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Correlating tropospheric column ozone with tropopause folds: the Aura-OMI satellite data

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OMI tropopause folds

Q. Tang and M. J. Prather

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

The geographic and temporal variations in tropospheric and stratospheric ozone columns from individual swath measurements of the Ozone Monitoring Instrument (OMI) on the NASA Aura spacecraft are reasonably well simulated by the University of California, Irvine (UCI) chemistry transport model (CTM) using $1^\circ \times 1^\circ \times 40$ -layer meteorological fields for year 2005. From the CTM we find that high-frequency spatial variations in tropospheric column ozone (TCO), including around the jet streams, are not generally correlated with variations in stratospheric ozone column, but instead are collocated with folding events involving stratospheric-origin, high-ozone layers below the tropopause. The CTM fold events are verified in many cases with available ozone sondes. Using the OMI Level 2 profiles, and defining tropopause height from our CTM using the European Centre for Medium-Range Weather Forecasts (ECMWF) fields, we find that most of the variations in TCO near CTM folding events are also not correlated with those in stratospheric ozone column. The absolute tropospheric columns from OMI and CTM agree swath-by-swath, pixel-by-pixel within ± 5 Dobson Units (DU) for most cases. Notable exceptions are in the tropics where neither the high ozone from biomass burning nor the low ozone in the convergence zones over the Pacific is found in the OMI observations, likely because of OMI's insensitivity to the lower troposphere. A separate bias is identified with the OMI profiles near the southern sub-tropical jet. The CTM has a high bias in stratospheric column outside the tropics, due to problems previously identified with the stratospheric circulation in the 40-layer meteorological fields. Overall, we identify ozone folds with short-lived features in TCO that have scales of a few hundred kilometers as observed by OMI.

1 Introduction

Stratosphere-troposphere exchange (STE) plays an important role in determining the chemical composition in the atmosphere, bringing O_3 -rich stratospheric air into the

ACPD

10, 14875–14896, 2010

OMI tropopause folds

Q. Tang and M. J. Prather

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



OMI tropopause folds

Q. Tang and M. J. Prather

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

troposphere (Danielsen, 1968), affecting the oxidative capacity of the atmosphere (Levy, 1972; Crutzen, 1973). Many studies have aimed to quantify the STE flux (e.g., Danielsen, 1968; Holton et al., 1995; Appenzeller et al., 1996; Stohl et al., 2003; Jaeger and Sprenger, 2009). This is a global problem that requires global observation and modeling. We identify STE O₃ flux with many tropopause folds (TF) in our chemistry transport model (CTM) and then show that these folds are observed as variations in tropospheric column ozone (TCO) on a daily global basis by the Aura Ozone Monitoring Instrument (OMI) satellite measurement.

Tropopause folding in the vicinity of both subtropical and polar jets have been observed to be a particularly important process leading to STE (e.g., Danielsen, 1968; Lamarque and Hess, 1994; Beekmann et al., 1997; Baray et al., 2000; Traub and Lelieveld, 2003). TFs facilitate great amount of STE flux, although not all of the material in the folds enters the troposphere (Hsu et al., 2005). We have looked for evidence of TF in the four Aura ozone instruments. In our model and ozone sonde data most folds are about 1–2 km thick and occur between 150–300 hPa. The Microwave Limb Sounder (MLS) with only three levels below 100 hPa is unable to resolve most folds. The High Resolution Dynamics Limb Sounder (HIRDLS) with higher vertical resolution still has no useful signal below 150 hPa. We are left with identifying TF in the tropospheric data from the Tropospheric Emission Spectrometer (TES) and OMI. Neither instruments, however, can resolve most folds vertically and thus identification requires matching geographic anomaly patterns in the tropospheric columns. The wide swath data from OMI is our first choice and is analyzed here.

First, we evaluate the CTM using ozone sonde data, finding good agreement for the 638 exact matches between 35° S and 40° N in year 2005. The criteria of detecting TF in the CTM are described at the end of Sect. 2. Good consistency between the OMI and CTM TCO and total ozone is shown in Sect. 3. In Sect. 3.1, we find TFs are correlated with anomalies in TCO but not in total ozone. Furthermore, we try to link TCO anomalies with STE O₃ flux in Sect. 4 and find good correlation in the vicinity of sub-tropical jets but not in high latitudes.

