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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Climatology of pure Tropospheric profiles and column contents of ozone and carbon monoxide using MOZAIC in the mid-northern latitudes (24 N to 50 N) from 1994 to 2009" *by* R. M. Zbinden et al.

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Reply to M. Osman (Referee)

We are very grateful to the Referee M. Osman for the time and the relevant comments he made on our paper. We have followed your suggestions which help to improve the quality of the manuscript.

Comments:

- Since z_{Ld} is the lowest height at which the MP and PTP deeply diverge and it illustrates the penetration depth of the tropopause height as well as stratospheric air contamination, the partial columns in the height between z_{Ld} and z_{DT} (i.e., UTC(X, t)) sounds more like the tropopause layer column.

Reply : *z_{Ld} highlights the impact of the penetration depth of the tropopause consistently between O_3 and CO. In Thouret et al. (2006), the tropopause layer is defined between z_{DT} - 15 hPa and z_{DT} + 15 hPa, with z_{DT} fixed at 2 pvu. As shown on Figure 10, and to compare with the tropopause layer defined by Thouret et al. (2006), the thickness of our UTC for Germany is 170-115 hPa, z_{Ld} varies from 6.6 to 8.1 km (∼435 to 351 hPa) and z_{DT} from 10.0 km to 10.8 km (∼264 to 234 hPa). For Japan, the thickness of our UTC is 194-45 hPa, z_{Ld} varies from 6 to 11.9 km (∼470 to 197 hPa) and z_{DT} from 9.3 to 13.6 km. Therefore and to conclude, contrarily to the tropopause layer, our UTC is much thicker, with a larger thickness in winter than in summer, and excludes the stratospheric air above z_{DT}. Undoubtedly, the occurrence of low tropopause events within a month affects the z_{DT} position.*

Below, modifications as suggested

⇒ We have added page 14717, Line 22 before "Finally" : *In Thouret et al. (2006), the tropopause layer has been defined between z_{DT} - 15 hPa and z_{DT} + 15 hPa, with z_{DT} fixed at 2 pvu. Our UTC thickness, shown only with respect to altitude on Fig. 10, is ranging 115-170 hPa, 52-214 hPa and 45-194 hPa for Germany, USeast and Japan, respectively. Therefore, contrarily to the tropopause layer, our UTC is much thicker and excludes the stratospheric air above z_{DT}.*

- Caution should be taken in determining z_{Ld} with few data (for example, Figure 6: China in January, February, April, December as well as Uaemi in October, November)

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since lack of enough data cause a significant error in the calculation of the monthly-averaged z_{Ld} height.

Reply : *We perfectly agree with you. We would like to insist on that point and emphasize that the section 4.3 is strictly limited to the USeast, Germany and Japan partial columns. We have never used and tested z_{Ld} elsewhere. Thank you to give us the opportunity to insist on such restriction.*

Below, modifications as suggested

⇒ We have added Page 14717, Line 6 : *z_{Ld} and partial tropospheric columns using z_{Ld} have been evaluated strictly for those 3 most documented sites in order to present significant results, and not elsewhere.*

⇒ We have removed Page 14717, Line 12 : *over USlake*, written by mistake.

- The PTP was calculated if $z_{top} < z_s < z_{DT}$ using $\mathrm{Mfit}(X, z_{\Delta_f}, s)$, the best-fitted line from MOZAIC data using a linear regression on $\overline{\mathrm{TP}}(X, z_{\Delta_s}, s)$ i.e. from 5 to 11 km for O_3 and from 8 to 11 km for CO. What is the reason for using different height ranges for O_3 and CO? Is there any particular reason for not using MP from the surface up to (z_{top}, if $z_{top} < z_{DT}$, and z_{DT}, if $z_{top} > z_{DT}$) ?

