General.

We would like to thank the anonymous Referee #1 for providing comments to improve and clarify our manuscript. We will revise the text by fully taking the comments into account. Please find our responses to the specific comments and questions below.

Comments of Referee #1 and our responses to them

Comment

This paper describes a method of splicing together in situ measurements from ships, from aircraft, and from the ACTM model to create vertical profiles of CO2 over the Pacific Ocean. The vertical profiles are integrated to calculate XCO2 values that are then compared with the OCO-2, ACOS-GOSAT, and NIES-GOSAT retrievals over the same region. It's not clear to me whether ACP is the correct journal for this publication; it seems as though AMT might be a better fit for the paper's stated goals.

Response

Our manuscript, which describes a method to derive XCO_2 by using ship, aircraft and model data, doesn't intend to solely focus on the technical and theoretical aspects (with a rigorous uncertainty analysis). In addition to the technical aspects, our manuscript presents a detailed analysis of the spatiotemporal variations of CO_2 of each in situ and satellite dataset over the Pacific Ocean (section 4.1 and 4.2). Furthermore, using the new constructed in situ XCO_2 dataset, we demonstrate its application as reference for XCO_2, which is not only of relevance for validating satellite data, but especially for carbon cycle studies. As a complement to TCCON data, we believe that the applicability as reference for XCO_2 over oceans is of immediate relevance to a wide interdisciplinary scientific audience in atmospheric chemistry and physical sciences. Because our goal is beyond the primarily technical aspects, we think ACP would fit our goals better.

General comments:

- There are multiple ATom and HIPPO profiles throughout the Pacific – it would very much strengthen this paper if you could find coincident data with HIPPO/ATom profiles and compare vertical profiles in detail.

Response

We fully agree with the Referee #1 that HIPPO (Hiaper Pole-to-Pole Observations) and ATom (Atmospheric Tomography Mission) profiles would be very valuable to strengthen our results. However, coincident profile data of the HIPPO and ATom campaigns between the years 2014 and 2017 and in the longitude–latitude range of 130° E to 173° E and 30° S to 40°N do not exist. The newest dataset of HIPPO covers the year 2011 (HIPPO 4, HIPPO 5, https://www.eol.ucar.edu/node/3402, 12/21/2020). Data of the campaign ATom 1 cover the time period from 07/29/2016 to 08/23/2016 (https://daac.ornl.gov/cgi-bin/dsviewer.pl?ds_id=1581, 12/21/2020). Unfortunately, the flight tracks closest to our study region are generally more than 20° East (Figure R1, purple line). The lack of coincident data is a drawback in strengthening our results, but emphasises the need to expand the amount of reference data.

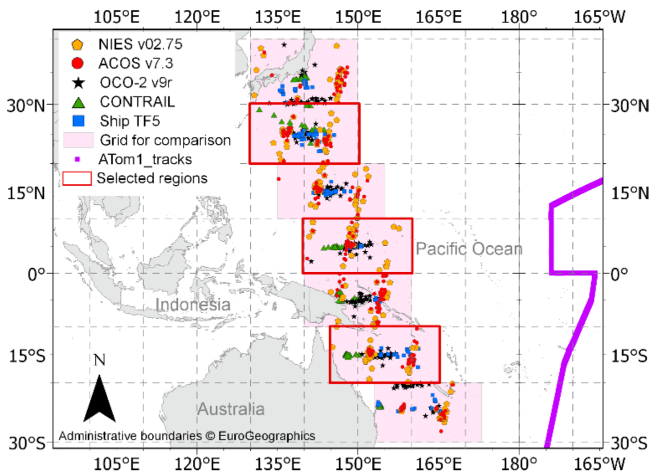


Figure R1. Comparison of the flight track of the Atom 1 campaign with the location of monthly averaged data of CO_2 from aircraft (CONTRAIL, green triangle), ship (Trans Future 5 - TF5, blue squares), the satellite retrievals from NIES (yellow diamonds), ACOS (red circles), and OCO-2 (black stars) between 2014 and 2017. Selected regions for the study within 10° latitude by 20° longitude boxes are shown in red frames. Administrative boundaries © EuroGeographics.

