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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Correlating tropospheric column ozone with tropopause folds: the Aura-OMI satellite data" *by* Q. Tang and M. J. Prather

Q. Tang and M. J. Prather

tangq@uci.edu

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We thank Reviewer #1 for the insightful comments and for pointing out a potential bias in our results. We have redone the entire analysis to address the problems of a shifting tropopause (see detailed discussion below). We respond to the three major comments below and see no problem with revising the paper to address the minor details.

1a. What is the effect of applying a tropopause from a relatively high resolution data set to the OMI ozone profiles with low resolution in the troposphere?

Given the coarser resolution of the OMI profiles, the application of an observed tropopause height (or modeled with EC fields in our case) to interpolate that profile and thus determine tropospheric column ozone (TCO) will of course have larger errors. It is not obvious whether these errors will be biased high or low. How to balance the OMI profile such that the right amount of O_3 is in the troposphere involves their retrieval algorithms, including a priori profiles, and is beyond the scope of this work. The best we can do in this analysis is to take the OMI profiles and interpolate linearly.

If there were a tropopause fold (TF), we assume that OMI sees enhanced zone somewhere in the troposphere, and that this enhanced ozone is exhibited as a perturbation in the OMI profiles. Given the coarse profiles, it is possible that enhancements in the upper troposphere near the tropopause might spill over into the stratosphere (and not be counted as TCO). Likewise, anomalies restricted to the lower stratosphere may spill over into TCO. In our high-resolution model, we separate these two situations and find no obvious correlation between them. In the OMI data we also find (as noted in the ACPD paper) that regions of high TCO variability do not significantly overlap with those of high variability in the total column, thus implying little spillover effect.

In terms of bias in monthly mean patterns, we have identified several, with the most obvious cause being OMI's insensitivity to lower tropospheric ozone. It is possible that OMI TCO biases high in the steep tropopause region or other places. But coarse resolution profiles mapped with high resolution tropopause could be biased in either direction. We see areas that OMI TCO is more or less than that of the model (Fig. 5c and 6c), but no global systematic bias in OMI TCO. This is why we feel that most of the TCO variability seen by OMI is due to TFs in the upper troposphere. The high biases shown in Fig. 5d correspond to tropical oceans, SH mid-latitudes, and Greenland. To understand the causes of this difference between satellite data and model requires detailed knowledge of OMI retrieval scheme, and would be a major undertaking.

1b. Doesn't this lead to an artificial ozone enhancement for the satellite data in regions of steep tropopauses or even in the whole data set (Figs 5d and 6d)?

This question gets to the core issue of this paper: Are the OMI TCO anomalies truly stratospheric intrusions? Is the large positive correlation between OMI TCO anomalies and the CTM resolved TFs real? Or is it an artifact of shifting tropopause heights? Indeed, if we were to impose large variability in the tropopause height in a region where O_3 is not changing, then we would generate large, correlated variations in TCO in both OMI and CTM. (This mistaken analysis would be independent of the OMI resolution.)

Clearly the regions of steep tropopause, near the subtropical jet, are vulnerable to such errors. About the jet we often find a double tropopause, separate by more than 1 km. The air between is clearly stratospheric with high ozone abundances. Thus we find that variations in tropopause height here generate apparent TCO anomalies. In our model such positive anomalies correspond to an increase in “tropospheric" air mass, but having O_3 abundances of more than 200 ppb. Thus we need to restrict our comparison of TCO anomalies to where the tropopause height is stable. We performed a statistical analysis of tropopause heights to exclude these fluctuations and limit our analysis to regions where a positive TCO anomaly occurs without significant change in tropopause height — this should be a clear indication of classic stratospheric intrusions.