Reply : *The preliminary climatology, $\overline{\mathrm{TP}}(X, z, s)$, is equivalent to an average of MP from the surface up to (z_{top}, if $z_{top} < z_{DT}$, and z_{DT}, if $z_{top} > z_{DT}$) on a seasonal basis and your suggestion should provide similar result, because the stratospheric air mass above tropopause on all individual profiles is discarded. Nevertheless, this latter labelling does not highlight the tropospheric major characteristic and could be somewhat confusing with an average on all MOZAIC profiles on a seasonal basis, $\overline{\mathrm{MP}}(X, z, s)$. The preliminary climatology, $\overline{\mathrm{TP}}(X, z, s)$, is aimed to further complete the unvisited tropospheric remainder of each profile whenever necessary (i.e. when $z_{DT} > z_{top}$) with contents strictly representative of tropospheric air (and not from an averaged of tropo-*

spheric and stratospheric air masses from above tropopause, in upper layers). Note the preliminary climatology $\overline{\mathrm{TP}}(X, z_{\Delta_s}, s)$ is calculated once over the time series, and has to be reprocessed when the time series are updated. The results are provided on a seasonal basis for all the sites to have a significant sampling, while, on a monthly basis, the study would have been limited to Europe, US (except Los Angeles) and Japan. The best fitted linear regression is estimated from $\overline{\mathrm{TP}}(X, z_{\Delta_s}, s)$ between 5 and 11 km for O_3 and 8 and 11 km for CO. These altitudes were fixed because, the O_3 amounts decrease almost linearly with altitude on $\overline{\mathrm{TP}}(\mathrm{O_3}, \mathrm{z}_{\Delta_s}, \mathrm{s})$ above the BL. Consequently, the regression linear fit should be determined between altitudes as far as possible from the polluted planetary boundary layer, but not too high to maintain significant sampling and, by the end, to be representative of the seasonal tropospheric amounts in upper layers. For CO, similarly, the limits were fixed at higher altitudes but within a narrow layer because CO does not decrease linearly with altitude, except above 7-8 km.

Below, modifications as suggested

$\Rightarrow$ We have added Page 14701 Line 9 : *These limits in altitude were chosen to be as far as possible from the polluted boundary layer, but not too high to maintain significant sampling and, by the end, to be representative of the seasonal tropospheric amounts in the upper tropospheric layers. For CO, similarly, the limits were fixed at higher altitudes but within a narrow layer because CO does not decrease linearly with altitude, except above 7-8 km.*

- In calculating the monthly-averaged $\mathrm{PTC}_m(X, t)$, the seasonal based z_{DT} is used instead of the monthly-averaged z_{DT}? Is it not possible to get the monthly-averaged z_{DT}? It would also be better if the author's reason out why they prefer to use seasonally averaged profiles but monthly-averaged columns and partial columns. Please include the reason in the introduction section to guide readers.

Reply 1: *Regarding your question on the seasonal based z_{DT} : In the paper, the z_{DT} is*

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fixed for each individual MOZAIC profile and the $\mathrm{PTC}(X,t)$ *is derived for each individual profile from the surface to its specific* z_{DT} *(Figure 1). After, the 176 averages by month are calculated from* $\mathrm{PTC}(X,t)$ *(not shown) and finally, from the last result, the seasonal cycle with* $\mathrm{PTC}(X,t)$ *is obtained by the end. The* $\overline{z_{DT}}$ *averaged by month or season is not introduced in the* $\mathrm{PTC}(X,t)$ *or* $\mathrm{PTP}(X,z,t)$ *calculations. It is only provided on the Figures as a guide line.* ***The delivered climatology results from profiles and column tropospheric contents on an individual*** z_{DT} ***basis.***

Reply 2: *Regarding your question on the possibility to obtain monthly-averaged* z_{DT} *: The Figure 1 shows that individual* z_{DT} *is the cornerstone of this tropospheric climatology. The monthly-averaged* $\overline{z_{DT}}$ *is only a guide line and given to illustrate the Figures 2, 4, 5, 10 and 11 while the seasonally-averaged* $\overline{z_{DT}}$ *is given to illustrate the Figures 7, 8 and 9.*

