

Dear Reviewer#2

We would like to thank you very much for reviewing our manuscript carefully and giving us many useful comments.

We agree your almost all of comments and suggestions, therefore the manuscript has been revised along your (and another reviewer's) comments. Please check the following our reply (black) of each your comments (blue).

In addition, we are sorry for late replying because it takes time to repair the data server which was broken down.

The major points that we deal with in the revised manuscript are as follows:

1. Figures were updated or removed.
 - ① Adding 2PVU (potential velocity unit) and OLR (outgoing longwave radiation) as the indicator of deep convection to Figures 1, 2, 3 and 5 of the revised manuscript, and zonal wind to Figure 2 of the revised manuscript
 - ② Figure 1 (plot of latitudinal distribution of increasing rate) in the previous manuscript was removed because Table 1 was enough information on it.
 - ③ Figure 3d (time-latitude cross section for 4-year at 250 and 500hPa) of the revised manuscript was drawn by the corrected data, because the previous figure was drawn by the data without the bias correction.
 - ④ Figure 5 (time-latitude cross section of inter-annual variation) in the previous manuscript was removed from the revised manuscript.
2. The discussion on CO₂ variation related with ENSO was moved to section of Discussion and Summary. Therefore the section name was changed to "Summary and Conclusion" to "Discussion and Summary" in the revised manuscript.
3. Many references recommended by reviewers were added to mainly the section of Introduction related with the *in situ* observational studies.

Review of Honda et al.,

First of all apologies for my late comment, which was due to unforeseeable issues. However I'm sorry to say, that I had a hard time reading the manuscript. The data set is very interesting and clearly of interest to the community. The discussion of the link to transport and dynamics is however misleading or partly wrong and neglects many aspects of transport (e.g.. the role of the tropopause as a barrier for transport and its effect on the CO₂ cycle amplitude or phase, which has already been discussed in several papers).

The paper presents observations of CO₂ from GOSAT from 2010 to 2013, which is analyzed at pressure levels of 500, 250, 150, and 100 hPa. The authors detrend the data by applying a simple linear empirical fit to build multiannual climatologies and anomalies. They show the zonal mean distributions of CO₂ and at the levels mentioned above to conclude on the transport processes, which cause the observed CO₂ distributions, but neglect relevant literature (Boering, Andrews, Sawa, Hoor, ...).

For the troposphere they state a transport time of two months from the LT (lower troposphere) to the UT (upper troposphere) and a dampening of 50% of the seasonal cycle amplitude. They relate this to the "absolute mixing ratio decreasing with altitude and to a lesser extent mixing with low CO₂ mixing ratio air mass".

Further they link tropical CO₂ mixing ratios to ENSO and identify interannual variability of the CO₂ in the monsoon region to different relations of vertical transport by convection and

horizontal transport "via anticyclonic circulation". The authors neglect relevant literature of CO₂ and its seasonal cycle from aircraft observations.

A: Thank you again for useful comments, especially providing us many related references. The results of previous studies have been added to the introduction section (the 3rd paragraph in page 2.) and proper location of the revised manuscript.

Figure 5 and related paragraphs with ENSO has been moved to "Discussion and Summary" section because, as you point out, long-term variability has many factors and has become presumptive discussion, so we decided to focus on the climatological descriptions of CO₂ at the revised manuscript.

The paper does not include any analysis of transport via e.g. Lagrangian methods, nor it shows links to surface observations or at least comparisons to the interannual variations of emissions or surface distributions or variability (zonally, globally or regionally, e.g. monsoon).

A: We agree that the discussion of UTLS CO₂ variation is needed to transport. As you pointed out, we added a description of the contribution of vertical and horizontal transport, and detailed descriptions of vertical and horizontal transport in the chapter on intra-seasonal variability with a short time scale (sub-section 3.4).

On the other hand, even though the results are very similar to ground observations, the upper air masses are the result of atmospheric transportation processes and mixing with other air masses. Therefore it is difficult to discuss UTLS CO₂ fluctuations from numerical calculations that cause chaotic errors, especially longer integration time causes larger error.