- It would further strengthen the paper if you could extend the most southern box another 4 degrees to 34S, where you could show that the combined in situ + ACTM total column matches that from the (coastal) Wollongong TCCON station (filtering for onshore wind direction, perhaps).

Response

We agree that this would be beneficial and of wide interest. Unfortunately, south of the latitude 28° S, the aircraft data of CONTRAIL between Narita, Japan, and Sydney, Australia, are only obtained over land (Figure R2, green triangle). Hence, an overlap with ship data over the ocean area is not given. By using our methodology and combining ship data from the open ocean area with aircraft data over land, no realistic CO_2 profiles can be obtained. Therefore, we cannot extent the study area to 34° S at present.

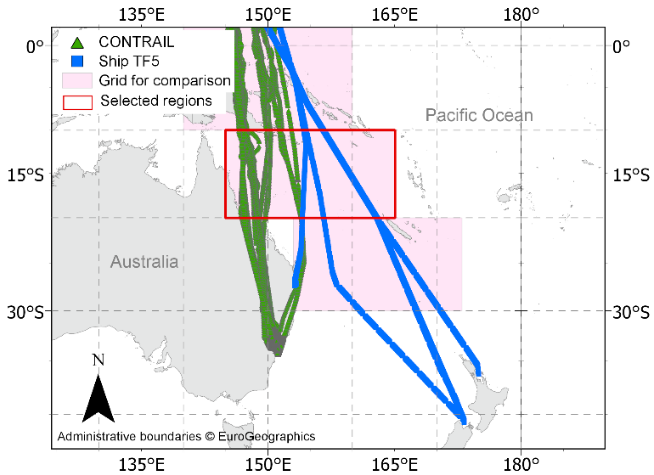


Figure R2. Location of the CO_2 data from aircraft (CONTRAIL, green triangle) and ship (Trans Future 5 - TF5, blue squares) between 2014 and 2017. Selected region for the study within 10° latitude by 20° longitude boxes is shown in the red frame. Administrative boundaries © EuroGeographics.

- I found the Results and Discussion section confusing in places (see Specific comments for details) and difficult to follow.

Response

We will revise the Results and Discussion section to clarify our statements. Please find our replies to the specific comments below.

- Uncertainties are large in the differences and trends, and yet conclusions were drawn about whether satellite measurements agreed with the ship+CONTRAIL+ACTM-derived XCO2.

Response

We agree that the uncertainties of the differences between the in situ derived XCO_2 and satellite XCO_2 shown in Table 3 and Fig. 5d-f are large, but significant in northern midlatitudes for ACOS (two-sided t-test, significance level α=0.05). The difference in the trends is not significant at northern latitudes, but at the equator for ACOS and OCO-2 (two-sided t-test, significance level α=0.05) Table 4.

Although uncertainties are not small, the comparison of the in situ derived XCO_2 dataset with satellite retrievals gives important indications on how good the retrievals currently are, and if newly revised retrieval algorithm are improved towards minimizing the difference or not. Figure A1 of Appendix A of the manuscript, as well as Figure R3 below, which shows the in situ derived XCO_2 and the data of OCO-2 v9 versus OCO-2 v10, illustrate the applicability of our new in situ derived XCO_2 dataset.

