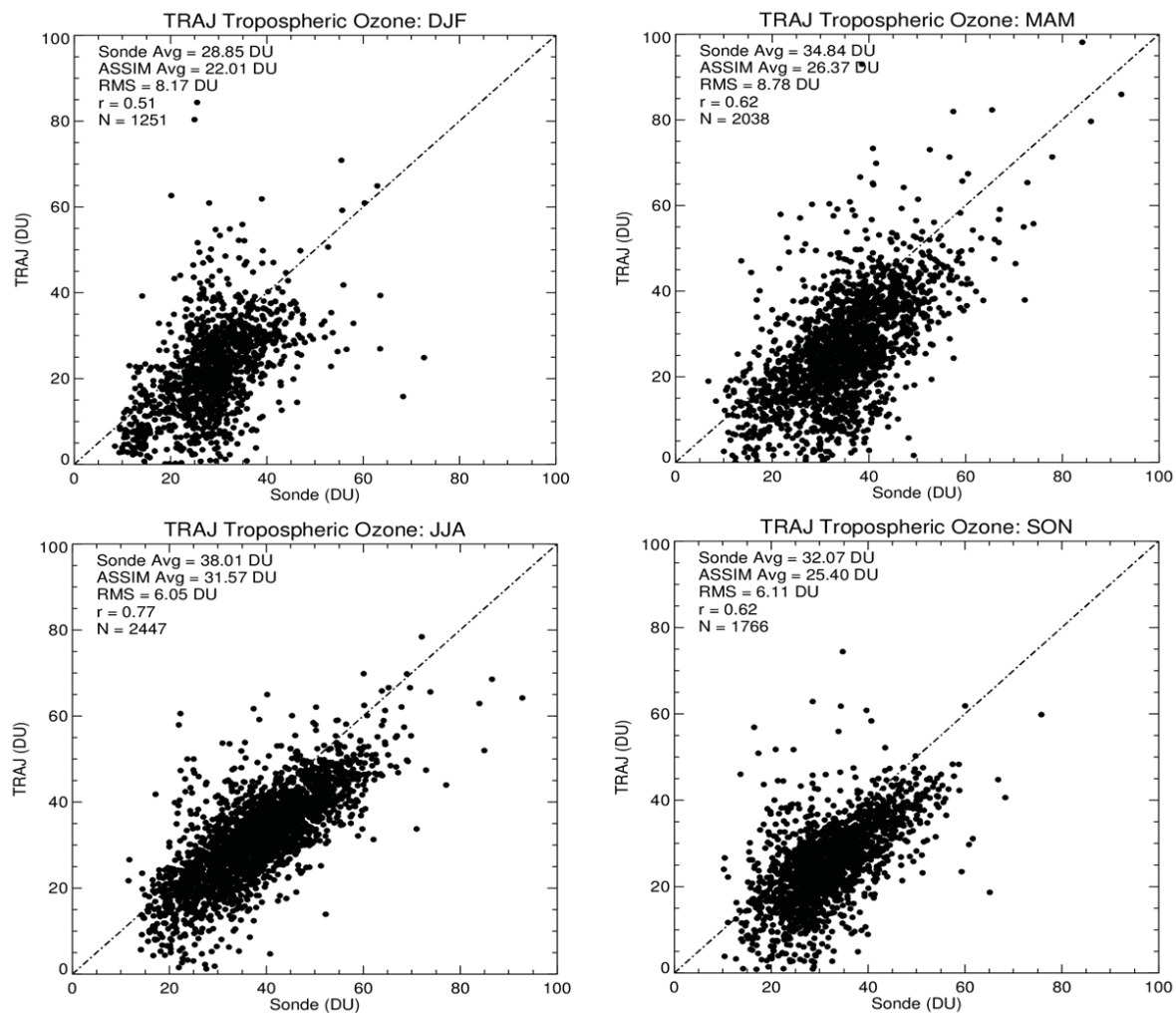


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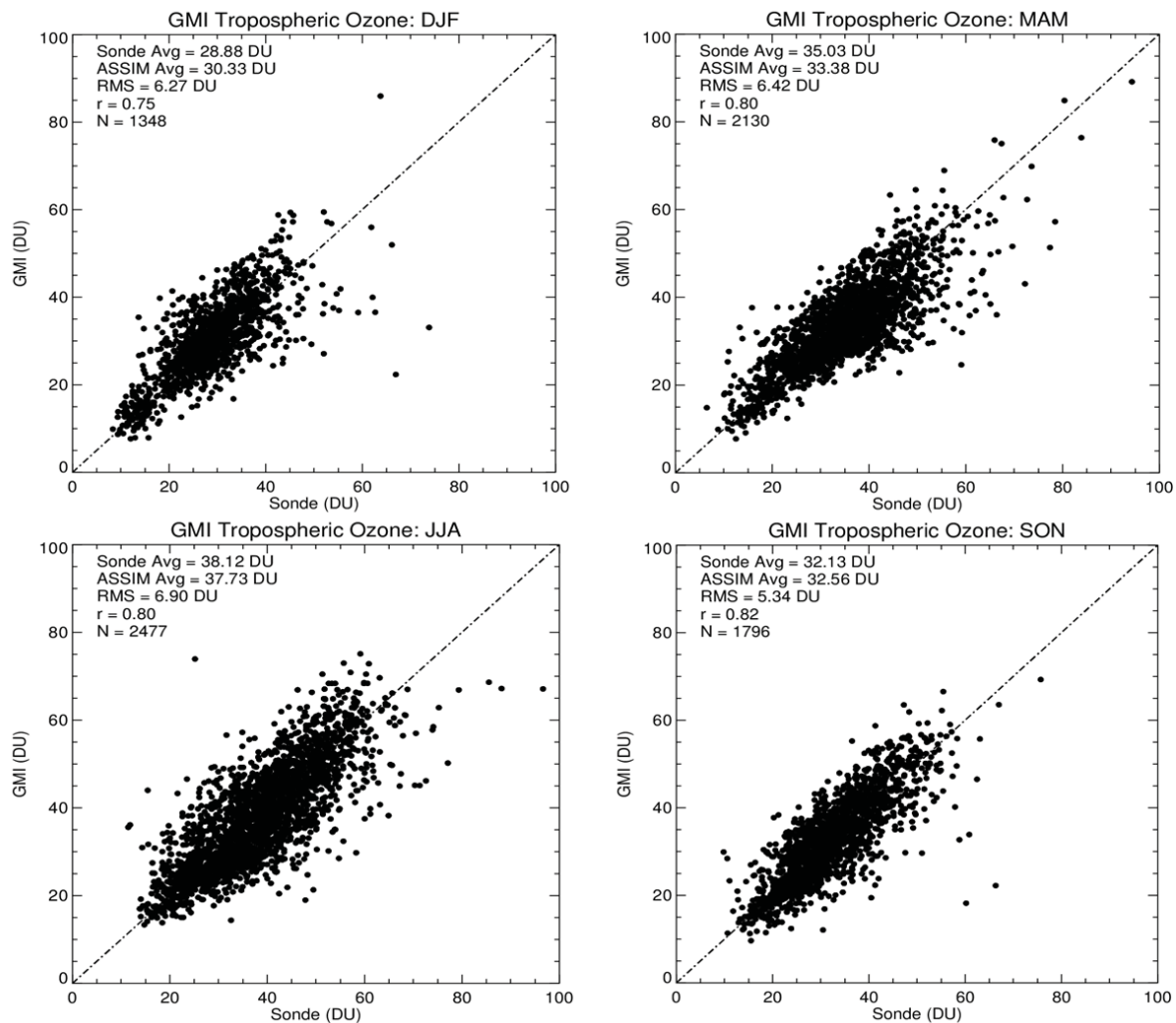
908 **Figure 4.** Daily TCO for GMAO data assimilation plotted versus coincident ozonesonde TCO  
 909 for the four 3-month seasons (indicated). Ozonesondes represent all available global station  
 910 locations. Included for each product in the scatter plots are their averages, difference RMS  
 911 value, correlation, and total number of data pairs. The 1-1 line is also shown. The years  
 912 included for ozonesonde comparisons for PROF are 2004-2008 and 2005-2010 for GMI, TRAJ,  
 913 and ASSIM.