Our new work:

Since TFs are associated with large TCO variations on synoptic scale, we focus on areas with large standard deviation ($\sigma_{TCO} >$ 5 DU) in the CTM TCO (Fig. A1 for January, Fig. A2 for June, Fig. A3 for September 2005) and then calculate the 2-D probability distribution functions (PDF) of TCO vs. tropopause height for the corresponding months (Fig. B1, 2, 3). The threshold of 5 DU selects the upper 16% locations. The variability of CTM TCO is generally independent of tropopause height from February to August 2005. In January and through September to December 2005, some TCO values increase with tropopause height (marked by the red boxes in Fig. B3). September is the worst month. The points within the red box in Fig. B3 generally reflect the change in tropopasue height and are not correlated with TF events. Therefore, these data should be excluded from the analysis of TCO variations. Given the location of these points and the magnitudes of TCO variation, we diagnose that this artificial TCO variation is caused by the displacement between tropical tropopause and extra-tropical tropopause about the sub-tropical jet.

The PDF of the global tropopause pressure (TPP) is bimodal as shown by Fig. C. We thus apply a tropopause filter, dividing the global TCO data set into two subsets: one with tropical tropopause and the other with extra-tropical tropopause. As a result, the tropopause height is restricted to vary within one mode. For September 2005, the PDF of tropopause height is fit by two normal distribution functions (mean$_{TP}$ = 95 hPa, σ_{TP} = 10 hPa for tropics (TP) and mean$_{EX}$ = 240 hPa, σ_{EX} = 50 hPa for extra-tropics (EX)). The data with TPP less than 105 hPa are collected into the tropical subset, while the ones greater than 190 hPa are classified into extra-tropical subset, and thus the data with TPP of 105–190 hPa are dropped. The filter excludes 17.4% of the data, corresponding to 22.5% area coverage. The σ_{TCO} is then calculated separately for both subsets (shown in Fig. D1, E1 for CTM, Fig. D2, E2 for OMI). The locations of large σ_{TCO} (e.g., over eastern Pacific, India Ocean, North America) are generally consistent between the CTM and OMI, but OMI variations are smaller in tropics and larger in high-latitudes.

The changes in CTM and OMI TCO are uncorrelated with tropopause shifting as shown by the wide spread distributions in Fig. E1, F1 for CTM and Fig. E2, F2 for OMI. OMI can not sense the lower troposphere, and because of the a priori profiles, OMI TCO values are all above ~25 DU. In Fig. E and F, the PDF patterns of OMI are quite similar to those of CTM. If the artificial bias, as suggested by the reviewer, has great influence on OMI TCO, the PDF patterns of CTM and OMI would show much greater differences. Therefore, we conclude that the bias in OMI TCO due to its coarse tropospheric vertical resolution is not systematic and its impacts on OMI TCO variability and SV are relatively small compared to TF events.

In the revised paper, however, we must redo our ACPD analysis of SV, since we must exclude the transient regions. We will note that OMI's coarse tropospheric resolution may be one of the reasons for the difference between OMI and CTM. But the derived

OMI TCO data set still captures the variations in the troposphere. The contents about tropopause filter will be added and thus Fig. 5a,b,e,f and 6a,b,e,f will be revised.

2. The authors are aware of a biomass burning effect on ozone as stated in the manuscript, but they do not try to disentangle it. Why did the authors not exclude biomass burning ozone by simply using a second tracer (e.g. CO) which should be available in the CTM? Those events could be marked differently in the plots and accounted for in the analyses or at least coincidence to folds could be indicated. Since the tropics and subtropics are affected by both biomass burning and folds, the authors could differentiate and calculate e.g the percentage of high TOC events, which are also affected by biomass burning (or exclude them).

Sources of TCO variations from biomass burning and industrial pollution could indeed be tracked with tracers like CO, C_2H_6 or other tagged tracers. There are interesting studies using this approach but beyond the scope of this paper. Here we focus on the areas with large TCO variations on synoptic scale, since tropopause folds occur on this scale. Biomass burning and pollution events enhance tropospheric ozone, but do not lead to large TCO variations on synoptic scale, at least in the CTM, as shown by the standard deviation of monthly TCO series (Fig. 5a, b and 6a, b). The analysis on the relative contributions of biomass burning and tropopause folds to high TCO events would be in a separate paper.

3a. It is surprising to see very low ozone fluxes in the winter hemispheres (Fig.7). Also the patterns of fold occurrence do partly not coincide with the fluxes (Fig.7) and the authors discuss a few reasons, including summer convection in the northern hemisphere (Fig 7).