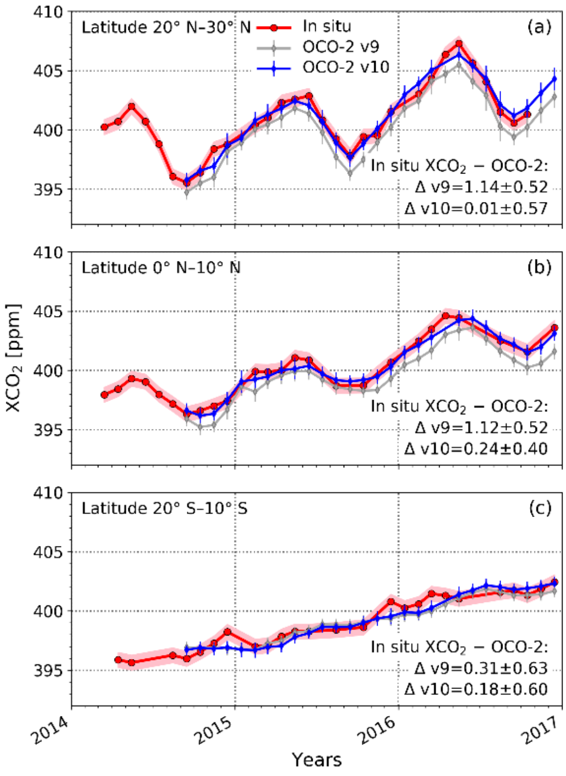


Figure R3. Comparison of the temporal variation of in situ XCO_2 (red) with XCO_2 derived from OCO-2 v9 (grey) and OCO-2 v10 (blue) for three selected latitude ranges. Red shaded areas are the uncertainty of the in situ XCO_2 which was obtained from the ±2 ppm variability in the in situ constructed CO_2 profile at ~850 hPa. Error bars show the standard deviation of the monthly averaged XCO_2.

We will add Figure R3 for illustration to Appendix A and clarify the uncertainties connected with our dataset and the uncertainties of the comparison in the revised manuscript as described below:

Line 302: *...and satellite* XCO_2 *in Fig. 5d-f.* ***The uncertainties of the in situ*** XCO_2 ***dataset are estimated to be 0.62 ± 0.01 ppm on average, which was derived from the ±2 ppm variation in the in situ adjusted*** CO_2 ***profile at 2 km above sea level (Lines 157-160).***

Line 308: *... average is found in the SH (Fig. 5c and 5f).* ***It is noted that the uncertainties of the differences between in situ*** XCO_2 ***and the satellite retrievals are large. However, the comparison indicates if the results of the current satellite retrievals tend to be higher or lower than in situ derived values. This is of importance for revising the retrieval algorithm in future.***

Specific comments:

- L38 – Why cite the 2018 value of atmospheric CO2? You could update this using the NOAA value for 2020.

Response

We revised the reference as follows:

Line 36: *Since the beginning of the Industrial Era in the 1750s, fossil fuel combustion and other human activities have increased the atmospheric concentration of* CO_2 *from approximately 277 ppm to more than 4****10*** *ppm in 20****20 (Ed Dlugokencky and Pieter Tans, NOAA/GML (www.esrl.noaa.gov/gmd/ccgg/trends/), 1/7/2021).***

- L108 – Why do you only use the tropospheric data in your analyses? Wouldn't the lower stratospheric data provide important constraints on the total column and provide a check on the stratospheric model?

Response

First, the CONTRAIL flights rarely went into the lower stratosphere during our study period. Therefore, we could have filled out the lower part of the stratosphere with aircraft data only occasionally. For our methodology, we think it is better to have a consistent stratosphere rather than using measurement data in a few profiles while most of the

remaining profiles use the results of the MIROC-4 ACTM only. Furthermore, the variation of CO_2 above the tropopause height varies much less than in the troposphere and can be successfully modelled (Figure R4).
Second, the aim of our study is not to provide a validation of the MIROC-4 ACTM in the stratosphere, which is already one of best validated stratospheric models at present using high altitude balloon-borne measurements of SF6 and CO_2-age-of-air (Patra et al., 2018).
We clarify the reason for excluding aircraft data of the stratosphere as follows:

Line 108: *Only those data which were obtained below the tropopause height during the cruise at around 11 km altitude are used.* ***Data of the lower stratosphere were only occasionally obtained. In order to have a consistent methodology for constructing CO_2 profiles as described in section 3.2, we screened out those data..***

- L125 – "By measuring the amount of light absorbed by CO2 and O2, the column average CO2 dry air mole fraction (XCO2) is estimated by taking ratio of the total column amounts of CO2 and O2, where O2 provides an estimate for the total column of dry air (Wunch et al., 2011)." This is true for TCCON, but I do not believe this is how the ACOS retrievals work. Please clarify.