2 Chemistry transport model and ozone sondes

The chemistry transport model (CTM) is driven by pieced-together, spun-up forecasts from the Integrated Forecast System of the European Centre for Medium-Range Weather Forecasts (ECMWF) developed in collaboration with University in Oslo. The CTM is run at $1^\circ \times 1^\circ \times 40$ -layer spatial resolution with ~ 1 km vertical resolution near the tropopause. The uppermost layer is exceedingly coarse (2–22 hPa). The chemistry scheme is a combination of the ASAD software package for the troposphere (Carver et al., 1997; Wild et al., 2003) and linearized ozone scheme for the stratosphere (Linov version 2) (Hsu and Prather, 2009). The ASAD algorithm has been entirely rewritten at UCI, including the steady-state assumptions to optimize the solver. We use the Year-2000 emissions from the EU Quantifying the Climate Impact of Global and European Transport Systems (QUANTIFY) project, with monthly biomass burning emissions from the average multi-year (1997–2002) Global Fire Emissions Database (Hoor et al., 2009). Lightning NO_x ($\text{NO} + \text{NO}_2$) is scaled linearly to the deep convective mass flux with annual total of $5.x \text{ Tg N yr}^{-1}$ for Year-2005. The resulting tropospheric ozone for year 2005 is typical: Northern Hemisphere (NH), 42.8 Dobson Units (DU); Southern Hemisphere (SH), 32.8 DU; STE flux, 590 Tg yr^{-1} ; surface deposition, 760 Tg yr^{-1} ; and thus net photochemical production, 170 Tg yr^{-1} .

To simulate the Aura ozone instruments, we store the CTM O_3 in 3-D for each OMI swath, saving two datasets 30-min apart to interpolate the exact time for each pixel ($\sim 63 \text{ GB yr}^{-1}$ in real*4 format). We also save $65^\circ \text{ S} - 65^\circ \text{ N}$ every two hours to match ozone sondes ($\sim 29 \text{ GB yr}^{-1}$ in real*4 format). Therefore, we can generate tracks for HiRDLS, MLS, OMI, and TES.

A critical evaluation of the modeled ozone, particularly with regard to TFs is found with the ozone sonde measurements. In this test we restrict our comparison range from 35° S to 40° N where the CTM predicts the preponderance of TF, giving 638 World Ozone and Ultraviolet Radiation Data Centre (WOUDC) ozone sondes from 20 stations for 2005 (HKO et al., 2009). Ozone sondes have much finer vertical resolution than the

OMI tropopause folds

Q. Tang and M. J. Prather

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

OMI tropopause folds

Q. Tang and M. J. Prather

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

CTM. Figure 1 shows four examples from Hong Kong (Fig. 1a), Ankara (Fig. 1b), and Huntsville (Fig. 1c, d). The CTM (blue solid line) agrees well with sonde (green dash line) in Fig. 1a, except for overestimation in the boundary layer, probably due to the redistribution of pollution plumes in the $1^\circ \times 1^\circ$ grid. Both CTM and sonde show fold structure near 251 hPa, but the CTM misses the fine structure reported by sonde at 140 hPa, leading to an offset of 50 ppb in the profile. In Fig. 1b, CTM and sonde closely match with a O_3 maximum at 400 hPa. In Fig. 1c, model and sonde are similar from the surface to 250 hPa with a fold structure near 158 hPa, although of different magnitude. Comparison in Fig. 1d is the worst: CTM predicts an increase while sonde detects a decrease around 250 hPa; fold structure can be found in both from 100–158 hPa; but the patterns differ enormously. Overall, we grade 50% of these 638 comparisons as “A” (e.g., Fig. 1a, b), 30% as “C” (e.g., Fig. 1c), and 20% as “F” (e.g., Fig. 1d).

The reported OMI profiles (de Haan and Veefkind, 2009) include 3–6 tropospheric layers, but really contain only one degree of freedom in the troposphere. Thus, TF detected by OMI would be represented as an overall enhancement in column, but not as a fold defined by sonde or CTM. OMI profiles (magenta dot dash line) generally underestimate ozone values in lower and middle troposphere but overestimate them in upper troposphere and lower stratosphere (Fig. 1).

An objective criteria to identify TF is developed for the CTM based on subjective evaluations of the 638 sonde comparisons. TFs are defined in a boolean sense (Yes or No) from the vertical profile of O_3 abundance: starting above 5 km, once the O_3 exceeds 80 ppb and then within 3 km above decreases by 20 ppb or more to a value less than 120 ppb. Note that TF detection depends in part on the vertical resolution of the CTM. Given this experiment the uppermost tropospheric layer in fold must be at least 1 km thick. Also note that by these criteria, TFs can be caused by STE events and/or biomass burning. For example, Fig. 2 shows simulated global TCO maps for two “daily” (25-h periods) OMI swath observations, in which the $1^\circ \times 1^\circ$ pixels with a TF event are marked with “+”.