The authors further discuss transport and mixing particular at 250hPa, but do not even mention the term "subtropical jet", mixing barrier, isentropic transport, and consequently do not discuss their roles for the propagation of the seasonal cycle. They also state that Theta=370 K "indicates the physical surface of the tropopause", which is simply wrong.

They fully dismiss the role of the extratropical tropopause as transport barrier, when discussing the timing of the seasonal cycle and its amplitude change at the barrier.

A: We thank for making us aware of above points. Along your recommendation, the jet and physical surface '380 K' were added to figures, and the related description were added to the result section.

They state, that the role of seasonal CO₂-cycles has not been studied and neglect significant corresponding work: For the stratosphere above 100 hPa: Andrews et al., 1999, Boering et al., 1994, 1996, Strahan et al., 1998.

For the UTLS and lower stratosphere: Hoor et al., 2004, Engel et al., 2007, Boenisch et al., 2009. For upper troposphere and the monsoon: Schuck et al., 2010, Gurk et al., 2008.

A: Also many thank you for giving us these information. Those previous papers were added to the introduction and its suitable sections.

All in all there are too many speculations when linking transport and CO₂ observations. I recommend to resubmit it focusing on the climatologies and the observations and carefully linking them to e.g. surface seasonal cycles from global observational network for the LT/MT data. For the UTLS there must be a correct treatment of the tropopause particularly for the 150 hPa and 250 hPa level. One could e.g. derive distinct seasonal cycles for tropospheric and

stratospheric data, which can be compared to existing data sets (see references). Speculations about transport mechanisms should be removed.

A: Thank you for your very reasonable opinion.

The speculative sentences and diagrams were removed and other variables (wind field and OLR as indicator of convection) which make it easier to understand the transportation were added.

Therefore I can't recommend the paper for publication in the current form.

I do highly suggest a resubmission with a different focus, since the data set as such is very valuable, but the discussion of potential links to transport and mixing is inappropriate. I encourage the authors for resubmission either sharpening the transport discussion or just focusing on the climatologies.

A: We agree the reviewer's comment, we said again that the speculative sentences and diagrams were removed, especially the inter-annual variation related with ENSO was moved to the section of "Discussion and Summary".

Minor points: line 109: What is the vertical resolution and how do averaging kernels look like?

A: The information of vertical resolution and the averaging kernel was added to revised manuscript, P.4, l.121- p.5, l.125.

The averaging kernel in Figure 1 of Saitoh et al. (2016) shows the sensitivity of UT, particularly at 300 -- 200hPa at lower and middle latitudes. The present study used the level between 287.30 and 90.85 hPa as the UT-LS region. The numbers of retrieved layers vary from 9 to 14 (see table 1 of Saitoh et al. (2016)), which yield lower (upper) pressure levels of 287.30 (237.14), 237.14 (195.73), 195.73 (161.56), 161.56 (133.35), 133.35 (110.07), and 110.07 (90.85) hPa, respectively.

Fig. 1: Gradients appear at the tropopause. These were not accounted for.

The discussion of trends and Figure 1 illustrates an example of the coarse and insufficient discussions and speculations: The trend figure is discussed without any mentioning of the tropopause and its role for e.g. the mid-lat trend. The according table 1 provides trends for different latitude ranges, different altitudes without consideration or discussion of the tropopause. What shall one learn from this?

A: Figure 1 was removed from the revised manuscript because the increasing rate are found by Table 1.

Fig.6 and related discussion (lines 261-268): The monsoon plays of course a role for the observed 250 hPa CO₂ in Fig.6, but there is no discussion of potential surface emission variations, change of ENSO-related tropospheric circulation patterns, change of emission patterns, the authors state without any supporting analysis, that the observed CO₂ variability is related to variability of deep convection. How do the authors come to their conclusion? How is emission variability differentiated from large scale transport variability or convection?

