SUPPORTING MATERIAL.

A. Validation and Adjustments Made to OMI/MLS TCO.

An OMI instrumental data artifact known as the "row anomaly" affects the quality of level-1B radiance measurements, and subsequently the quality of the orbital level-2 and gridded level-3 ozone measurements. The row anomaly is an instrumental blockage in the optical path that expanded greatly in late January 2009 to affect greater than one-third of all 60 scan positions for ozone retrieval.

The OMI standard total ozone product has extensive corrections made for the row anomaly problem, but we have found that for TCO there still remains a small error of ~0.5 to 1.0 DU over the Aura record. We have evaluated the OMI/MLS TCO product for the OMI row anomaly and have made corrections for this. Figure S1 compares monthly time series differences of OMI/MLS standard product TCO minus OMI/MLS row-isolated TCO (i.e., only OMI rows 3-18 included for total column ozone in orbital measurements) (see figure caption). The result of this row anomaly evaluation is that the OMI standard total column ozone used to derive TCO has a small average artificial drift of ~+0.5 to +1.0 DU-decade^{-1} due to the OMI row anomaly. An incremental change is visible around January 2009 in Figure S1 when the row anomaly problem became escalated. As a conservative approach we applied a mean -1.0 DU-decade^{-1} adjustment to OMI/MLS TCO. This adjustment is relatively small compared to the ~+3 DU-decade^{-1} or greater trends calculated over India/East Asia in Figure 1 for OMI/MLS TCO.