3 Satellite observation and tropospheric columns ozone

The tropospheric column ozone (TCO) requires knowledge of the tropopause. The UCI CTM uses an artificial tracer emitted uniformly at the surface with uniform e-fold decay of 90 days (e90) to determine the boundary between troposphere and stratosphere.

5 The e90 tracer allows for dynamic allocation between stratospheric and tropospheric air masses instantaneously in 3-D for the purpose of the chemical model and diagnostics. It also provides a measure of the tropopause that is consistent with the traditional definition based on the 1-D temperature lapse rate and matches the sondes. Folds typically have tropospheric air masses overlying stratospheric air, and we identify the tropopause as the upper boundary of the uppermost model layer identified as tropospheric by its mean e90 abundance. Thus, our TCO includes all O₃ below this uppermost tropopause. The CTM TCO is shown in Fig. 2, beginning at 00:00:00 UTC for 25 h on 10 June and 3 December 2005. Note that anomalies of high TCO are often co-located with TF events.

15 The OMI instrument aboard the NASA's EOS Aura satellite has measured atmospheric composition from a sun-synchronous polar orbit since 9 August 2004 (Schoberl et al., 2006; OMI Team, 2009). With a 2600-km swath width, OMI essentially provides daily global coverage. Ziemke et al. (2006) derived global TCO from the OMI level 3 (L3, gridded/daily) and MLS level 2 (L2, orbit/swath) datasets, following the tropospheric ozone residual method (Fishman et al., 1990) by subtracting MLS stratospheric column from the OMI total column. The tropopause pressure was determined from NCEP re-analysis data. The resulting TCO cannot retain the heterogeneous spatial and temporal structures in OMI L2 data, which provide important clues to the existence of TFs. The use of OMI L3 data diffuses TCO features at high latitudes because different swath data (at least 90-min apart) are averaged. Further, in the overlap region necessary to achieve daily global maps, averaging is done over swaths 24-hour apart, even in the tropics. The MLS L2 data (6-km cross-track width) (Livesey et al., 2007) must be extended across the OMI swath (2600-km cross-track width), usually by

OMI tropopause folds

Q. Tang and M. J. Prather

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



interpolation with the next orbit, thus some of the geographical variations in TCO are actually aliased from the variability in the OMI stratospheric column O_3 .

In this study, we use the OMI L2 ozone profile dataset (OMO3PR V003) for year 2005 (de Haan and Veefkind, 2009). The 18-layer ozone profile is based on the Optimal Estimation method (Rodgers, 2000) and derived from the signals of UV1 channel (270–308.5 nm) and part of the UV2 channel (311.5–330 nm), covering from the surface to 0.3 hPa. The Differential Optical Absorption Spectroscopy (DOAS) algorithm is implemented to retrieve the total ozone (Bhartia, 2002; Veefkind et al., 2006). Horizontal resolution of the OMI profiles (used here for TCO) is 13 km×48 km (along-track × cross-track) and that of the column (used here for total column ozone) is 13 km×24 km with a cross-track swath width of 2600 km. Gaps between adjacent pixels in the raw OMI data disappear when accumulated onto the 1°×1° grid of the CTM. We select the same tropopause height as in the CTM simulation for each 1°×1° pixel and interpolate the OMI profile to get TCO.

3.1 Swath-by-swath comparison

OMI L2 swath data allows us to study TFs on an hourly basis, which is important since they are not static over a day. Generally there is very good agreement between the modeled and measured ozone columns, both total and TCO. Importantly, the TCO anomalies are not correlated with total ozone anomalies which would be the case if we were aliasing upper tropospheric meteorologies (ridges and troughs) as STE intrusions. Thus, we find TFs drive variations in TCO but not in total column.

Analysis of the 25-h swaths beginning at 00:00:00 UTC for 10 June are shown in Fig. 3a, c and Fig. 4a, c and for 3 December in Fig. 3b, d and Fig. 4b, d. The total ozone columns for OMI are shown in Fig. 3a, b, while that for the CTM are in Fig. 3c, d. The UCI CTM successfully reproduces the patterns, but overestimates in most extra-tropical regions by about 15%. We recognized this problem with the stratospheric circulation of the 40-layer ECMWF wind field (Hsu and Prather, 2009).