The low ozone fluxes during the winter are consistent with our previous publications using the EC met-fields (*Hsu et al.*, 2005, Fig. 6) and (*Hsu and Prather*, 2009, Fig. 7 and 8). In these studies, the method of diagnosing ozone flux is entirely self-consistent and is based on the flux across ozone isentropes. The locations of ozone fluxes identify where stratospheric ozone mixes down to tropospheric abundances. TF frequency indicates the possibility of stratospheric air entering the troposphere and the stratosphere troposphere exchange (STE) flux is diagnosed later as and if the fold is mixed and dispersed. Therefore, the TF frequency and ozone fluxes do not always coincide.

3b. Fold occurrence for deep folds in the extratropics should be strongest in winter / spring at least according to the Lagrangian studies (e.g. Sprenger, Croci-Maspoli, Wernli, 2003, SCW03 in the following). Note, that the TF frequency largely matches the summer shallow exchange case in SCW03 (their Figure 3b), but not for winter. Note also that the Lagrangian mass fluxes in the summer NH (SCW03, Fig.7) are not too different from the June patterns of ozone flux in Tang and Prather, whereas the winter fluxes and patterns are totally different (of course mass and ozone fluxes are not the same, but should be related to some extent).

The differences in winter TF frequency noted here are primarily due to the shift in scales used in Fig. 3 of SCW03 (*Sprenger et al.*, 2003, to be added to paper). In this paper Fig. 7 uses the same scale for all TF and hence the low-frequency, deep folds are not shown. In SCW03, the maxima of medium and deep TF frequency are 5% and 1.2%, respectively. If we change the color scale of Fig. 7 to 0–10%, these low frequency regions emerge (e.g., North America). Note that these are infrequent events and hence less important for the STE flux. In Fig. 7, only ozone fluxes greater than 2 $\mathrm{g\,m^{-2}\,yr^{-1}}$ are marked by dots. Large mid-latitude areas over Southern Hemisphere and North Pacific and Atlantic have ozone fluxes around 1 $\mathrm{g\,m^{-2}yr^{-1}}$. A serious study comparing TFs diagnosed from ozone vs. PV using the same met-fields would be very interesting as a collaborative effort among the different groups. We could diagnose differences and causes.

3c. Is it possible, that the fold occurrence frequency is biased by the method of fold detection and/or the choice of the ozone thresholds? Eventually the 5km criterion is too high as lower cut-off since it systematically removes deep folds,

which are of potential importance. Are there any sensitivity studies possible?

The fold frequency is a relative number used here to compare different regions. Of course the absolute frequency depends on resolution, thresholds, sampling interval, and etc, e.g., sonde data give much high TF frequency. The TF frequency will change if we choose different thresholds. The main reason for the 5 km criterion is to exclude the high ozone values in pollution plume and biomass burning (but these still contribute to TCO variability).

3d. Note also the large differences of fold frequency occurrence (Fig.7) around south America compared to SCW03, which might be from biomass burning highlighting the importance to account for the latter.

This is possible. But again the variability in lower troposphere is less frequent. For the same reason as the answer above, it does not affect the main conclusion of this paper.

3e. Further, if convection is the reason for the continental summer ozone fluxes, could the authors give reference to measurements which clearly show ozone enhancements from convection due to associated downdraft (due to mass conservation) as suggested at the end of section 4? It is correct that upward mass flux must be balanced by downdraft somewhere, but what is the experimental evidence for locally associated tropospheric ozone enhancements from the stratosphere?

Measurements by *Baray et al.* (1999) and *De Bellevue et al.* (2006) directly link downdraft induced by deep convection with tropospheric ozone enhancement. They provide observations of tropospheric ozone enhancement in the vicinity of convection on mesoscale. This work supports our proposal but the major work here will be in our subsequent paper. The citation to their papers will be added in section 4.