**Figure 5.** Same as Figure 4 but for direct profile retrieved TCO.

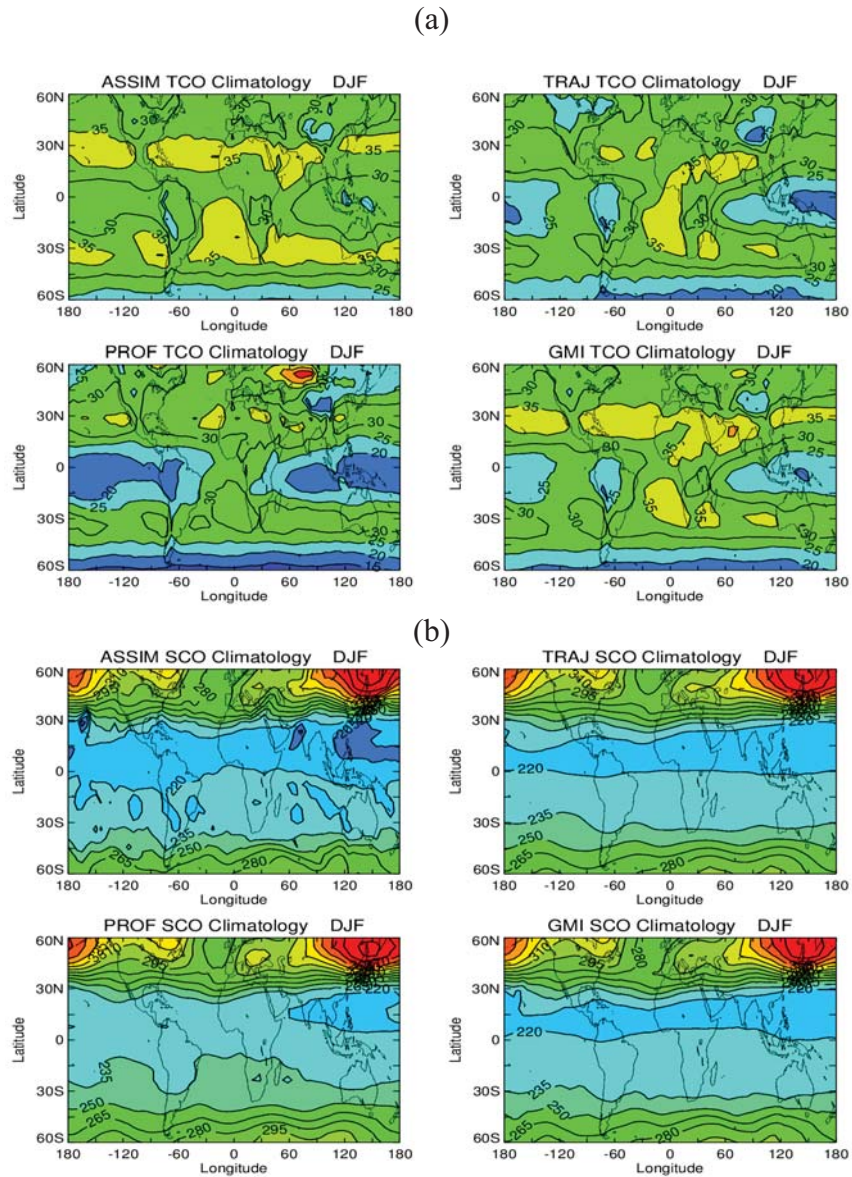


**Figure 6.** Same as Figure 4 but for trajectory mapped TCO.



**Figure 7.** Same as Figure 4 but for GMI model TCO.





**Figure 8.** (a) Seasonal climatology of TCO (in Dobson Units) for the December-February season for each of the three products and GMI model (indicated). (b) Similar to (a) but for SCO. For this visualization comparison of spatial variability a constant offset of +4 DU, +7 DU, -2 DU, and 0 DU (based on ozonesonde differences in Figure 2) was applied to all TCO measurements for ASSIM, TRAJ, PROF, and GMI, respectively. These constant offsets were applied only for plotting TCO here and were not applied to the products in calculating the spatial statistics listed in Tables 1-3.