The TCO patterns (Fig. 4) are also quite similar, with smaller absolute error than for

OMI tropopause folds

Q. Tang and M. J. Prather

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



total ozone. In both OMI and CTM a band of high TCO (~ 40 DU) at 30° N spreads from the Eastern Asia, across the Pacific and North America, to the central Atlantic. In December, the maximum regions to the east of Australia also appear in both. At 28° S, however, OMI TCO has a narrow band of high TCO (~ 45 DU) across all longitudes, which seems unphysical and has no analog in the CTM. This difference might be due to the climatological a priori applied in the retrieval algorithm (de Haan and Veeffkind, 2009). The O_3 -rich areas over tropical Africa and South America due to biomass burning in the CTM are missing in OMI, most likely because OMI is less sensitive to lower tropospheric O_3 (Zhang et al., 2010). Likewise, OMI does not detect the very low TCO over equatorial western and central Pacific, where the low O_3 abundances are near the surface.

3.2 Bias and variability in the CTM tropospheric ozone

Analysis of the time series of TCO for the months of June and December are shown in Figs. 5 and 6, respectively. The patterns of the monthly mean differences (CTM–OMI) show smooth large-scale features that change only little from month to month as shown in Figs. 5c and 6c. For most of the daylit globe (56% in June and 65% in December), the differences are within ± 5 DU. Larger, positive biases exist over Africa and South America. In June, the greatest differences are located at southern Africa, while in December large biases concentrate over central Africa, following the seasonality of biomass burning. Likewise, differences over South America are more enhanced in December than in June. Significant low biases occur over western equatorial Pacific for both months related to the ozone loss in the marine boundary layer, where the CTM matches the typically low 5–15 ppb observed in the tropical Pacific (Davis et al., 1996; Crawford et al., 1997; Browell et al., 2003). In June, there is an extensive longitudinal extent of low CTM biases between 30° N and 40° N off the east coast of the continents, and this may be related to the similar problem of southern sub-tropical jet in December.

Consistency between the instantaneous CTM and OMI TCO is shown by the 2-D probability density function (PDF) (Figs. 5d and 6d), which represents two million in-

OMI tropopause folds

Q. Tang and M. J. Prather

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



stant, individual comparisons per month. The highest densities lie along the 1:1 line (black bold line) and errors are generally symmetric, showing little overall bias. The probability of each comparison is weighted by the pixel area and frequency of observation as a function of latitudes. High latitude swaths overlay one another and result in oversampling. There is a low bias in the CTM for OMI ranges 20–35 DU in June, corresponding to tropical oceans, SH mid-latitudes, and Greenland.

Removing the monthly mean TCO at each $1^\circ \times 1^\circ$ pixel, we can test the meteorologically driven variations. In terms of the monthly standard deviation (σ , STD), the CTM matches OMI over much of the globe (Figs. 5a, b and 6a, b). Both generally agree on the locations of larger σ , primarily near the jet streams at 30° S and 30° N. There are examples where the CTM overestimates (e.g., north Pacific Ocean in June) and underestimates (e.g., central Asia in June). These hot spots of large σ are generally caused by a single, very large folding event that the CTM apparently overestimates. The large σ observed at high latitudes cannot be explained by the model.

To analyze how well we predict the synoptic variability of TCO, we note that if the residuals, $CTM' = CTM - \overline{CTM}$ and $OMI' = OMI - \overline{OMI}$, are uncorrelated, then the variance of the error less the means, $(CTM' - OMI')^2$, would be $\sigma_{CTM}^2 + \sigma_{OMI}^2$. Thus, our index of matching the observed synoptic variance,

$$SV = 1 - \frac{\overline{(CTM' - OMI')^2}}{\sigma_{CTM}^2 + \sigma_{OMI}^2} \quad (1)$$

is a measure of the fraction of variance that is accurately simulated. The average and σ are weighted by the pixel area and observation frequency as above. By definition, SV ranges from negative (when CTM' and OMI' are anti-correlated) to +1 (when CTM' and OMI' are identical). The global mean SV is 0.39 for June and 0.37 for December. Figures 5e and 6e show the geographic patterns of CTM monthly mean TCO, where those locations with SV greater than 0.70 are marked by black dots. Note that in areas where the CTM matches the high frequency variability of OMI, the σ is also large,

OMI tropopause folds

Q. Tang and M. J. Prather

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



particularly along the subtropical jet streams near 30° S and 30° N (Figs. 5a, b and 6a, b).

Figures 5f and 6f present the cumulative distribution function (CDF) of the index SV weighted by area (i.e., the percentage of area whose SV is greater than or equal to the value on the X-axis) for the globe (blue line), tropics (25° S–25° N, red line), NH mid-latitudes (25° N–50° N, green line), and SH mid-latitudes (50° S–25° S, black line). Independent of seasons, the SV is best in SH mid-latitudes, moderate in NH mid-latitudes, and worst in the tropics.