Reply 3: *Regarding your question on the reason to prefer seasonally-averaged profiles and monthly-averaged columns and partial columns, there is no technical major reason to limit the study to seasonally-averaged profiles, except when the monthly sampling is rather poor. Providing the monthly-averaged profiles could be possible but the study should have been limited to Europe, US (excepted Los Angeles) and Japan. Seasonally-averaged profiles are more synthetic results allowing to illustrate clearly the* O_3 *seasonal dichotomy and associated anomalies in the mid-northern latitudes.*

Below, modifications as suggested

⇒ We have added Page 14701, before Line 20 : *From* PTC*(X,t), we calculated the monthly times series (not shown) and finally the monthly-averaged* $\mathrm{PTC}_m(X,t)$ *as shown in section 4.1. The* $\overline{z_{DT}}$ *by month or season is not introduced in any of the* $\mathrm{PTC}_m(X,t)$ *or* $\mathrm{PTP}_s(X,z,t)$ *calculations; it is only provided on the Figures as a guide line. Consequently, the delivered climatology results from profile and column tropospheric contents on an individual* z_{DT} *basis. Providing the monthly-averaged profiles could be possible but the study should have been limited to Europe, US (excepted Los*

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Angeles) and Japan, should obviously be less synthetic and should less highlight the seasonal dichotomy and seasonal anomalies discussed later in section 4.2.

- Please provide equation # for all equations in the paper.

Reply : *This has been done.*

Below, modifications as suggested

$\Rightarrow$ We have replaced Page 14701, Line 12-19 by :

$$\mathrm{PTP}(X,z,t) = \mathrm{MP}(X,z,t) = \mathrm{TP}(X,z,t) \quad (1)$$
$$\mathrm{PTP}(X,z,t) = \mathrm{TP}(X,z^\star,t) + \overline{\mathrm{TP}}_s(X,z_{\Delta_s},t) \quad (2)$$
$$\mathrm{PTP}(X,z,t) = \mathrm{TP}(X,z^\star,t) + \overline{\mathrm{TP}}_s(X,z_{\Delta_s},t) + \mathrm{Mfit}_s(X,z_{\Delta_f},t) \quad (3)$$

We have used the equation :

(1) when $z_{DT} < z_{top}$.

(2) when $z_{top} < z_{DT} < z_s$, with $z^\star = [z_0, z_{top}]$ and $z_{\Delta_s} = [z_{top}, z_{DT}]$.

(3) when $z_{top} < z_s < z_{DT}$, with $z^\star = [z_0, z_{top}]$, $z_{\Delta_s} = [z_{top}, z_s]$ and $z_{\Delta_f} = [z_s, z_{DT}]$.

$\Rightarrow$ We have replaced Page 14702, Line 9 by :

$$\mathrm{PTC}(X,t) = \sum_{z=z_0}^{z_{DT}} \mathrm{PTP}(X,z,t) \quad (4)$$

$\Rightarrow$ We have replaced Page 14703, Line 24-27 by :

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$$\mathrm{MWPTP}(\mathrm{O_3}, z, t) = \mathrm{TP}(\mathrm{O_3}, z, t) = \mathrm{PTP}(\mathrm{O_3}, z, t) \quad (5)$$

$$\mathrm{MWPTP}(\mathrm{O_3}, z, t) = \mathrm{TP}(\mathrm{O_3}, z^\star, t) + \overline{\mathrm{WPTP}}_{m'}(\mathrm{O_3}, z_\Delta, t) \quad (6)$$

The equation equa5 is used when $z_{DT} < z_{top}$ while equa6 is used when $z_{DT} > z_{top}$.