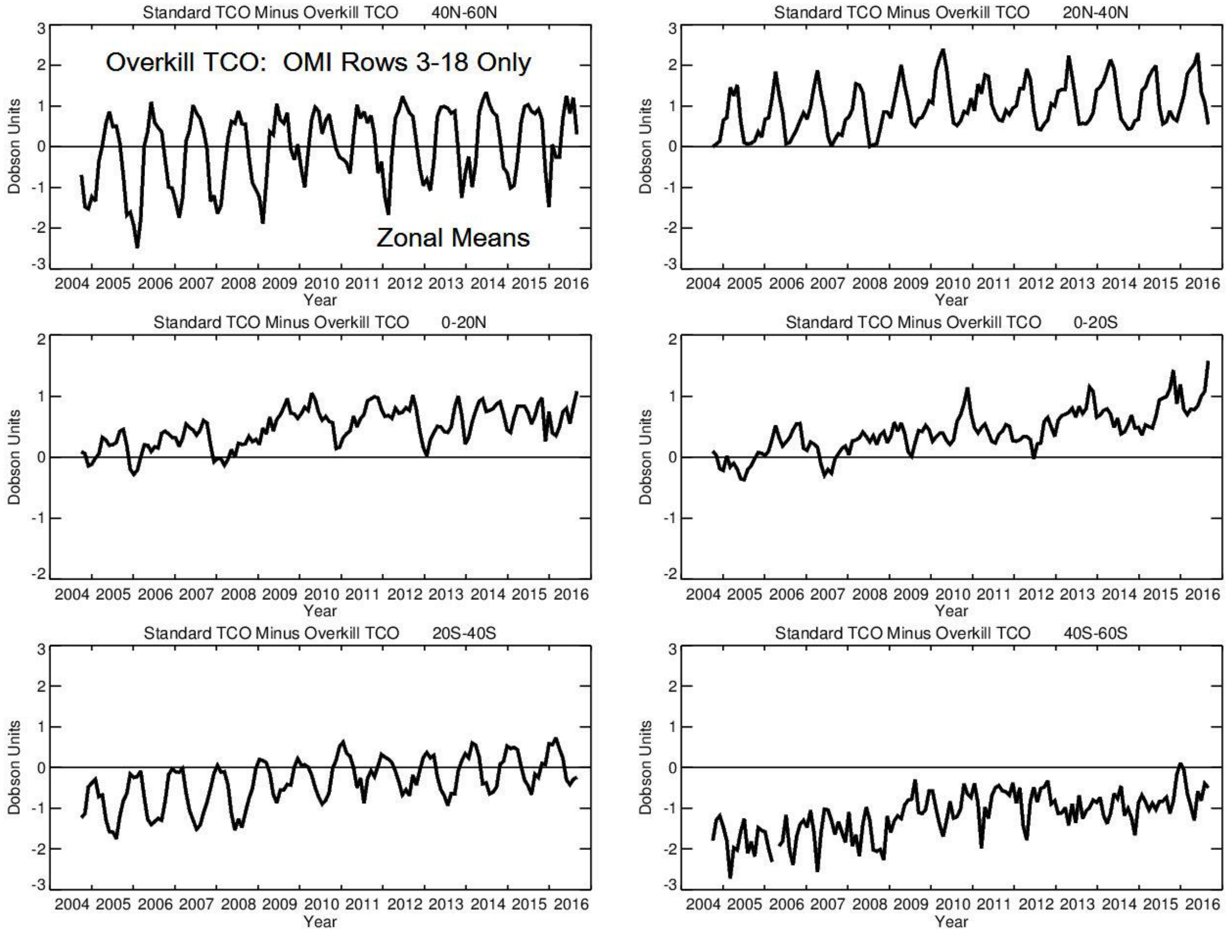

Figure S1. Monthly time series differences of OMI/MLS standard TCO minus OMI/MLS row-isolated TCO (i.e., only OMI rows 3-18 included for total column ozone for entire record) for estimating artificial drift due to the row anomaly error in standard OMI total ozone measurements. Differences are averaged over 20^o latitude bands (indicated). These differences indicate that the standard product TCO from OMI/MLS has an artificial drift varying from about +0.5 to +1.0 DU-decade^{-1} with a small incremental change visible around January 2009 when the row anomaly problem became escalated.

We have compared the row anomaly adjusted OMI/MLS TCO with global ozonesondes for several sites over the globe. A total of 29 ozonesonde stations were selected based on having a sufficiently large number of daily ozone profiles for the comparisons during October 2004 through May 2016 (Figure S2). The number criterion was based on at least 50 (20) daily 1-1 co-located matchups for the first and last 5-year periods for 12-month averages (June-September averages). The differences (final 5-year minus beginning 5-year average) invoked a t-test (e.g., Wolf, 1962).

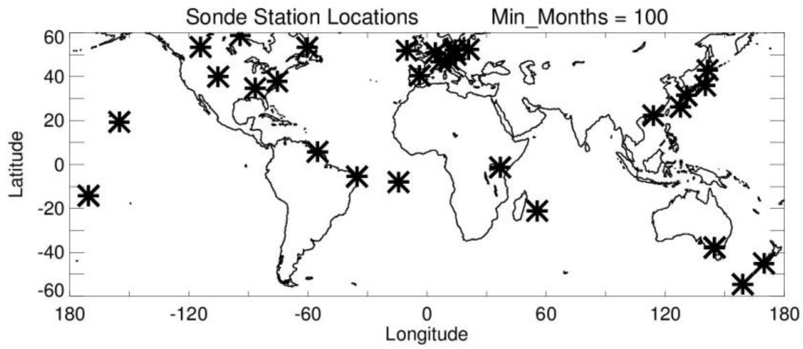

Figure S2. Locations for the ozonesonde stations for the ozonesonde measurements used to evaluate the OMI/MLS TCO product for potential drift and/or offset. Each station has at least 100 months of measurements (and at least 3 daily profiles each of these months) covering the period October 2004 through May 2016.

Figure S3 shows OMI/MLS TCO comparisons with ozonesonde TCO to test the OMI/MLS product for potential long-term measurement drift and mean offset. Nearly all ozonesonde measurements are from Electrochemical Concentration Cell (ECC) instruments. All co-located TCO for each of the 29 stations were first measured daily and then averaged over the beginning and ending 5-year records for October 2004 – May 2016 (see figure caption).