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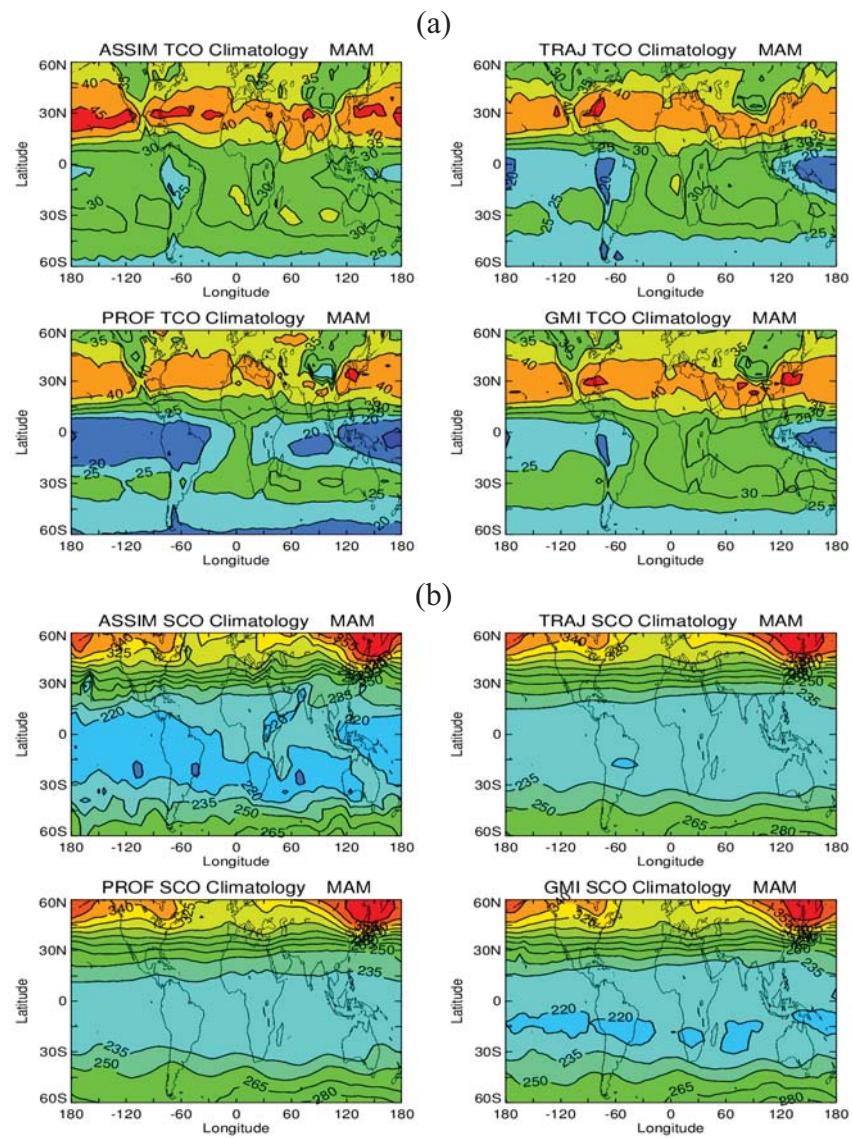
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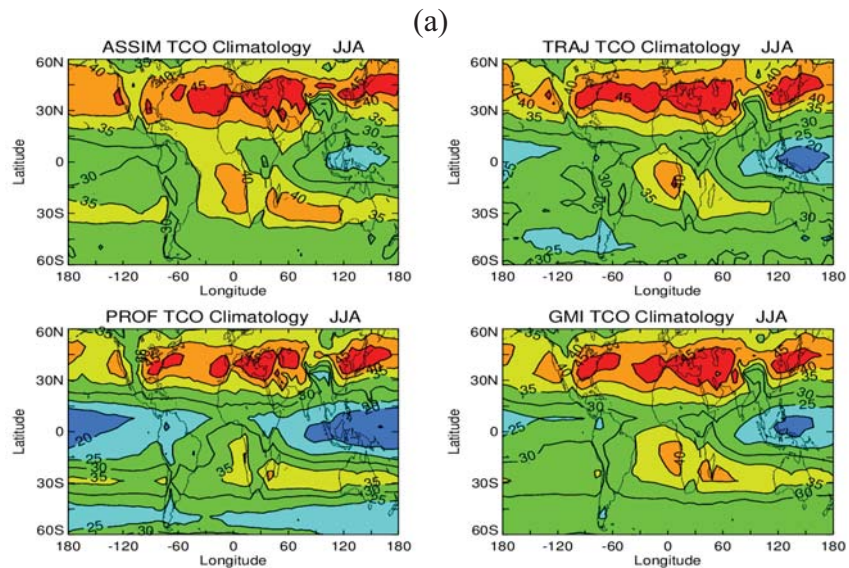
**Figure 9.** Similar to Figure 8 but instead for March-May season.

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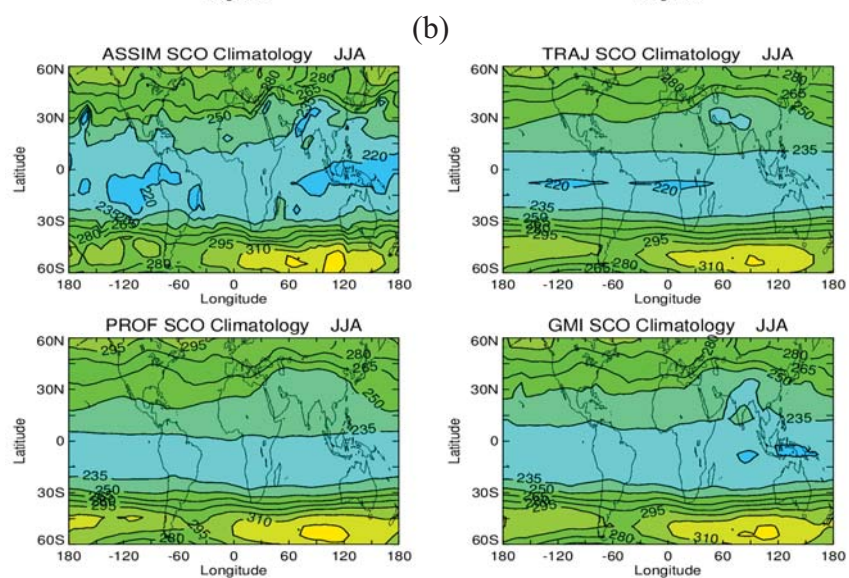
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**Figure 10.** Similar to Figure 8 but instead for June-August season.