- It would be great if the author could mention in the paper the percentage of the MP data with 1. $z_{DT} < z_{top}$; 2. $z_{top} < z_{DT} < z_s$; 3. $z_{top} < z_s < z_{DT}$

Reply : *Percentage has been added. Thank you for this very interesting suggestion.*

Below, modifications as suggested

$\Rightarrow$ Add Page 14701, before Line 20: *The Table S1 provides the percentage of profiles corresponding to the 3 cases encountered at the 11 sites.*

- Page 14705 Line 25 and Page14706 Line 1: "in USsouth and Uaemi between 1 and 4 km all over the months due to intense domestic traffic." This sentence needs clarification. I took me a while to understand what it meant. Did you mean that most of the data were collected using small aircrafts that flew well below the tropopause? Please rewrite this sentence because it is really vague. Can you please mention what percent of data of other sites were collected using such domestic flights and international flights.

Reply : *Thank you to allow us to clarify. The MOZAIC equipment is only on board of AIRBUS A340. Nevertheless, when the flight connects two close airports, like Abu Dhabi and Dubai ($\sim$ 150 km distant) or Dallas and Houston ($\sim$ 400 km distant), the AIRBUS A340 will not reach the cruise level and the flight is limited to a short ascent and short descent. These flights are considered as 'domestic traffic' by opposition to*

Table S1. Percentage of MP profiles for the 3 cases where $z_{DT} < z_{top}$, $z_{top} < z_{DT} < z_s$, $z_{top} < z_s < z_{DT}$.

	Percentage of MP with		
Sites	$z_{DT} < z_{top}$	$z_{top} < z_{DT} < z_s$	$z_{top} < z_s < z_{DT}$
Los Angeles	24.3	31.3	44.3
USeast	41.9	31.9	26.2
USlake	43.5	34.3	22.1
USsouth	10.5	26.6	62.8
Paris	46.0	46.0	8.0
Germany	49.5	44.4	6.1
Vienna	58.4	36.3	5.3
Eastmed	34.8	16.3	48.9
Uaemi	4.5	5.1	90.4
Beijing	48.2	36.0	15.8
Japan	31.4	29.3	39.2

international distant flights. At Los Angeles, Eastmed, Beijing and Vienna no domestic flights are included, they represent less than 1% for Germany, USeast, USlac, Japan and Paris but 29% of the MOZAIC traffic for USsouth and 39% for Uaemi. We could have discarded these flights but they are providing valuable observations on the highly variable BL, thus we did not choose this option. Moreover, we found they document areas where no ozonesondes or carbon monoxide measurements are existing (except in case of research field campaign).

Below, modifications as suggested

⇒ We have replaced Page 14705 Line 25 and Page 14706 Line 1 by : *. . . in USsouth and Uaemi between 1 and 4 km all over the months due to intense domestic traffic. Note, the domestic flights connect two close airports, like Abu Dhabi and Dubai (∼ 150 km distant) or Dallas and Houston (∼ 400 km distant). Consequently the cruise*

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level is never reached and the flight is limited to a short ascent and short descent. At Los Angeles, Eastmed, Beijing and Vienna no domestic flights are included, while they represent less than 1% for Germany, USeast, USlac, Japan and Paris and 29% of the MOZAIC traffic for USsouth and 39% for Uaemi. We did not discarded these profiles because they were documenting the highly variable BL and areas where no regular ozonesondes or carbon monoxide measurements are existing.

- Page 14708 Line 16-17: "Intense photochemical activity is detected there in spring" Who detected it? How detected it? Please cite reference

Reply : *The text has been fully changed.*

Below, modifications as suggested

⇒ Text Page 14708 Line 16-17 has been replaced by : *The difference between* $TC_m(O_3)$ *and* $PTC_m(O_3)$ *is less than 2 DU in spring, compared to 4 DU in summer where the tropospheric column height has a significant contribution. In May, the* $PTC_m(O_3)$ *is more than 5 DU higher than Germany and is even higher than Beijing. These findings suggest favourable photochemical conditions allowing this local* O_3 *production, as shown further in section 4.2.3 and Fig. 9.*

- Page 14710 Line 9-11: " Over all these sites, a sharp May-June CO depletion highlights the intense photochemical activity" How did you know? Please cite reference