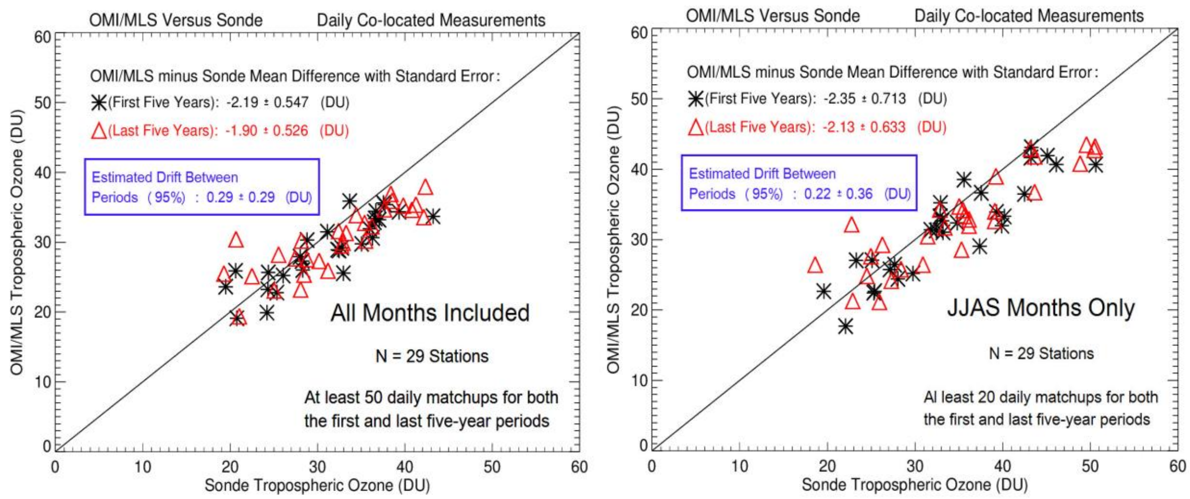

Figure S3. (Left) OMI/MLS TCO versus ozonesonde TCO for the 29 stations in the analysis. The ozonesonde TCO measurements each day used the same NCEP WMO 2K-km^{-1} lapse-rate tropopause as used for OMI/MLS. The ozonesonde record of daily coincident TCO is 1 October 2004 to 31 May 2016. All OMI/MLS and ozonesonde measurements for each station were daily and then averaged over the beginning 5-year record (black asterisks) and ending 5-year record (red triangles). The estimated relative drift (OMI/MLS minus ozonesonde) including $\pm 2\sigma$ between the beginning and ending 5-year periods is +0.29 DU ± 0.29 DU. A two-sided t-test was used for these difference calculations. (Right) Same as left panel, but including only daily measurements for the months June-July-August-September (JJAS). Estimated drift for JJAS is +0.22 DU ± 0.36 DU.

The main conclusion from Figure S3 is that according to the ozonesondes there is only a very small positive drift detected in OMI/MLS TCO of around +0.2 to +0.3 DU between beginning and ending 5-year records that is not statistically significant when based upon a difference t-test. The ozonesondes also indicate that OMI/MLS TCO is too small by about 2 DU. We have included a mean +2 DU constant offset adjustment to the OMI/MLS TCO measurements based on the ozonesondes.

We have also tested possible relative drift between OMI and MLS ozone retrievals by comparing their independent measurements of SCO. Figure S4 in the left panel shows OMI SCO from the

CCD method (black solid curve) plotted with MLS SCO (red dotted curve). All values represent monthly means and are averaged over the tropical Pacific (indicated). The right panel in Figure S4 shows the difference of these two curves (MLS minus OMI) along with a low-pass filtering of this same difference curve for visualization. This test implies that OMI and MLS ozone measurements are well behaved over long record with no obvious relative drift other than ~0.5 DU-decade^{-1} which is not statistically significant. Most differences in Figure S4 are QBO related with one instrument measuring the QBO signal in SCO at nadir (OMI) and the other from limb (MLS).