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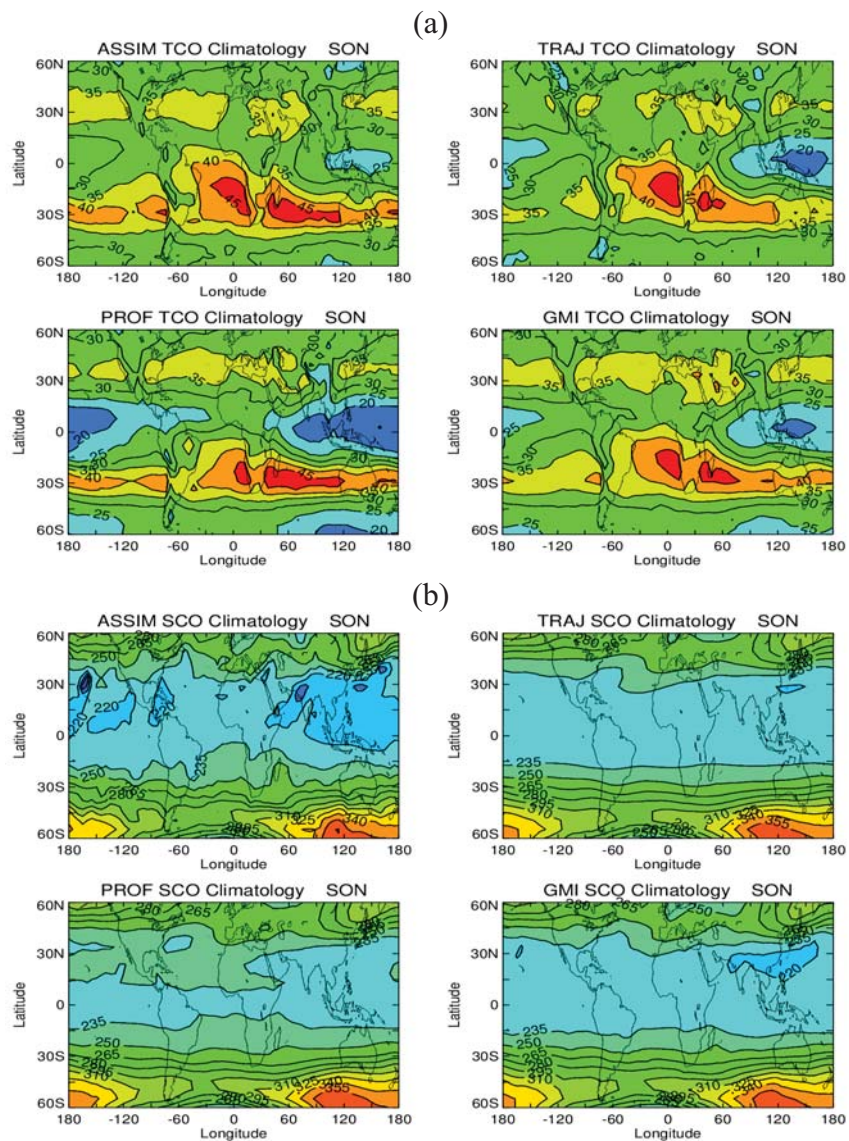
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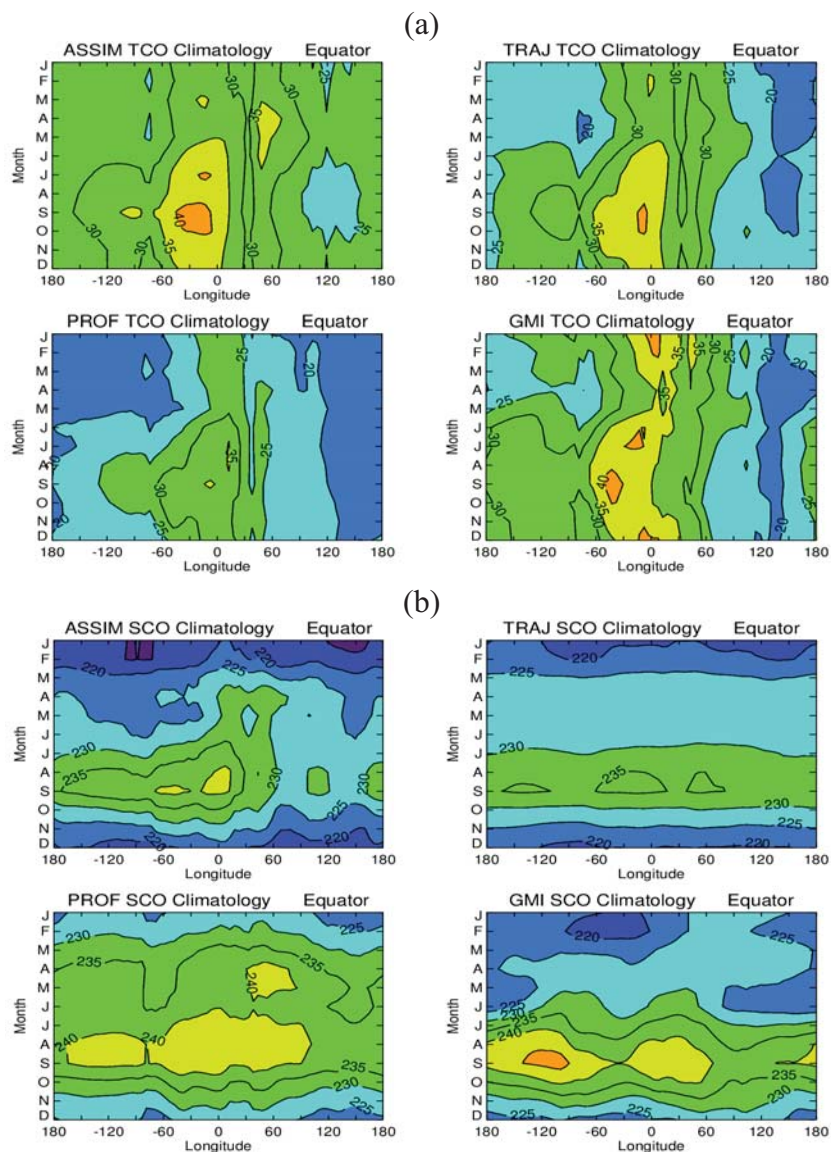
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**Figure 11.** Similar to Figure 8 but instead for September-November season.