Reply : *The text has been modified.*

Below, modifications as suggested

⇒ Text Page 14710 Line 9-11 has been replaced by : *Over all these sites, a sharp May-June CO depletion highlights the powerful OH cleansing efficiency regulated by NOx (Lamsal et al., 2010)*

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⇒ We added the reference : Lamsal L. N., Martin R. V., Donkelaar A. van, Celarier E. A., Bucsela E. J., Boersma K. F., Dirksen R., Luo C., and Wang Y., Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI satellite instrument: Insight into the seasonal variation of nitrogen oxides at northern midlatitudes, J. Geophys. Res., 115, D05302, doi:10.1029/2009JD013351, 2010.

- P14710L16-17: " The July bump over USeast results from the impact of North American boreal fires, during the summer of 2004 (Turquety et al., 2007)". Is the July bump ONLY as a result of the fire in 2004? There were other intensive wild fires during the period of this climatology study, for example in 1995 and 2006. I expected a July bump in the USlake as well but I did not see it. Why?

Reply : *In Canada, wild fires occur every year, but the one in 2004 has the greatest extent* [1]*, and this extent was so huge that Morris et al. (2006) reports on a MOZAIC flight with a CO anomaly down to Dallas on 18 July 2004 attributed to the Alaska and Yukon wildfires. We have added the Figure S2 to evaluate the 2004 CO anomaly in July over USeast and to show that USeast has been more on the CO plume pathway (or branch of pathway) than the USlake. This finding and difference appear in agreement with what MOPITT has captured* [2]*. As this point has been also highlighted by the Anonymous referee 1, more details are given there with the modifications*.

We have included in the text all your minor corrections and modified the Figure 4, 5 and 6. Thank you for your very constructive remarks.

Figure captions

[1]http://cwfis.cfs.nrcan.gc.ca/en_CA/nfdb/poly
[2]http://www.ucar.edu/news/releases/2005/wildfires.shtml

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Fig. S2 : Monthly mean tropospheric profiles of the CO July 2004 anomaly (i.e. the average of (2002, 2003, 2005, 2006) minus 2004) at USeast (red line) and USlake (dotted blue line) in mol/cm^2.

Fig. 4 : Validation and impact of Δ on the pure tropospheric columns based on USeast, Germany and Japan seasonal cycles by comparing: $\overline{\mathrm{TC}}(\mathrm{O}_3, m)$, exactly what MOZAIC has measured in the troposphere (dark blue line); $\overline{\mathrm{PTC}}(\mathrm{O}_3, m)$, the MOZAIC pure tropospheric ozone column (red line); $\overline{\mathrm{MWPTC}}(\mathrm{O}_3, m)$, the composite MOZAIC-WOUDC tropospheric ozone column (blue line). All columns are expressed in DU. Δ is the layer between z_{top} (dotted green line) and z_{DT} (solid green line), in km (right green vertical axis).

Fig. 5 : Cycles of $\mathrm{TC}_m(\mathrm{O}_3)$, in blue, and $\mathrm{PTC}_m(\mathrm{O}_3)$ box and whisker, in red, expressed in DU by referring to left vertical axis for USeast, USlake, USsouth, Los Angeles, Germany, Paris, Vienna, Japan, Beijing, Uaemi and Eastmed. z_{DT} is the thick green line and z_{top} the thin green line, both referring to the right vertical axis in km. Monthly sampling frequency of each site is provided above the X axis. Box uses the quartiles [Q25, Q50, Q75]. The end of box whiskers are the $\geq$Q25-1.5IQR or $\leq$Q75+1.5IQR.

Fig. 6 : Same as Fig. 5 but for CO, expressed in x10^{18}mol/cm^2. Note that only Beijing is plotted with a specific scale.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C8180/2013/acpd-13-C8180-2013-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 14695, 2013.