A: The description on ENSO was removed from the revised manuscript and was moved to discussion section because there are many factors that affect long-term fluctuations, and it is not possible to discuss all of them in this study.

Fig.2 : Concerning the bias correction, which is also mentioned in the manuscript: Which role does the isentropic CO₂ gradient at the extratropical and subtropical tropopause play for the

bias correction? Did the authors consider the tropopause when calculating the bias?

A: The isentropic surface (380K potential temperature) and PVU=2 line were added to the figures (Figures 1,2,3, and 5) of the revised manuscript. With the revision of figure, the description of Figure 1 was modified in Sub-section 3.2, for example, line 190-192 in p.7.

This revised figure is easier to understand the horizontal and vertical transports, for example at 250 hPa, the CO₂ maximum at the lower latitude vary with vertical transport, on the other hand it is suggested that the extension of CO₂ higher mixing ratio to the higher latitudes at northern hemisphere across +2PVU line was affected by the horizontal transport.

Fig 2d) How do the cycles (e.g. at point Barrow and Mauna Loa) fit to the GOSAT observations at 500 hPa. Highest CO₂ at high latitudes should occur later than at low latitudes, since biological activity is delayed. How does this fit to Fig.2d? also line 165-168.

A: The following figure (FigureA) shows the CO₂ variations at MLO site, the GOSAT TIR CO₂ data was taken +/-5 degrees around MLO site. At 500hPa, the seasonal changes from GOSAT TIR are almost the same, and the amplitude decreases with increasing altitude, and the maximum peak timing is delayed by about one month. It was found that the UTLS CO₂ derived from GOSAT TIR shows the similar results with the previous studies.

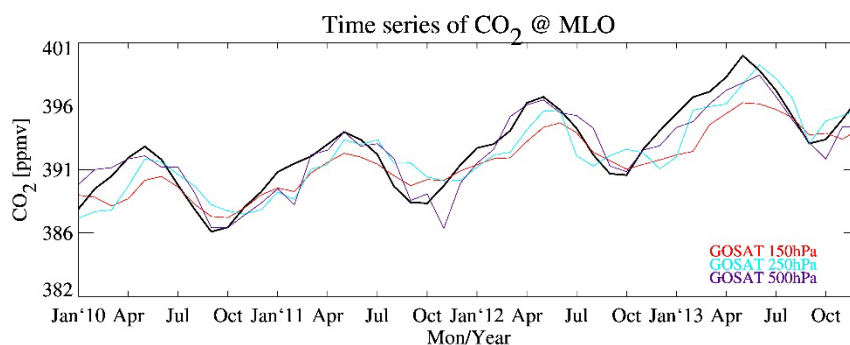


Figure S2: Time series of CO₂ at MLO. The black line shows the surface observation at MLO, the red, light blue and purple lines show the GOSAT observation averaged +/- 5 degree box around MLO at 150, 250 and 500 hPa, respectively.

1.184: The 370K isentrope defines the physical surface of the tropopause.

This statement is simply wrong. The tropopause is no way defined by isentropes (read Holton, 1995, Hoskins, 1991, Bethan, 1996...

A: The isentropic surface was modified as 380K potential temperature through the revised manuscript.

Fig. 3: How well is the troposphere resolved in the CO₂-data (vertical resolution, kernels, degrees of freedom)?

A: From the Table 1 and Figure 1 of Saitoh et al (2016), the vertical resolution and the averaging kernel (AK) were enough to discuss the vertical transport. The AK explanation was added to the revised manuscript, p.4, 1.121-122.

‘Caption’: Replace ‘vertical velocity’ with ‘pressure tendency’ – they have different signs.

A: The term was modified, p19.

Fig.4b)c): The data at 250 hPa are affected by the tropopause location, which inhibits quasi-horizontal (quasi-isentropic) transport. The phase propagation therefore is different from 100 hPa or 500 hPa (see Sawa et al., 2008, Hoor et al., 2004). Please add the (mean) 2 PVU and 4 PVU contour (also Fig. 6a)-d) and Fig.5 a)

A: The 2 PVU line was added to figures. With the revised figure, the description was modified in sub-section 3.3, p.8-9.