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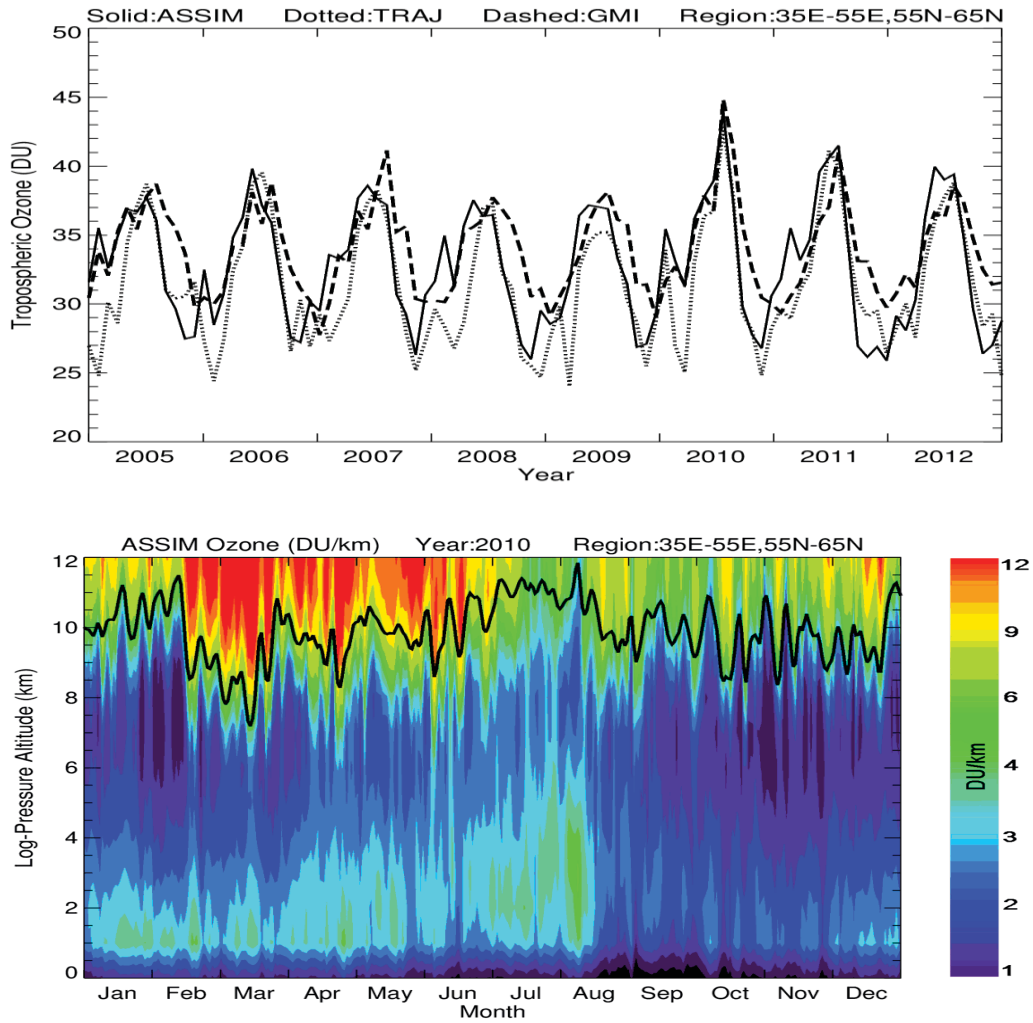


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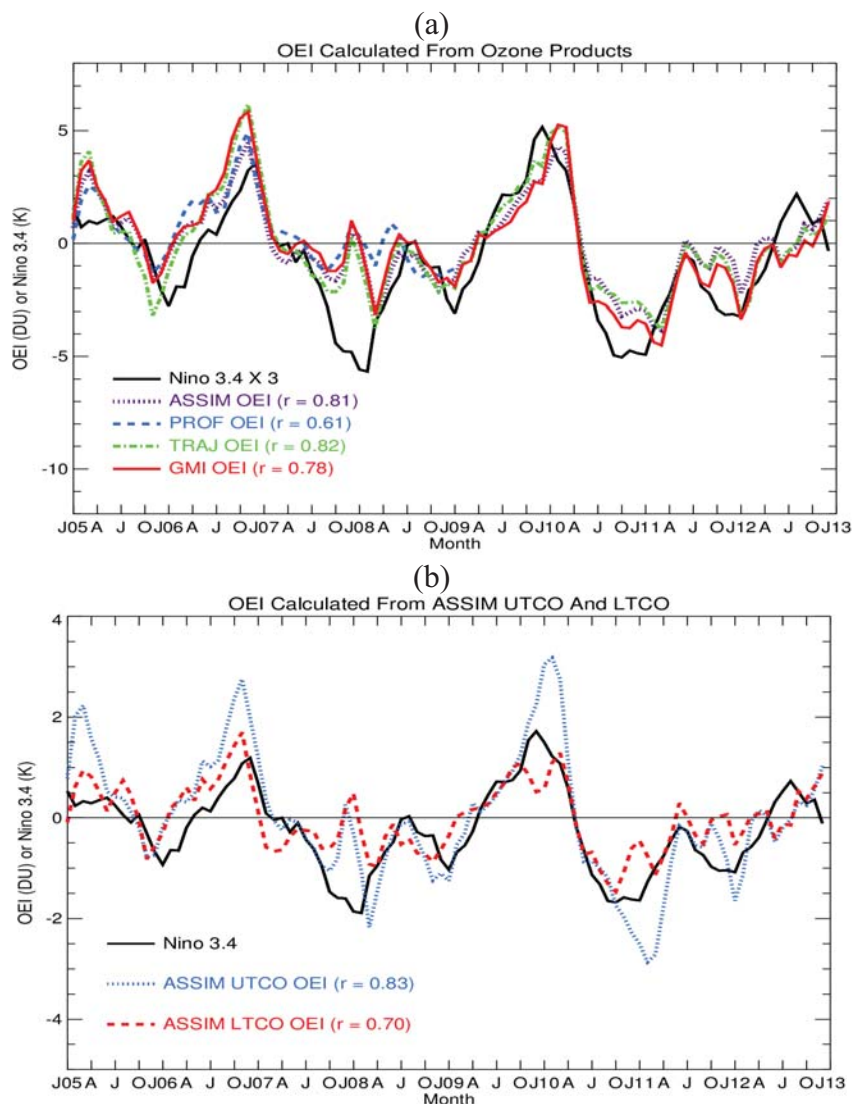
**Figure 12.** (a) Hovmoller plots of month versus longitude monthly TCO climatologies of the three data products and GMI model along the equator. (b) Same as (a) but instead for SCO. All column amounts are in Dobson Units.