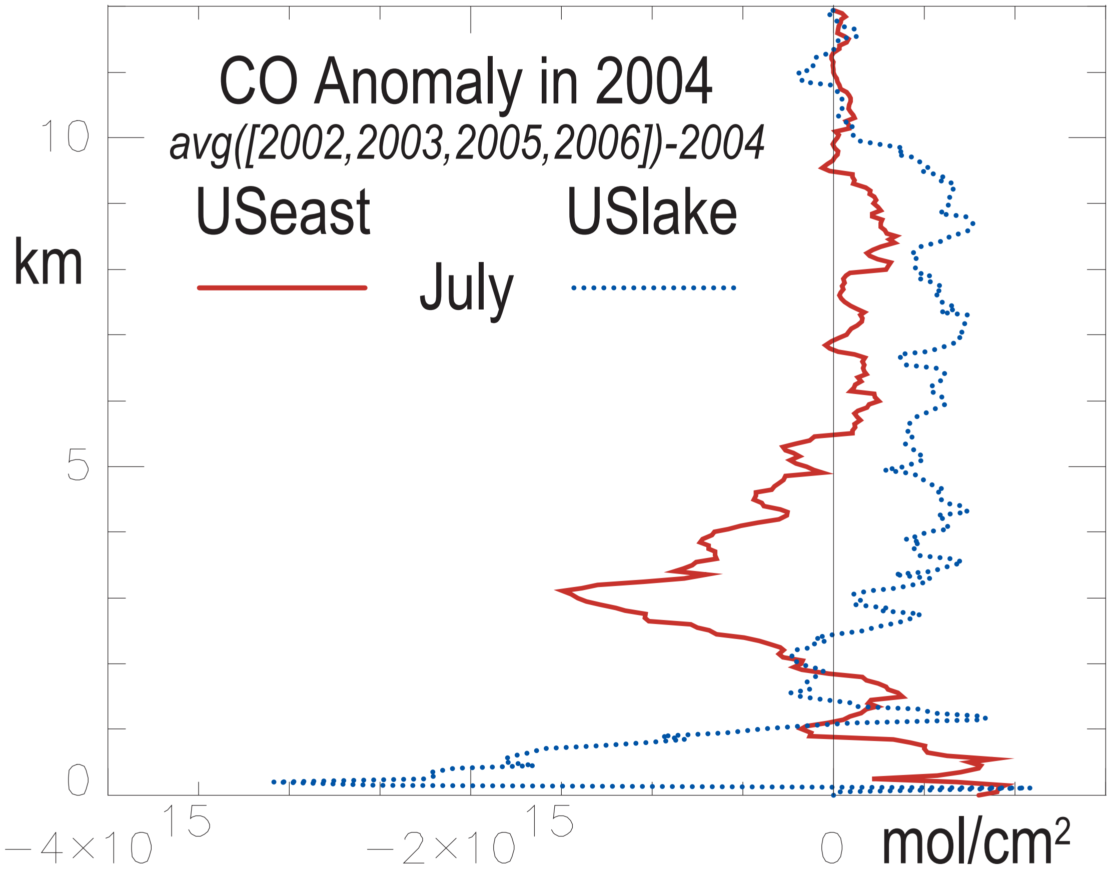

Fig. S2. Added as Figure S2.

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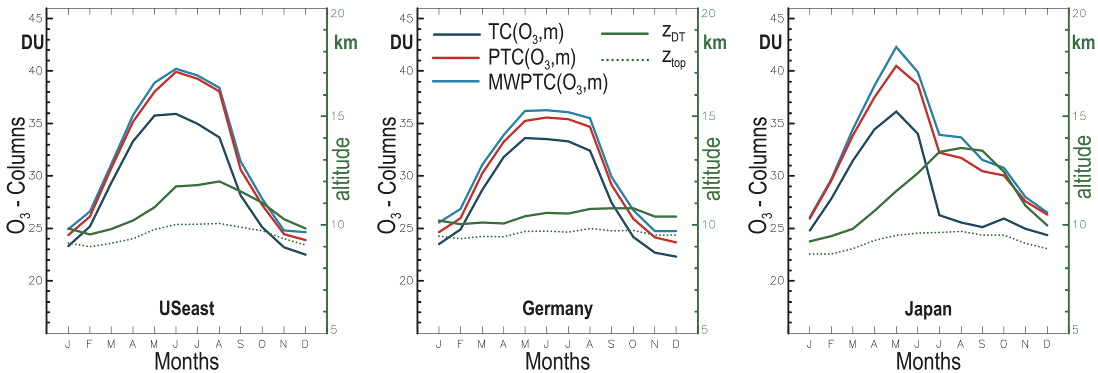

Fig. S2. to replace - Fig. 4.