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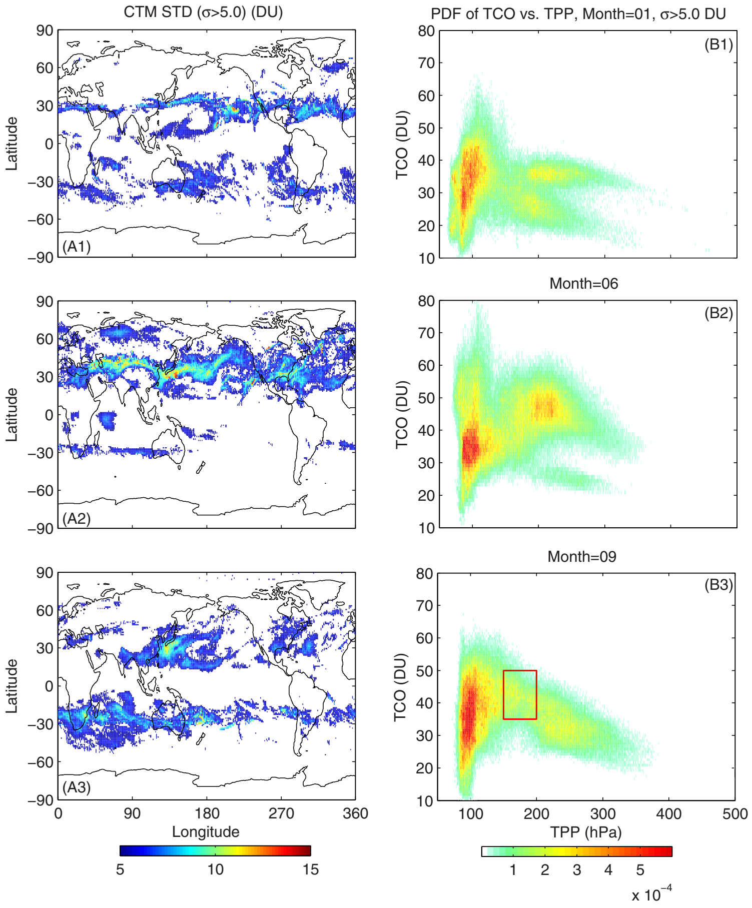
CTM STD (σ>5.0) (DU)
PDF of TCO vs. TPP, Month=01, σ>5.0 DU
Month=06
Month=09
(A1)
(A2)
(A3)
(B1)
(B2)
(B3)
Latitude
90
60
30
0
−30
−60
−90
Longitude
0
90
180
270
360
TCO (DU)
80
70
60
50
40
30
20
10
TPP (hPa)
100
200
300
400
500
5
10
15
1
2
3
4
5
x 10^−4

Fig. 1.