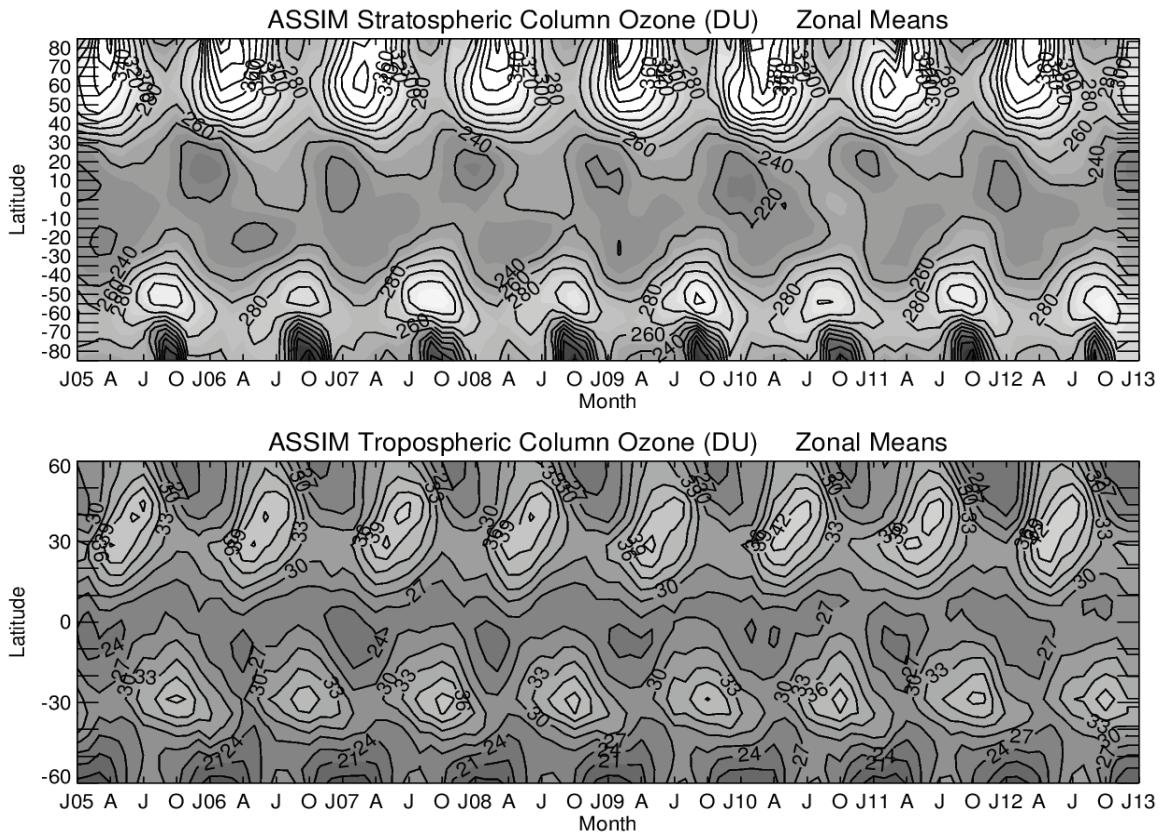


**Figure 13.** (top) Monthly mean TCO time series measurements averaged over western Russia from ASSIM (solid), TRAJ (dotted), and GMI model (long dashed). TCO for the three products was averaged over the region  $35^{\circ}\text{E}$ - $55^{\circ}\text{E}$ ,  $55^{\circ}\text{N}$ - $65^{\circ}\text{N}$ . Anomalous increases in TCO over the region occurred in July 2010 for all three TCO time series in conjunction with record dry conditions involving high tropospheric temperatures and uncontrolled wildfires. (bottom) Contour plot of daily ozone profiles (calculated vertical gradients in  $\text{DU}\cdot\text{km}^{-1}$ ) from ASSIM for year 2010 over the same broad region. The thick black curve is NCEP tropopause pressure in log-pressure altitude.

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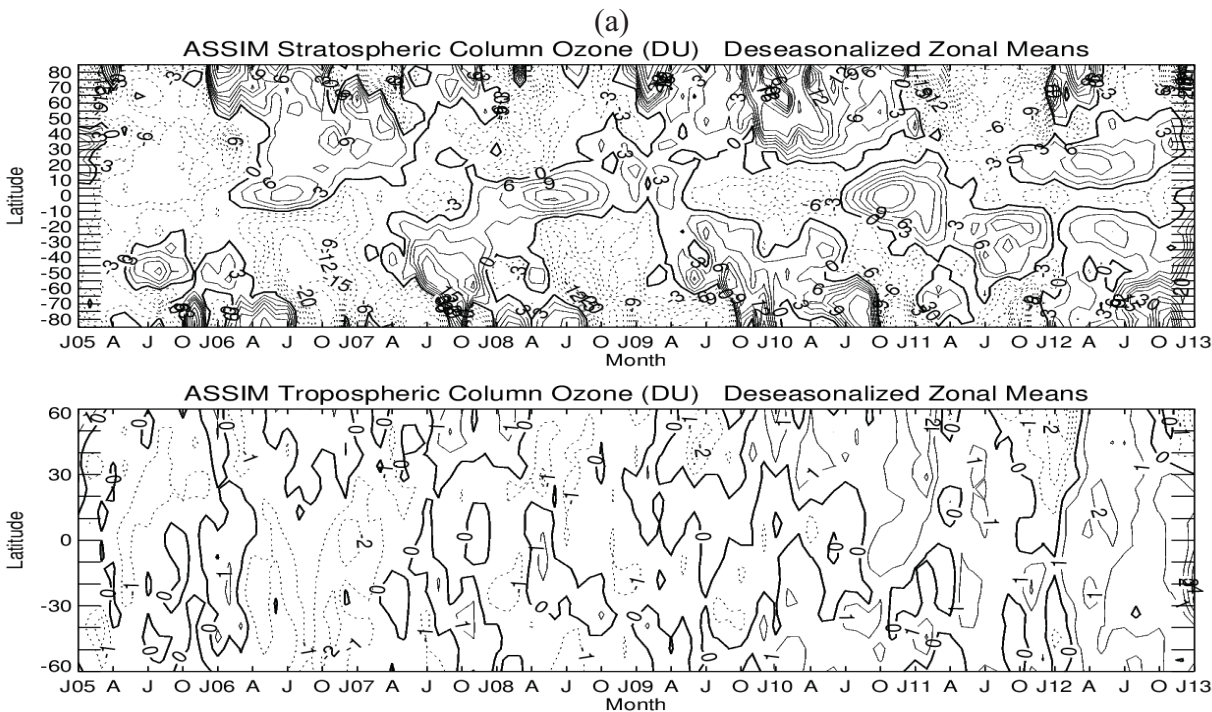


**Figure 14.** (a) Monthly tropospheric Ozone ENSO Index (OEI) derived separately from ASSIM (dotted purple curve), PROF (dashed blue curve), TRAJ (dotted-dashed green curve), and GMI (solid red curve). Also shown is the Niño 3.4 monthly temperature anomaly ENSO index (solid black curve) along with listed time series correlations “ $r$ ” between the Niño 3.4 index and OEI’s from the three products and CTM. OEI time series are given in Dobson Units while the Niño 3.4 units are in K. All four OEI’s extend from January 2005 through December 2012 except for PROF OEI which ends December 2008. (b) ASSIM upper tropospheric column ozone (UTCO, dotted blue curve) is plotted with lower tropospheric column ozone (LTCO, dashed red curve) and Niño 3.4 (solid black curve). Upper and lower tropospheric column ozone are defined as the ozone column from 500 hPa to tropopause and 500 hPa to ground, respectively.