4 TF anomalies vs. STE ozone flux

We expect regions with TF to be associated with STE O₃ flux as the folds are sheared and mixed with tropospheric air. The TF frequency is calculated from the 2-h global O₃ fields from the CTM (Fig. 7). Two high TF frequency bands are located near the subtropical jet stream in each hemisphere. The high frequency regions over equatorial Africa and South America are actually aliased from biomass burning plumes, as our TF criteria detect biomass burning plumes rising above 5 km as TF events. The STE O₃ flux is diagnosed by following the dilution of stratospheric O₃ (Hsu et al., 2005; Hsu and Prather, 2009), and, here, we define a chemical tropopause of 120 ppb of O₃. Since the 1° × 1° 60-layer meteorology field is not available, the STE flux is calculated using meteorology at T42 horizontal resolution with 60 vertical layers. This 60-layer field is basically the same EC meteorology as the 1° × 1° 40-layer, but has much better stratospheric circulation and STE O₃ flux. The STE O₃ flux is larger in NH than in SH, and in the summer of each hemisphere. The TF frequency and STE O₃ flux are better coincident in SH than in NH, probably due to the more zonal structure in the SH, as large scale planetary waves may drive the location of final STE mixing away from the jet in the NH. From June through August, large NH STE flux occurs over almost all of the Asian and North American continents and is not associated with TF. Over Asia the large STE region expands beyond 60° N, while high TF frequency area

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



only reaches 40° N. This STE O₃ flux is associated with deep convection. Convection over summer continents extends into the lower stratosphere, dragging O₃-rich air into the troposphere. Over the summer, approximately 5% of continental convection in the CTM reaches O₃ levels above 120 ppb. Additionally, the summer monsoon might contribute to this large STE flux, but the latitudinal extent is too large to be attributed to the monsoon alone.

5 Conclusions

Comparing the CTM profiles with WOUDC ozone sondes reveals that the model matches sonde measurements and is capable of locating and resolving tropopause fold events. In the CTM, high ozone anomalies in tropospheric column are correlated with TF events and occur most frequently near the subtropical jet streams, which is consistent with previous studies (Baray et al., 2000; Traub and Lelieveld, 2003). Over the whole year, the TF's frequency is 12% in the NH and 9% in the SH, with 13% and 11% of the area in NH and SH covered by TFs, consistent with the inter-hemispheric differences in STE O₃ flux of 300 Tg and 220 Tg, respectively.

The real-time tropospheric and total ozone columns measured by OMI are simulated by the UCI CTM for year 2005. The modeled ozone columns show very good agreement with coincident high frequency OMI observations, both in terms of the geographical patterns and variability. Model results are generally better in extra-tropics than in tropics, in part because of the problems with the CTM's tropical stratospheric meteorology, in part because OMI is not sensitive to O₃ from biomass burning. Except for biomass burning regions in Africa and South America and certain regions over ocean, the CTM monthly TCO means fall with ±5 DU range of OMI values. Further analysis shows that the high frequency variations in TCO observed by OMI are very well matched outside the tropics by the UCI CTM using the high resolution ECMWF pieced-forcast fields.

Having demonstrated the link between OMI anomalies in the tropospheric col-

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



umn ozone (TCO) and tropopause folds, we seek to extend it to the stratosphere-troposphere exchange (STE) ozone flux. The STE flux in the vicinity of the sub-tropical jets can possibly be measured with TCO anomalies, but the large regions over the summer continents in the NH occur through deep convection and are less apparent in the OMI ozone columns.

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OMI tropopause folds

Q. Tang and M. J. Prather

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



OMI tropopause folds

Q. Tang and M. J. Prather

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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OMI tropopause folds

Q. Tang and M. J. Prather

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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OMI tropopause folds

Q. Tang and M. J. Prather

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



H. E., Schnabel, C., Siegmund, P., Tobler, L., Trickl, T., Wernli, H., Wirth, V., Zanis, P., and Zerefos, C.: Stratosphere-troposphere exchange: A review, and what we have learned from STACCATO, *J. Geophys. Res.*, 108, 8516, doi:10.1029/2002JD002490, 2003. 14877