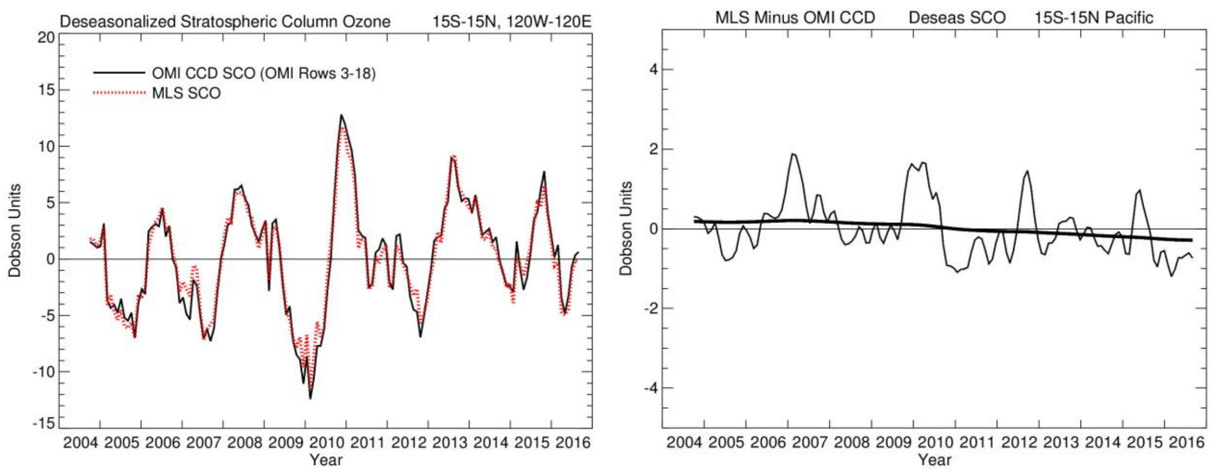

Figure S4. (Left) Monthly time series (in DU) of OMI SCO derived from the CCD method (solid black curve) over-plotted with MLS SCO (dotted red curve) beginning October 2004. All measurements were averaged over the tropical Pacific (indicated) where the CCD measurements of SCO are optimal for comparing with MLS SCO. The OMI measurements used only OMI scan rows 3-18 (out of a total of 60 rows for each side-to-side scan) to avoid OMI row-anomaly problems with retrieved ozone. (Right) The time series difference of MLS SCO minus CCD SCO (thin black curve) and a low-pass filtering of the difference curve (thick black curve). There is no statistically significant drift measured between MLS and OMI SCO.

In summary, our cross-evaluations with ozonesondes and independent SCO measurements from MLS and OMI, as well as the OMI row anomaly shows that the OMI/MLS TCO data product is well behaved for the entire Aura time record for evaluating trends. Corrections made to the

OMI/MLS TCO were very small. These corrections included a +2 DU offset adjustment (via ozonesonde comparisons) and a conservative -1.0 DU-decade^{-1} drift adjustment (via row anomaly analysis).

B. OMPS Nadir-Mapper/Limb Profiler Tropospheric Ozone.

An important yet small addition to this study is to show some evaluation of TCO derived from Suomi NPP OMPS measurements. We produced global monthly mean measurements of OMPS TCO for January 2012 through June 2017. OMPS TCO is measured similar to OMI/MLS TCO. OMPS v2.5 limb-profiler (LP) ozone profiles for all three combined slit measurements are first integrated vertically each day to determine SCO. The SCO fields are then filled in each day using 2D Gaussian + linear interpolation. The SCO is then subtracted from OMPS nadir-mapper v8.6 total ozone measurements to derive gridded TCO. An example of monthly-mean OMPS TCO for July 2016 is shown in Figure S5.

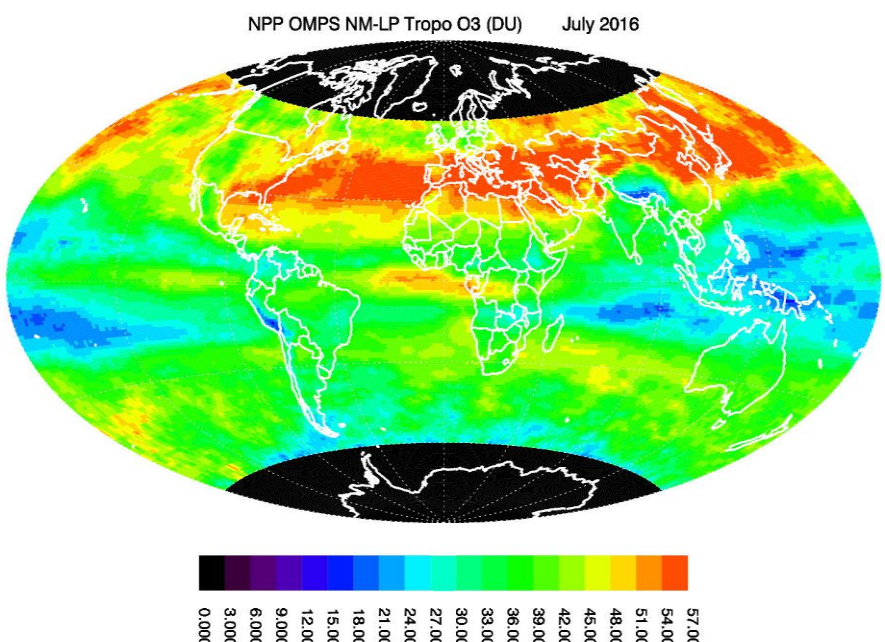

Figure S5. TCO (in DU) averaged for July 2016 from combined OMPS nadir-mapper and OMPS limb-profiler measurements (see text).