Response

The Referee #1 is correct. Generally, XCO_2 quantifies the average mixing ratio of CO_2 in a column of dry air extending from the Earth's surface to the top of the atmosphere. It is derived by taking the ratio of the column integrated number densities of CO_2 and the total column of dry air. For the satellite retrievals, the total column of dry air is primarily derived from the surface pressure, which is mainly retrieved from the O_2 A-band in case of OCO-2, ACOS (O'Dell et al., 2012, 2018), and the GOSAT retrieval from NIES (Yoshida et al., 2011, 2013). Furthermore, the definitions of XCO_2 vary in how the dry air column is estimated and in how the vertical weighting is done (Crisp et al., 2012; O'Dell et al., 2012).
We revised the sentence as follows:

Line 125: *By measuring the amount of light absorbed by CO_2 and O_2, the column average CO_2 dry air mole fraction (XCO_2) is estimated by taking ratio of the total column amounts of CO_2 and the total column of dry air* (O'Dell et al., 2012, 2018; Wunch et al., 2011; **Yoshida et al., 2011, 2013**).

- Figure 2 – How does this profile compare with the GINPUT profile?

Response

Figure R4 shows the monthly averaged CO_2 profile calculated by GINPUT version 1.0.6 (purple line) in addition to the result of the MIROC-4 ACTM Model and the in situ derived CO_2 profile of Figure 2. Especially in the lower troposphere, the GINPUT profile differs. This is explained by the fact that the TCCON prior do not try to capture the effect of emissions and do not ingest global flux dataset nor any longitudinal dependent behaviour. In contrast, the MIROC-4 ACTM Model uses realistic flux and transport simulations and therefore, the ACTM derived profile is close to that derived from in situ measurements. The difference in the stratosphere is small as compared to the troposphere.

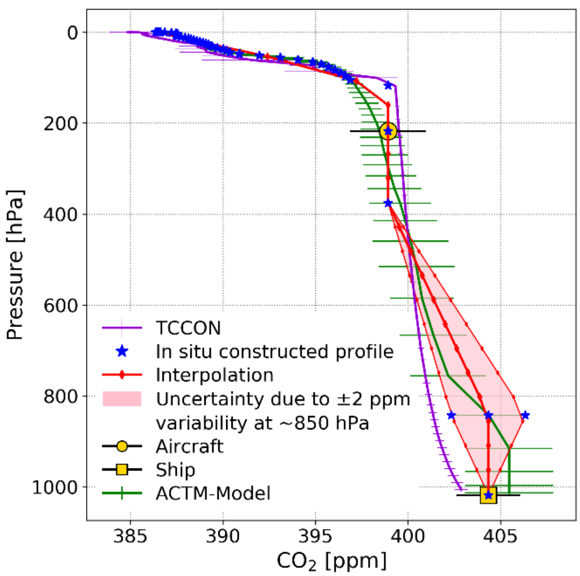


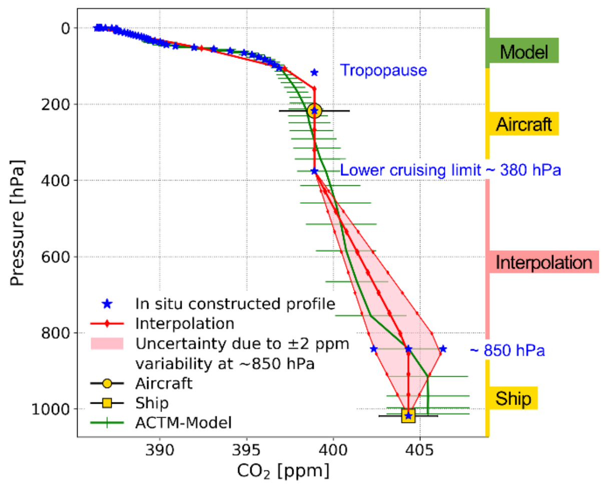
Figure R4. Comparison of the in situ constructed CO_2 profile (blue) with that obtained from the ACTM-Model (green) and the TCCON a-priori profile of CO_2, calculated by GINPUT version 1.0.6 (purple line). The example is obtained at the latitude 20° N–30° N, March 2014.

We clarify why we are using the results of the ACTM-model instead of those from GINPUT as stated under the comment L172 below.