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OMI tropopause folds

Q. Tang and M. J. Prather

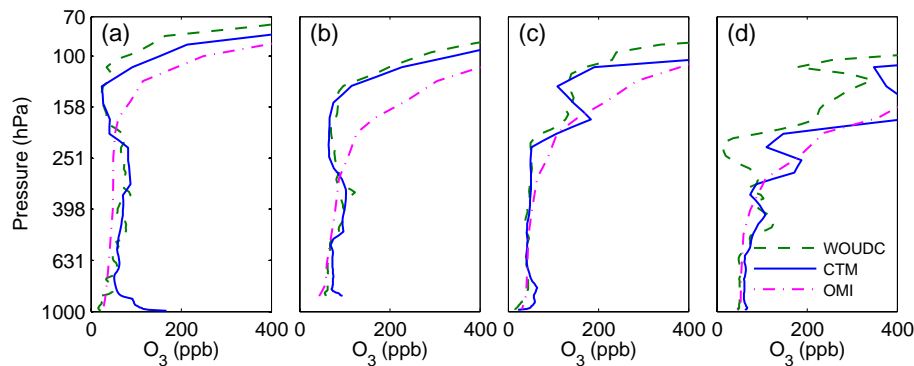


Fig. 1. O_3 profiles (unit: ppb) from WOUDC, CTM and OMI as a function of pressure (unit: hPa). **(a)** Hong Kong (22.31° N, 114.17° E, STN 344), China, 7 September 2005. **(b)** Ankara (39.97° N, 32.86° E, STN 348), Turkey, 17 August 2005. **(c)** Huntsville (34.72° N, 86.64° W, STN 418), USA, 3 December 2005. **(d)** Huntsville for 5 March 2005.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


OMI tropopause folds

Q. Tang and M. J. Prather

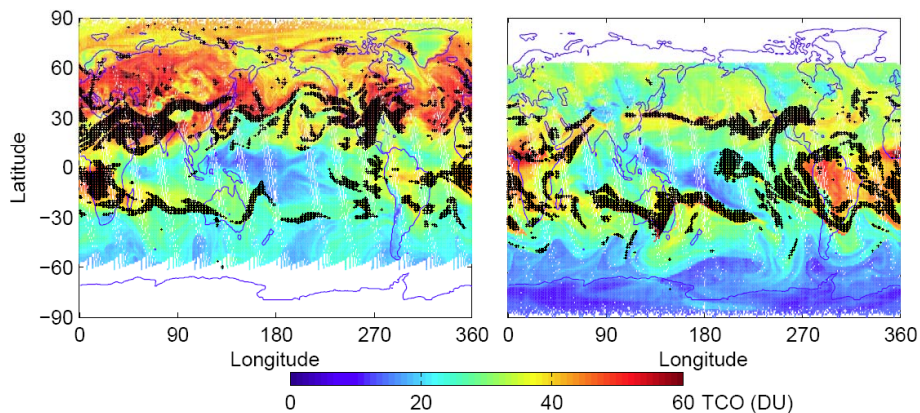


Fig. 2. CTM TCO (color, unit: DU) correlated with TF events (black “+”) for 25-h periods beginning 00:00:00 UTC on 10 June (left) and 3 December (right) 2005.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

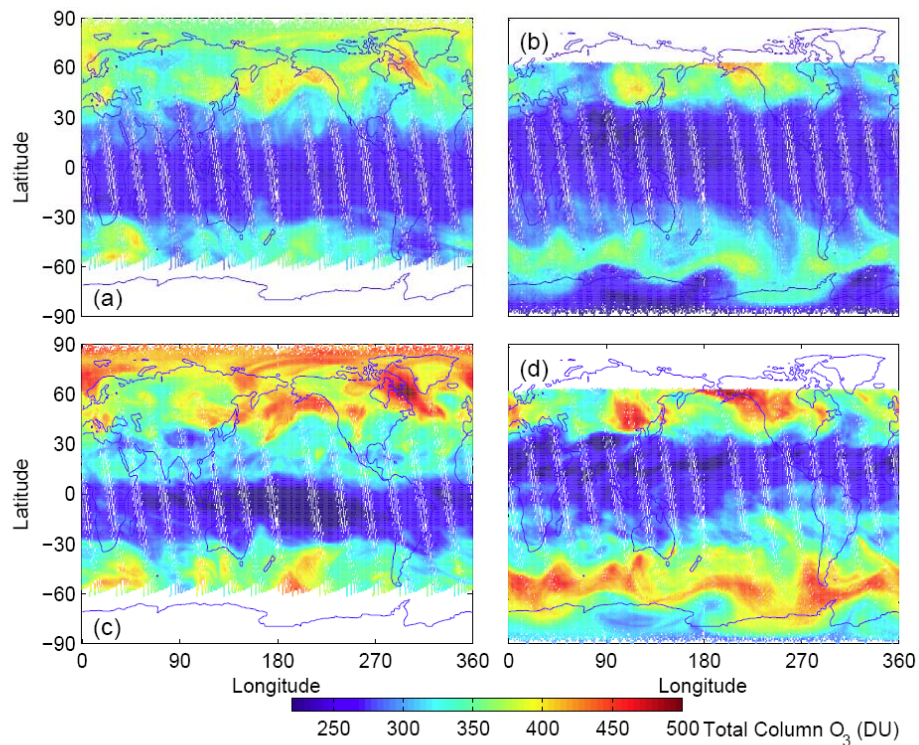


Fig. 3. Swath-by-swath comparison of OMI (top) and CTM (bottom) in total O_3 (**a** vs. **c** for 10 June and **b** vs. **d** for 3 December, unit: DU) in 2005.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