Figure S6 shows differences in monthly zonal means for OMPS and OMI/MLS averaged over January 2012 through June 2017. A generally constant offset of about 4 DU (OMPS being higher) occurs over most of the useful latitude range. Following this +4 DU offset between OMPS and OMI/MLS, the OMPS tropospheric ozone was adjusted at all latitudes by -2 DU following the +2 DU ozonesonde offset applied to OMI/MLS TCO.

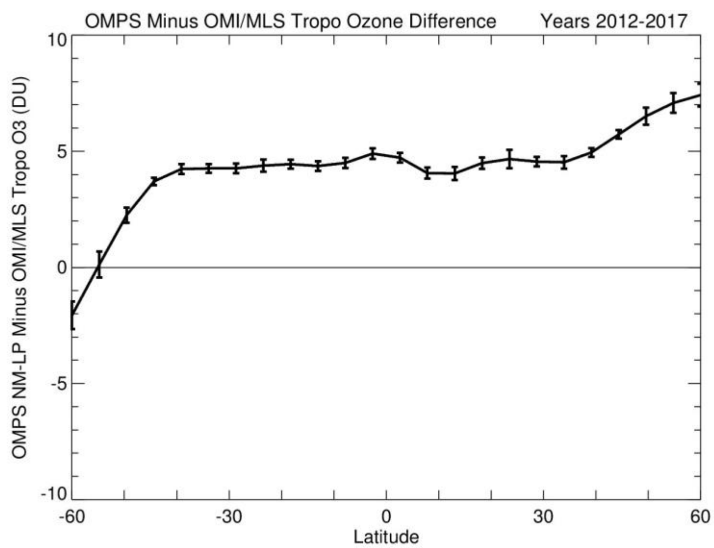

Figure S6. Differences of monthly zonal-mean OMPS TCO minus OMI/MLS TCO for the data overlap period of January 2012 through June 2017.

Figure S7 shows comparisons of time series for OMPS TCO (blue curves) and OMI/MLS TCO (dotted red curves) for selected sites. Included sites are Java, Brazil, Washington DC, and Beijing. In mid-latitudes the dominant variability is the seasonal cycle, but in the tropical latitudes there is considerable inter-annual change, in particular for Java due to ENSO events.

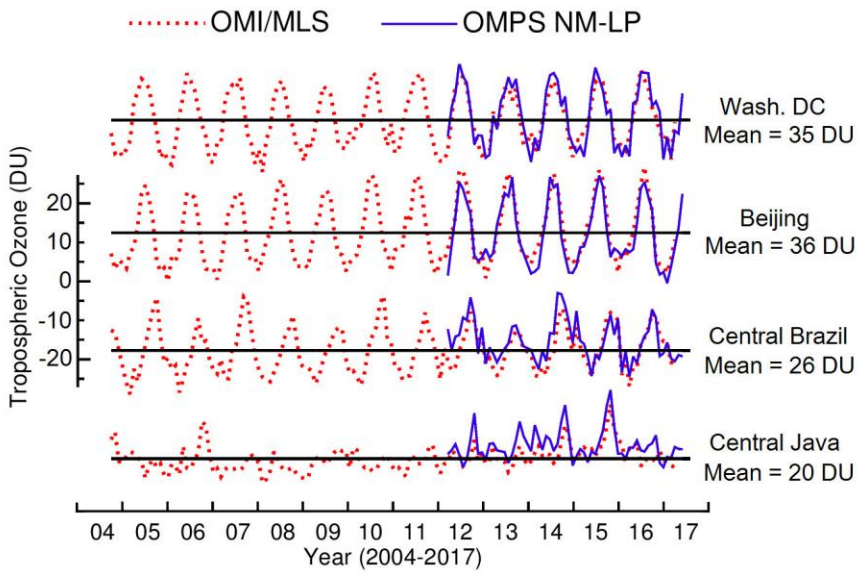

Figure S7. Monthly-mean time series of OMI/MLS TCO and OMPS TCO at several sites plotted together following the offset adjustment of Figure S6.