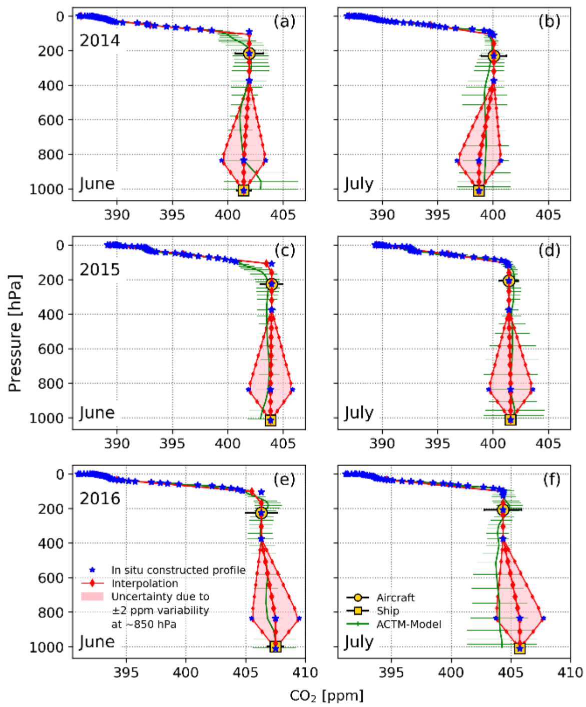
- If I understand correctly, the blue stars are a combination of model, in situ, and extrapolated data, is that correct? If so, calling it the “in situ” profile is misleading.

Response

The Referee #1 is correct, the blue stars are a combination of model, in situ, and extrapolated data. We intended to make it clear by using the terms "in situ constructed" in Figure 2 and "in situ adjusted profile" in the Figure caption. We revised the labelling in Figure 2, as well as in Figure A3 of Appendix A. To be consistent with the text, we use the term "in situ constructed profile" in the Figure caption. Furthermore, we clarify the definition of the "in situ constructed profile" in the Figure caption as follows:



Line 169: *Figure 2. Construction of the in situ* ***constructed*** *CO_2 profile (blue)* ***obtained*** *by using ship (SOOP) and aircraft (CONTRAIL) data (yellow) together with the results of the ACTM (green), and the interpolation (red). The example is obtained at the latitude 20° N–30° N, March 2014.*



Line 430: *Figure A3. In situ constructed CO_2 profiles (blue)* ***obtained by*** *using ship (SOOP) and aircraft (CONTRAIL) data (yellow), together with the results of the ACTM (green), and the interpolation (red) for the month June and July in 2014 a), b), 2015 c), d), and 2016 e), f) at the latitude range 20° N–30° N.*

- L172 – Why use the MIROC-4 ACTM for the stratosphere instead of the GINPUT stratosphere? How do they compare?

Response

First, using the MIROC-4 ACTM means that our method is fully independent of TCCON which is important for using our methodology as complement for evaluating satellite retrievals. If TCCON and our method are independent, and satellite retrievals show a similar bias to both datasets, then the observed difference is a bias of the satellite data. In addition, the GINPUT prior is used in the OCO-2 v10. Therefore, having an independent stratosphere is a good cross check.

Furthermore, the MIROC-4 ACTM simulates the realistic CO_2 fluxes and transport processes as described above. GINPUT start from the average CO_2 mixing ratio measured at Mauna Loa and American Samoa and then imposes a latitudinally dependent seasonal cycle to derive the specific CO_2 profiles. Nevertheless, the CO_2 mixing ratios in the

stratosphere are varying much less than in the troposphere and therefore, the results of the MIROC-4 ACTM and GINPUT are similar as seen in Figure R4. We clarify the choice of the MIROC-4 ACTM as follows:

Line 172: *To account for the stratospheric partial column, we used results of the MIROC-4 ACTM (Patra et al., 2018) above the TROPPB* ***(Fig. 2) instead of the results from GINPUT. First, by using the MIROC-4 ACTM, our method is fully independent of TCCON which is important for using our methodology as complement to evaluate satellite retrievals. Second the MIROC-4 ACTM uses realistic flux and transport simulations and is one of the best validated stratospheric models at present.***

Line 196: *Second,* ***as mentioned earlier,*** *the MIROC-4 ACTM is among the best validated stratospheric models* ***using high altitude balloon-borne measurements of SF6 and CO_2-age-of-air*** *(Patra et al., 2018).*

- L335 – "Hence, even though no assumption was necessary at that period, the negative bias persists (Fig. 5d, Fig. 6e), which indicates that the difference between in situ and satellite XCO2 can be linked to measurement uncertainties of the satellites." I do not follow this logic. Why couldn't the bias be caused by a bias in the ACTM stratosphere and not in the satellite retrievals?