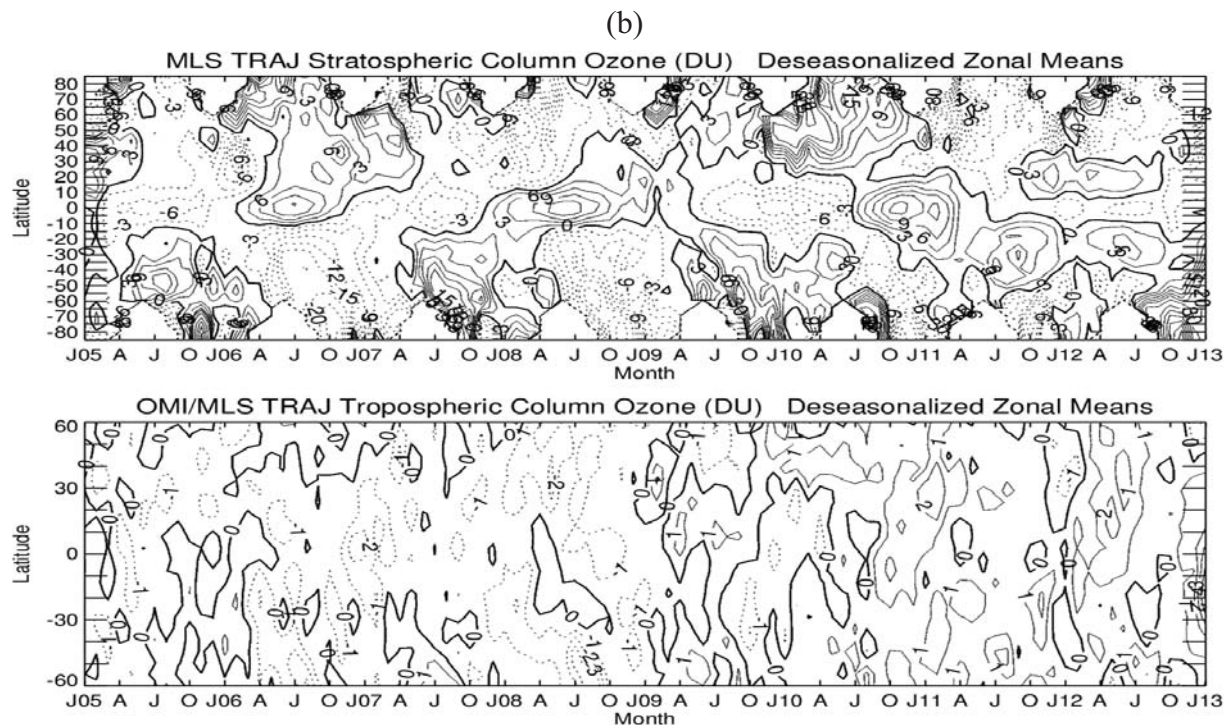


**Figure 15.** (top) Monthly zonal mean SCO in Dobson Units for 2005-2012 from the data assimilation product. (bottom) Same as top panel but instead for TCO. Dark to light shading in each panel designates smaller to higher column amounts, respectively. The contour values for the top panel (bottom panel) increments by 20 (3) Dobson Units.

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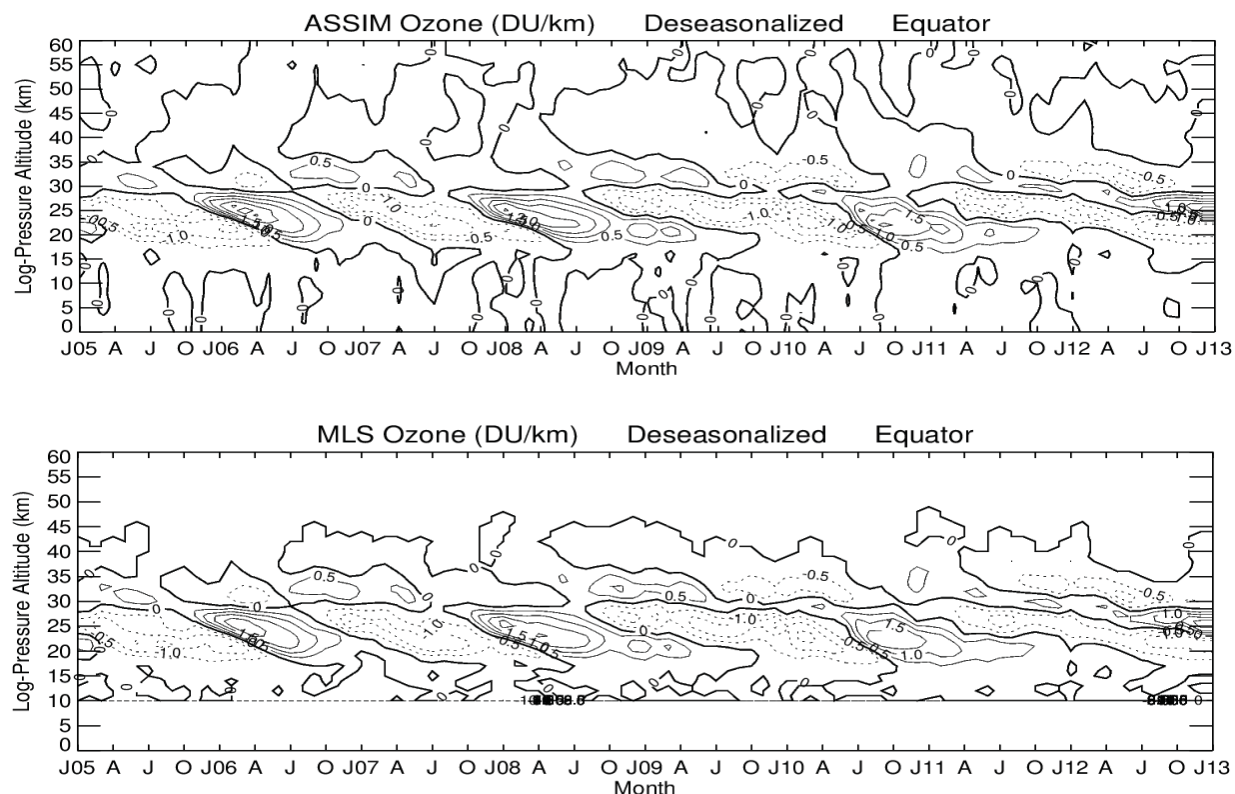


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**Figure 16.** (a) Contour diagrams of the same ozone data plotted in Figure 15 but with all measurements deseasonalized. The contour increments for the top panel (bottom panel) is three (one) Dobson Units. (b) Same as (a), but instead for TRAJ SCO and TCO measurements.