C. Validation of TOMS TCO Measurements.

We noted in section 2 that the TOMS TCO measurements included validation assessments by Ziemke et al. (2005, and references therein). For TOMS TCO the main difficulty with validating against ozonesondes is obtaining a sufficient number and long record of good quality ozonesonde data back to the 1970's and 1980's. Ziemke et al. (1998) compared CCD TCO for the Nimbus-7 record (1979-1993) with ozonesondes that were mostly entirely of the Brewer-Mast instrument design and had large profile correction factors (often ~30%). Those comparisons were at best only useful for evaluating basic properties of seasonal cycles. Ziemke et al. (2005) used SHADOZ ozonesonde comparisons for the Earth-Probe record from 1996-2005. A large fraction of SHADOZ ozonesondes, particularly over the last decade, are of the much more accurately-measuring ECC design. Figure S8 shows SHADOZ/TOMS comparisons of TCO monthly time series.

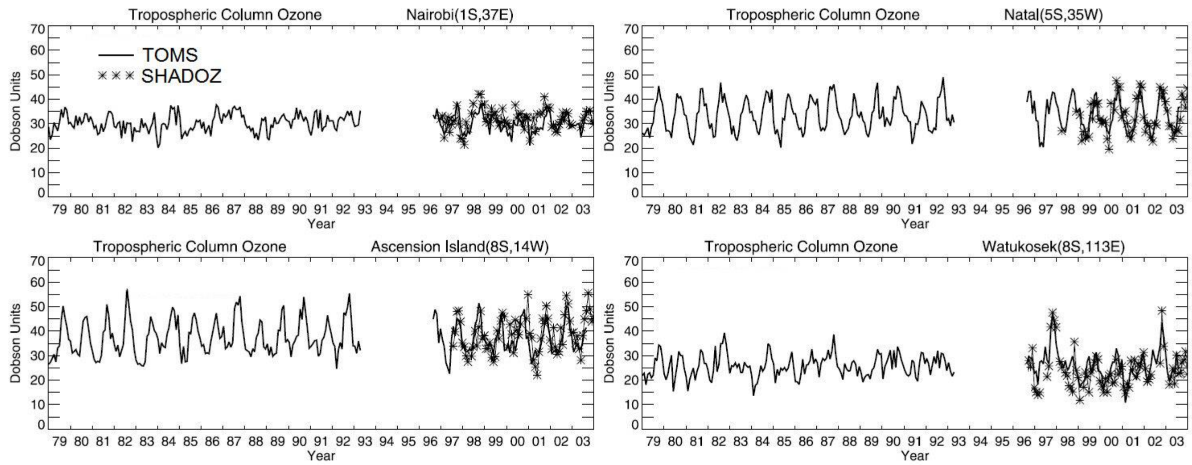

Figure S8. Coincident time series of TOMS CCD (dark solid curves) and SHADOZ (asterisks) tropospheric column ozone in Dobson Units. The stations plotted are Nairobi, Natal, Ascension Island, and Watukosek. This figure is adapted from Ziemke et al. (2005).

D. Trend Comparisons Between Ozonesondes, OMI/MLS, and GMI Tropospheric Ozone.

We calculated linear trends in column ozone from sondes for 2005-2016 and compared these with the linear trends calculated from OMI/MLS and GMI TCO in Figure 1. Figure S9 shows the ozonesonde stations sites for these comparisons. In total there were 27 sites based upon minimum statistical conditions including at least a total of 2 ozonesondes per given month extending over at least 8 years for the 12-year record. Most all of the ozonesonde measurements are from ECC instruments that tend to be generally well calibrated over long record and between different stations.