Response

The variation of CO_2 in the stratosphere is much less as in the troposphere and can be simulated with high precision by the MIROC-4 ACTM (Patra et al., 2018). Furthermore, as described in Lines 197 – 199, our sensitivity test revealed that the impact of the stratospheric part on the calculated XCO_2 is as small as 0.2 ± 0.1 ppm on average. Based on the error induced by the stratosphere and the uncertainty derived from the variability in the in situ constructed CO_2 profile at ~850 hPa (0.62 ± 0.01 ppm), the largest assumed reasonable bias is 0.9 ppm. This bias is not enough to explain the observed average negative discrepancy of 1.2 ± 0.4 for ACOS and OCO-2 from June to September in 2014 to 2017. We add the impact of the stratosphere as follows:

Line 335: *Niwa et al. (2011) found similar straight vertical profiles between June and September in East Asia, based on aircraft observations and model results.* ***Furthermore, any bias due to errors in the MIROC-4 ACTM stratospheric CO_2 profile are smaller than the average difference of 1.2 ± 0.4 ppm between the in situ constructed XCO_2 and satellite observations of ACOS and OCO-2 between June and September (section 3.2).***

- L353 – "The consistency with long-term studies support the correctness of the in situ XCO2, which implies that satellite XCO2 sometimes show a delayed response to CO2 changes." Again, I do not follow this argument. The satellites measure the total column in the atmosphere at the time of the measurement. Are you saying that the satellite measurements are wrong?

Response

As described in Lines 350-353, long-term in situ measurements in the upper troposphere and at surface level report maxima and minima of CO_2 not later than May and September, while the satellite retrievals show the extreme values sometimes one month later. Based on the long-term in situ datasets, maxima in June and minima in October are too late. We do not intend to say that the satellite measurements are wrong. Our observations suggest that these positive phase shifts of the satellite data are caused by remaining uncertainties which are introduced by limitations in the retrieval algorithm or the lack of validation data. The lack of validation data makes it difficult to characterize and correct these uncertainties. We know that from GOSAT and OCO-2 the retrieval algorithm to obtain XCO_2 from the measured radiance are undergoing rapid progress with almost one new version per year for OCO-2. We are hoping that the highly accurate ship and aircraft data over a unique geographical region will help us to build the capacity for the validation of satellite XCO_2 retrievals.
We clarify our statement as follows:

Line 353: *The consistency with long-term studies support the correctness of the in situ XCO_2, which implies that satellite XCO_2 sometimes show a delayed response to CO_2 changes****, which might be caused by remaining uncertainties introduced by limitations in the retrieval algorithms or the lack of validation data.***

- L359 – "In contrast, a significant increase of 3.84 ± 0.65 ppm yr−1 is observed by in situ XCO2 from 2015 to 2016, which is by _10% larger than that observed by satellites (3.39 ± 0.03)." Firstly, I don't see 3.39 ± 0.03 in Table 4 – is this a typo? Secondly, these numbers do not differ by 10% - their uncertainties overlap and therefore you cannot say anything conclusive about how they differ.

Response

On average, the increase of the mean values of all three satellite retrievals is 3.39 ± 0.03. That means, the value 3.39 ± 0.03 is the average increase and its standard deviation of the mean values of all satellite retrievals in the period 2015 to 2016. We added a column to Table 4 with the average values as shown below.