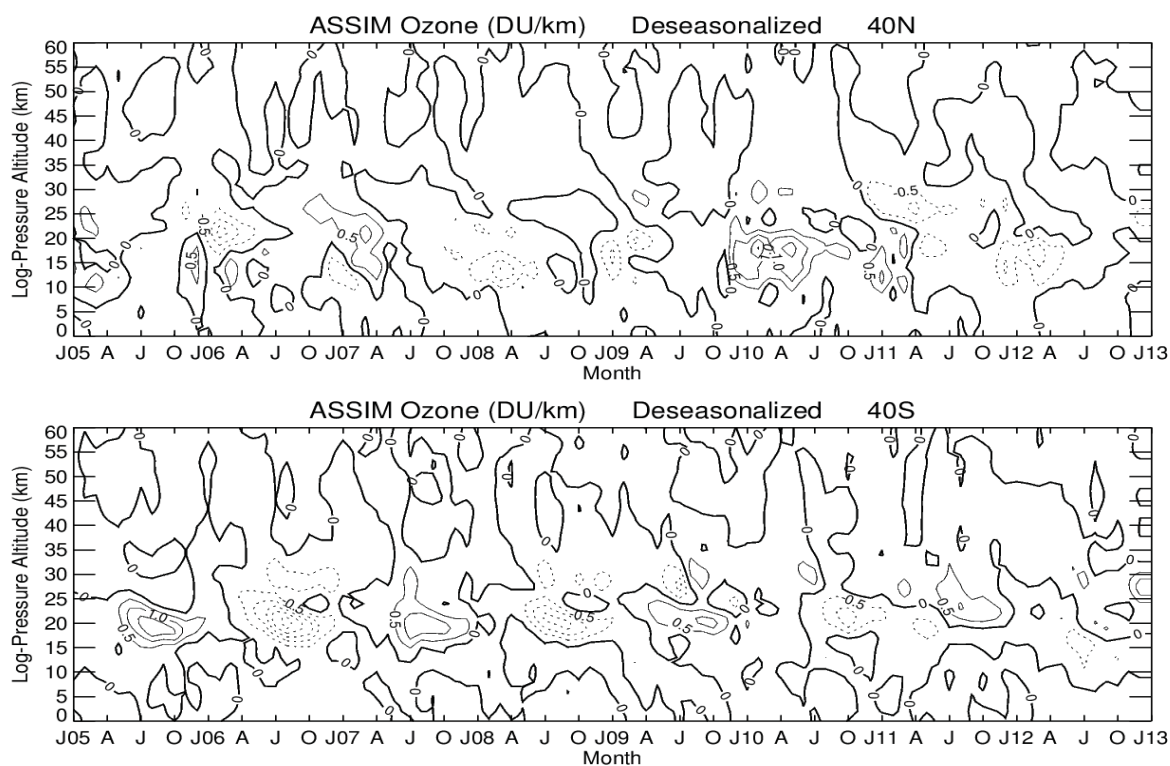
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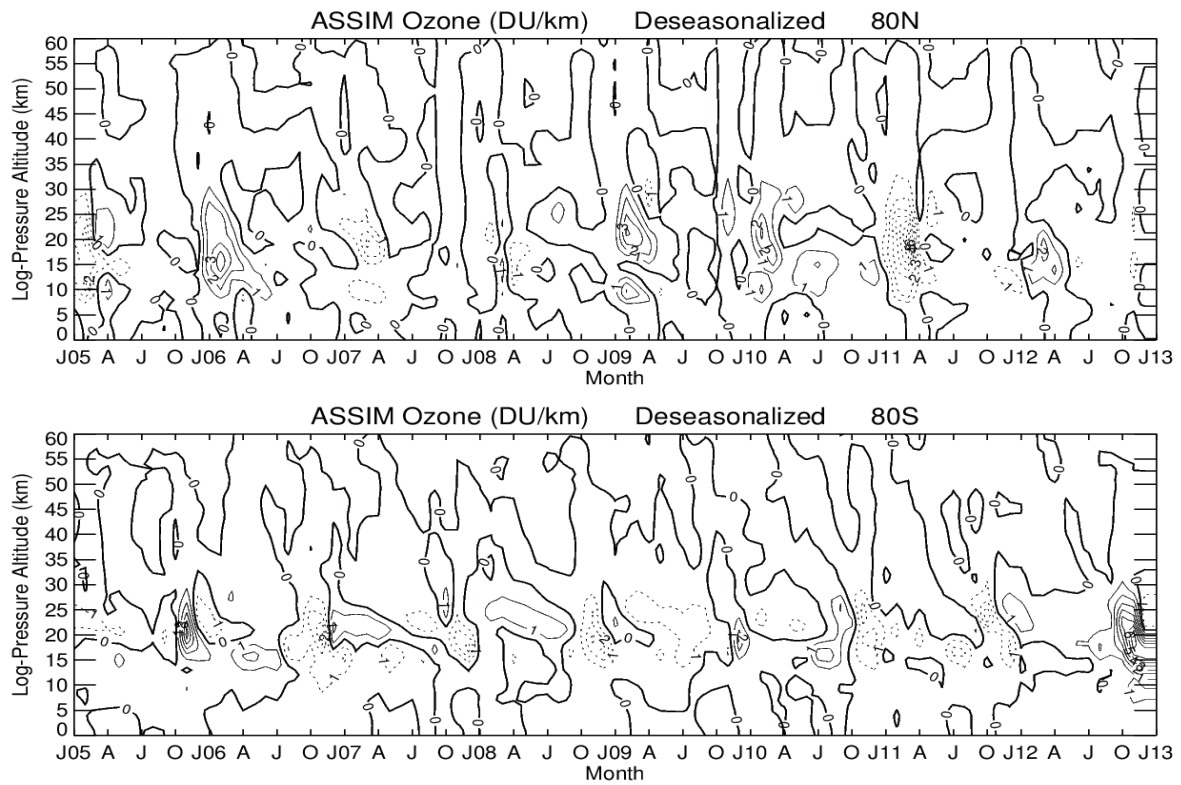


**Figure 17.** (top) Equatorial ozone vertical profile gradients (in units  $\text{DU}\cdot\text{km}^{-1}$ ) from the data assimilation product plotted as log-pressure altitude (in units km) versus month. The vertical gradients are derived from monthly means by interpolating the product log-pressure surface ozone measurements to pressure levels  $1013.25 \cdot \exp(-z/7)$  where  $z$  is log-pressure altitude in units km and the pressure is in units hPa. (bottom) Same as (top) but for MLS ozone profiles.





**Figure 18.** (top) Same as Figure 17 for ASSIM ozone but instead at latitude 40°N. (bottom) Same as Figure 17 for ASSIM ozone but instead at latitude 40°S.



**Figure 19.** (top) Same as Figure 17 for ASSIM ozone but instead at latitude 80°N. (bottom) Same as Figure 17 for ASSIM ozone but instead at latitude 80°S.