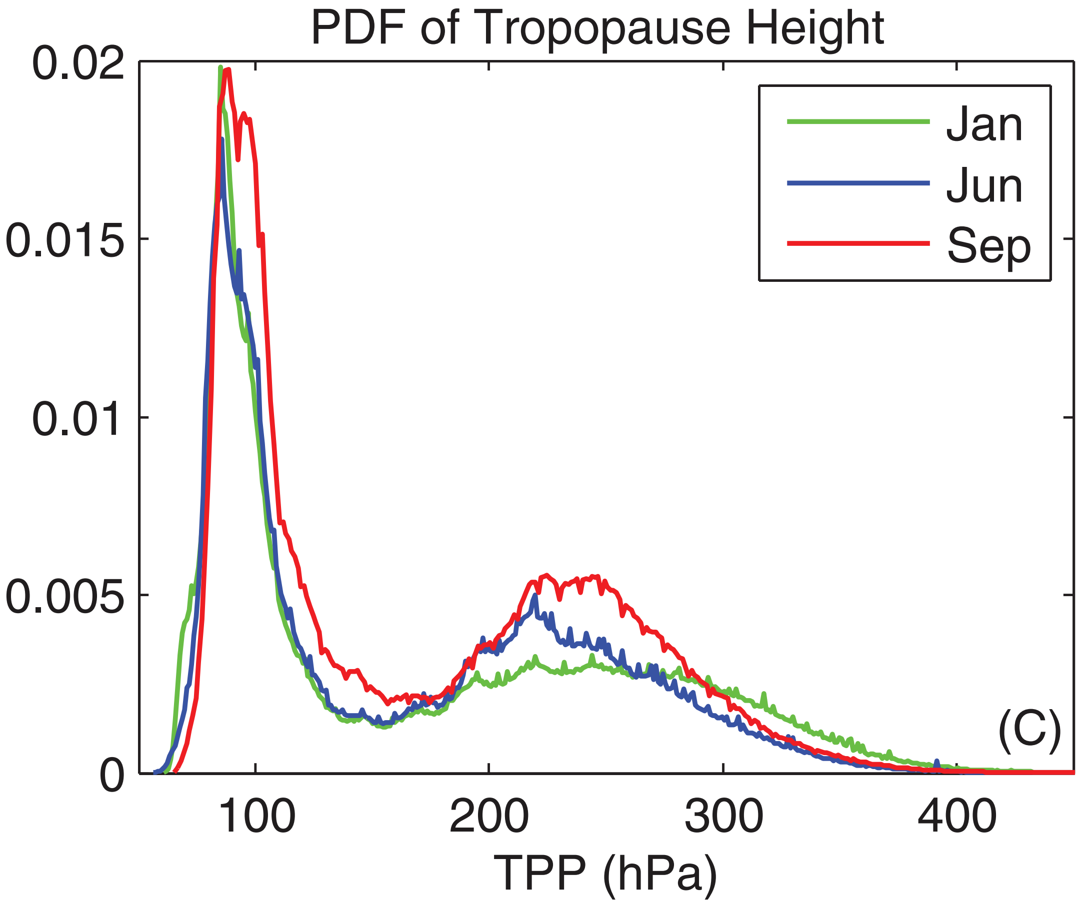
PDF of Tropopause Height
0.02
0.015
0.01
0.005
0
100
200
300
400
TPP (hPa)
Jan
Jun
Sep
(C)

Fig. 2.