Figure S10 plots the trend comparisons. The ozonesonde trends in Figure S10 show large spread in both sign and magnitude in the NH compared to either OMI/MLS or GMI trends which are generally consistent between them everywhere except in the SH extra-tropics. In the SH extra-tropics the ozonesondes indicate no viable trend, similar to GMI TCO which suggests that the near-zero trends throughout the SH extra-tropics determined by GMI in Figure 1 are more correct than OMI/MLS. It is noted however that the positive trends for OMI/MLS TCO in the SH extra-tropics are primarily over remote ocean regions and not in vicinity of the sondes station sites. In the tropics there is one (highlighted) station, Costa Rica, where the ozonesonde trend is clearly negative and opposite the positive trends measured for both OMI/MLS and GMI TCO. A reason for the negative ozonesonde trends at Costa Rica relates to volcanic SO_2 within the sonde detector during the latter years that reduced the detected ozone in the 2-7 km altitude range (e.g., Witte et al., 2018).

A main result from the ozonesonde trend evaluation in Figure S10 is that the short Aura record combined with limited measurements for the ozonesondes precludes any definitive quantitative trend determination and trend comparisons with OMI/MLS and GMI TCO. Yet, there is still a general consensus in Figure S10 of an overall increase in tropospheric ozone from OMI/MLS, GMI, and the ozonesondes.

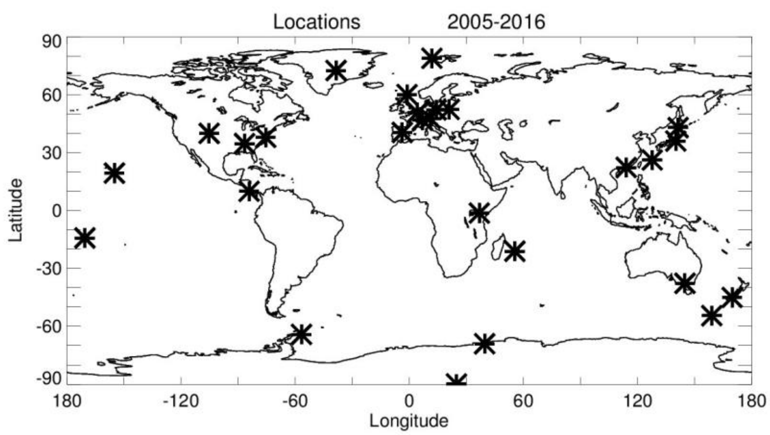

Figure S9. Ozonesonde station sites (27 in total) for ozonesonde linear trend calculations (see text).

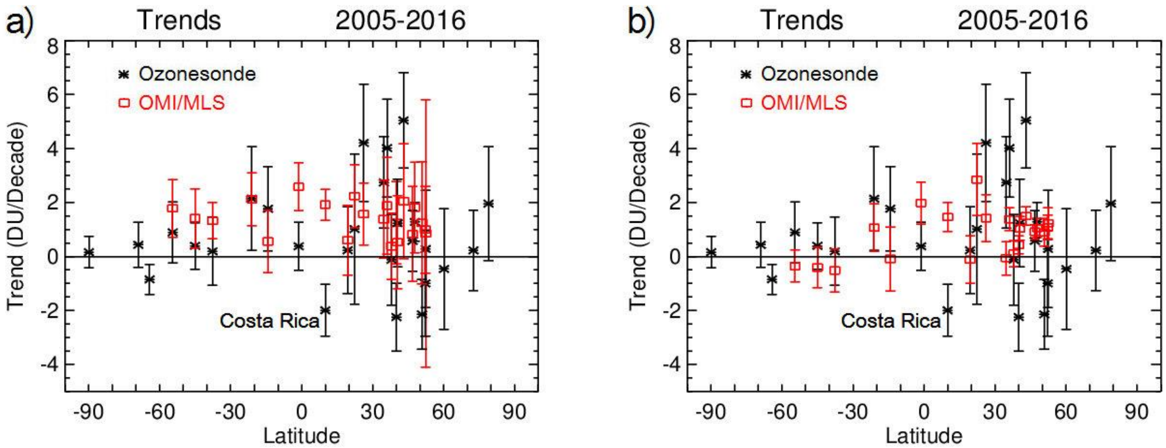

Figure S10. (a) Linear trends in tropospheric ozone for 2005-2016 calculated for OMI/MLS monthly mean TCO (red squares) and ground-to-8 km column ozone from ozonesondes (black asterisks). The trends for OMI/MLS TCO used the MLR method discussed in section 2.3 while trends for the ozonesondes were calculated from a line fit regression using the daily profile measurements. All calculated trends include ±2σ statistical uncertainties. (b) Same as (a) but instead for MERRA-2 GMI monthly mean TCO.