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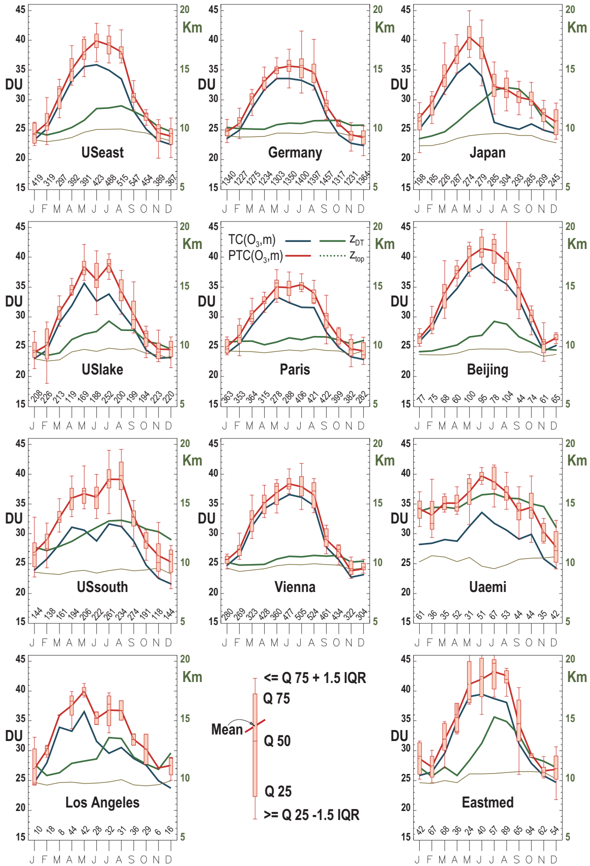

Fig. S2. to replace - Fig. 5.

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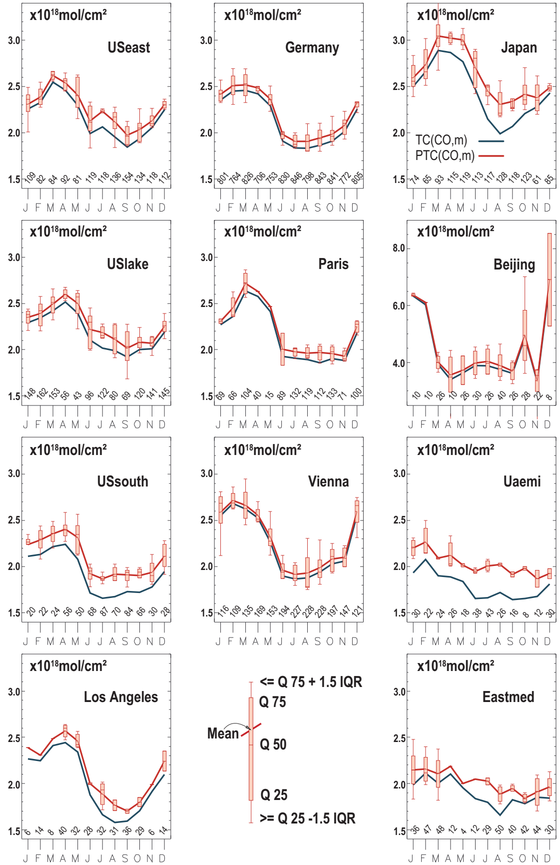

Fig. S2. to replace - Fig. 6.

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