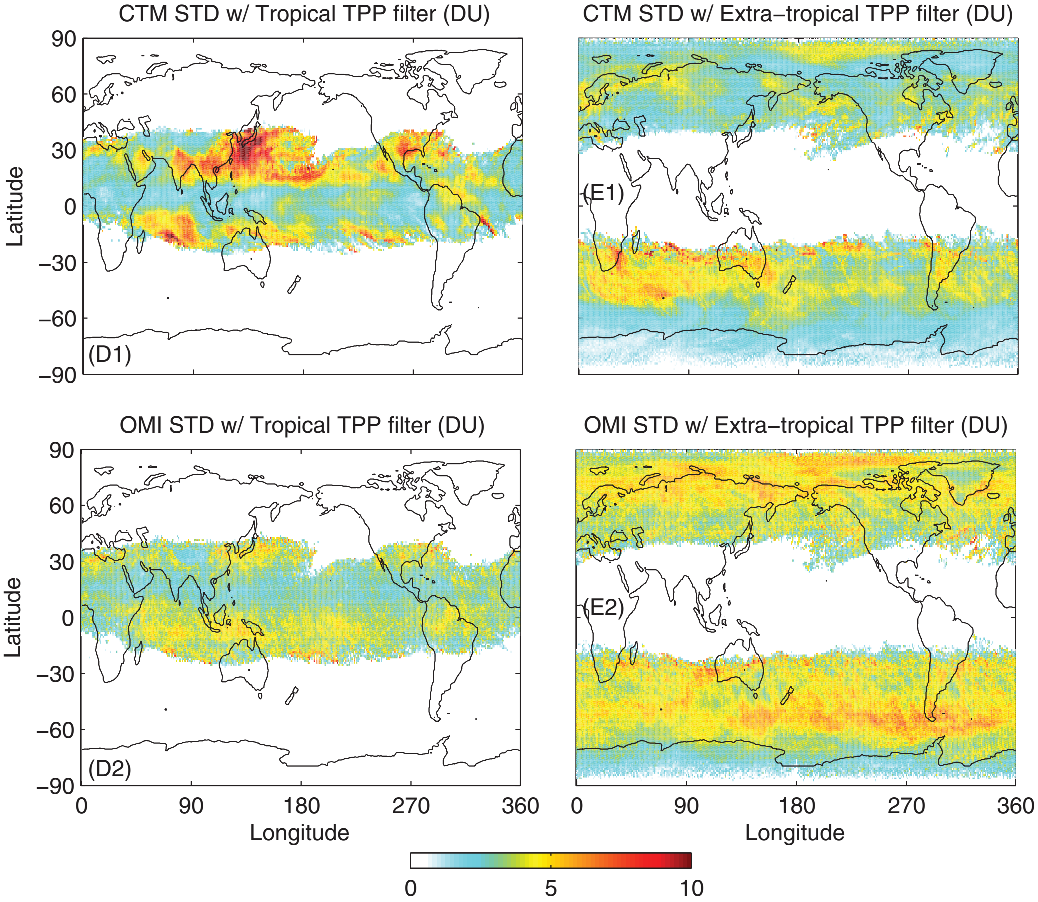

Fig. 3.

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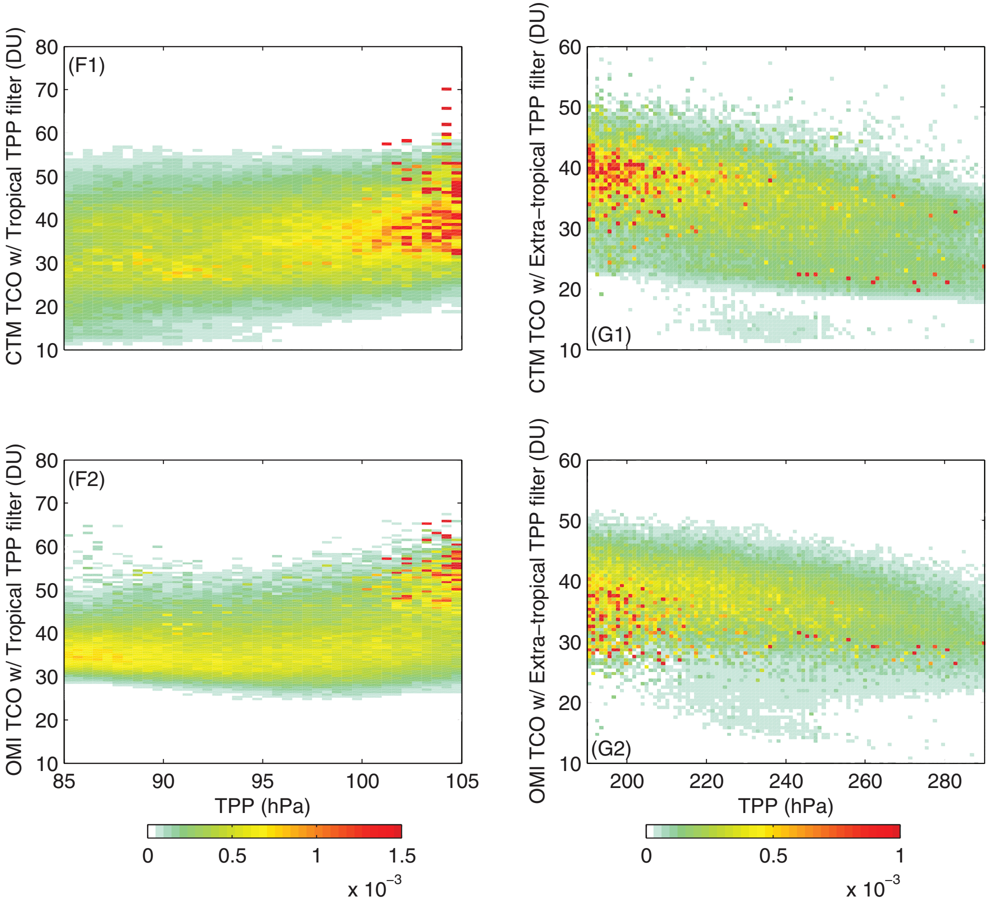

Fig. 4.

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