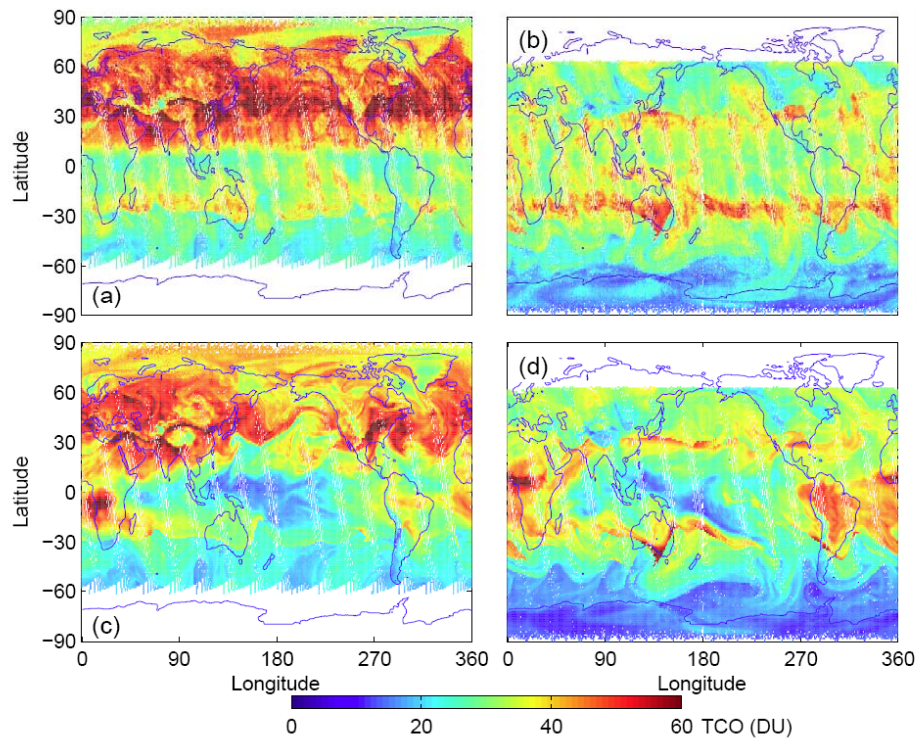


Fig. 4. Swath-by-swath comparison of OMI (top) and CTM (bottom) in TCO (**a** vs. **c** for 10 June and **b** vs. **d** for 3 December, unit: DU) in 2005.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