1.208/209: Which vertical gradient? Please calculate or plot (e.g. for different latitudes).

A: The following figure (FigureS3) shows the latitudinal distribution of the vertical gradient between 250hPa and UT/LS pressure levels. It was found that the latitudinal gradient between 10S and 30S in the southern hemisphere was steeper than that in northern hemisphere (10N-30N), and the temporal variation of latitudinal gradient in northern hemisphere was larger than that in southern hemisphere at each pressure levels.

The following sentence was added to the revised manuscript, p.8, 1.235-239.

“The latitudinal gradient between 250 hPa and 100 hPa in the Southern Hemisphere was steeper than in the Northern Hemisphere and, for example, the CO₂ mixing ratio decreased 1.45 ppmv from 10S to 30S and 0.71 ppmv from 10N to 30N in annual mean. However the latitudinal gradient in the NH varies seasonal (approximately 2.8 ppmv); the gradient in July (January) was steeper (gentler), on the other hand the seasonal variation of gradient in SH was smaller (approximately 0.9 ppmv).”

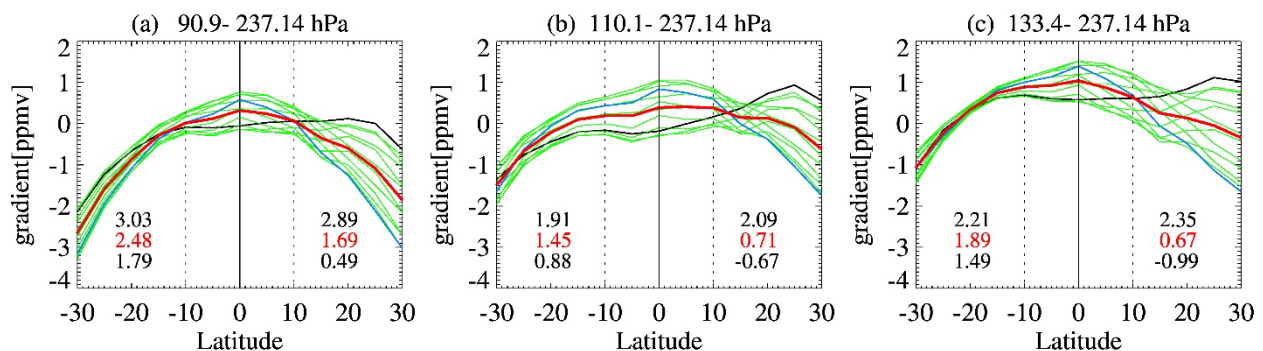


Figure S3: The latitudinal distribution of vertical gradient of CO₂ [ppmv] between 237hPa and 90 hPa (a), 110 hPa (b) and 133 hPa (c). The green lines show each month except January (black) and July (blue), the red line shows annual mean. The values in each panels indicate the maximum (top), average (red) and minimum (bottom) of latitudinal gradient between 10S(N) and 30S(N) at left (right) side of each plot.

1.210: This statement holds for any tracer and is very unspecific - the distribution of anything in the UTLs depends on vertical and horizontal transport in the troposphere.

A: This sentence was removed to avoid the confusion.

Fig.5: 500 hPa shows a trend of the anomaly at higher latitudes. Is this due to the (possibly wrong) linear trend estimate to derive the anomaly (eqn.1)?

A: Figure 5 was removed from the revised manuscript.

Also Figure 7: Why is the CO₂ maximum related to deep convection? Why do the data not

show any accumulation effect inside the anticyclone (see e.g. Baker, 2013, Schuck, 2010)?

A: The figure (Figure 6 in the revised manuscript) was modified. The revised figure shows the high mixing ratio was located within the Asian monsoon high at the June and July. And the explanation of this figure was written more carefully in Section 3.4, at 1.290-303 in p.10.