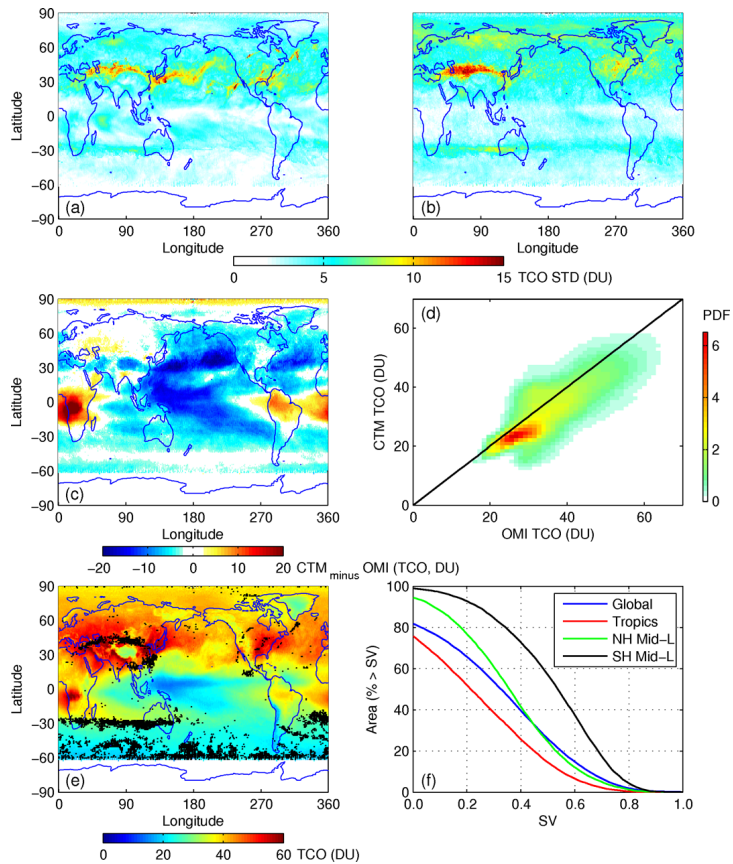


Fig. 5. For June 2005, **(a)** and **(b)** monthly σ of TCO (unit: DU) for CTM and OMI; **(c)** monthly mean bias (CTM–OMI) (unit: DU); **(d)** 2-D PDF of TCO (unit: 10^{-3} DU^{-2}); **(e)** monthly mean CTM TCO (unit: DU) with the locations of $SV \geq 0.70$ marked by black dots; **(f)** CDF of SV.

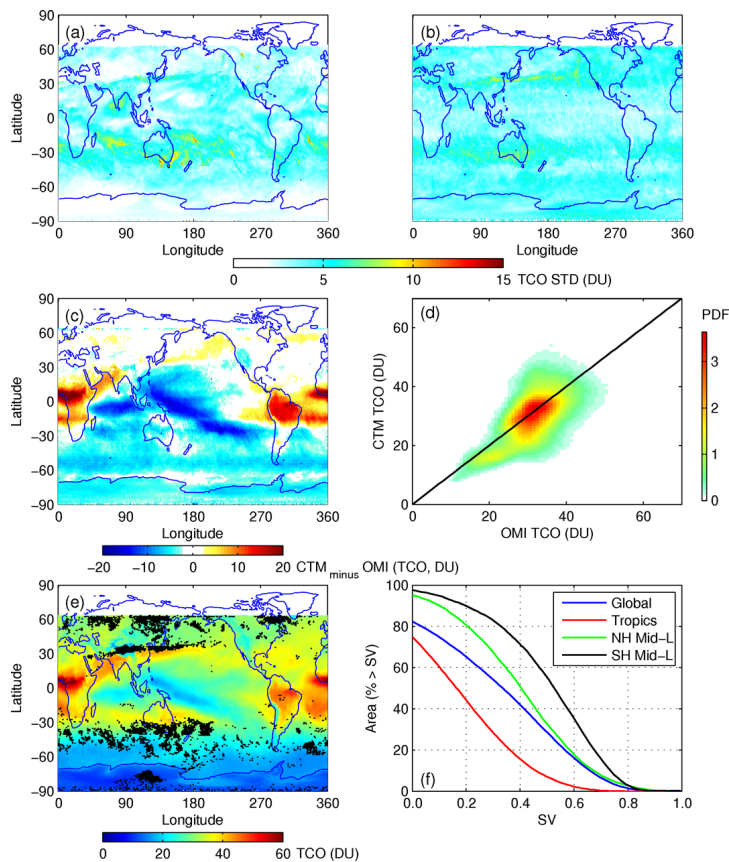


Fig. 6. For December 2005, same as Fig. 5.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



OMI tropopause folds

Q. Tang and M. J. Prather

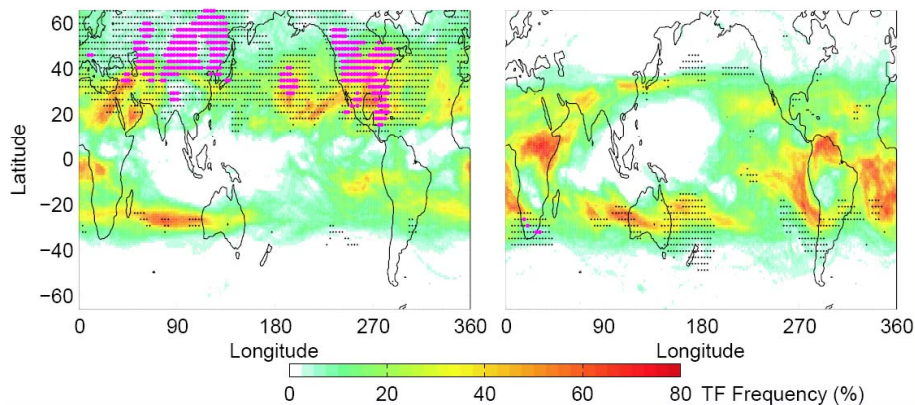


Fig. 7. TF frequency (color, unit: %) for June (left) and December (right) 2005 with STE O_3 fluxes (unit: $\text{g m}^{-2} \text{yr}^{-1}$) shown by black dots ($2 \leq \text{STE} < 4$) and magenta dots ($\text{STE} \geq 4$).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)