The Referee #1 is correct that the uncertainties overlap. The difference in the increase of the in situ derived XCO_2 and that of the satellite XCO_2 isn't significant at northern latitudes, but the increase of the in situ derived XCO_2 tends to be slightly higher. At the equator, the increase of the in situ XCO_2 is significantly higher than that of ACOS and OCO-2 (two-sided t-test, significance level α=0.05). We revised this part as follows:

Line 359: *In contrast, a significant increase of 3.84 ± 0.65 ppm yr^{-1} is observed by in situ XCO_2 from 2015 to 2016.* ***The average increase of the mean values of all satellite retrievals is 3.39 ± 0.03. This*** *rapid increase is also seen near the equator,* ***where the increase of the in situ XCO_2 is significantly higher than that of ACOS and OCO-2 (two-sided t-test, significance level α=0.05). S****imultaneously**, a larger negative bias of the satellite XCO_2 in 2016 as compared to the previous years* ***is observed*** *(Figs. 5b and 5e).*

Table 4. *Increase of XCO_2 between peaks of consecutive years and the standard error of the difference seen by in situ and satellite XCO_2 of GOSAT (NIES, ACOS) and OCO-2 between 2014 and 2017. Peak values are defined as mean of the three consecutive highest monthly averages during spring of each year. In 2016, the mean of ACOS and that of in situ XCO_2 at 0° N–10° N is based on 2 months due to limited data. "–" indicates missing data.* ***The right column shows the average increase of all satellite means and its standard deviation.***

	In situ XCO_2 (ppm yr^{-1})	**NIES (ppm yr^{-1})**	**ACOS (ppm yr^{-1})**	**OCO-2 (ppm yr^{-1})**	**Avg. all satellites (ppm yr^{-1})**
			20° N–30° N		
2014–2015	1.45 ± 0.63	1.42 ± 0.60	1.95 ± 0.54	–	**1.68 ± 0.26**
2015–2016	3.84 ± 0.65	3.37 ± 0.43	3.43 ± 0.40	3.36 ± 0.38	**3.39 ± 0.03**
			0° N–10° N		
2014–2015	1.72 ± 0.22	–	1.99 ± 0.30	–	–
2015–2016	3.87 ± 0.09	–	2.82 ± 0.37	3.52 ± 0.16	**3.17 ± 0.35**

Technical comments:

- L55 – change "improves" to "improve"

Response

We revised the sentence as follows:

Line 54: *These observations are most sensitive to the lower troposphere where CO_2 is most variable (Patra et al., 2003) and therefore, are able to improve the knowledge on local CO_2 emission and sinks (Connor et al., 2008).*

- L56 – change "the second NASA" to "NASA's"

Response

We revised the sentence as follows:

Line 56: *Japan's Greenhouse gases Observing Satellite (GOSAT), and the second NASA**'s** (National Aeronautics and Space Administration) Orbiting Carbon Observatory (OCO-2) are dedicated to inferring the concentration of GHGs from high-resolution spectra at NIR and SWIR wavelengths.*

- L71 – TCCON has a very limited number of sites observing *the atmosphere over*open oceans. I'm not sure how you define this, since there are several coastal and island TCCON stations (e.g., Réunion Island, Ascension Island, Izaña, Burgos, Darwin, Wollongong) and the TCCON footprint is large enough that it would be sensitive to CO2 over oceans.

Response

The Referee #1 is correct that there are TCCON stations at coastal and island sites. However, wide areas over the open ocean, which we define as the area outside the coastal region, are not covered by TCCON stations as shown in Figure R5. Therefore, we speak of "very limited number". Even though the footprints of TCCON stations are large, reference data measured directly above open water areas provide a valuable complement to TCCON stations.

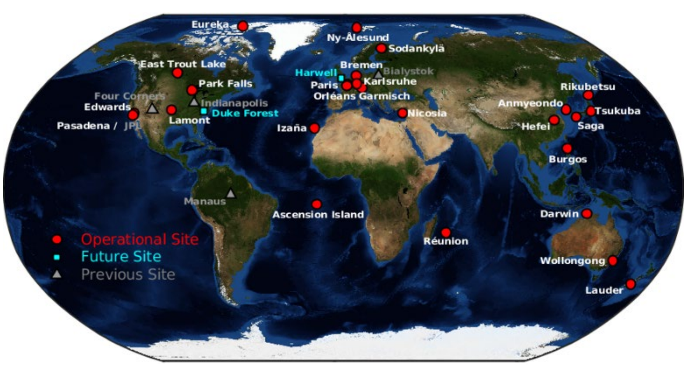


Figure R5. Location of current, future, and previous TCCON stations. Data are from 2/11/2020 (https://tccon-wiki.caltech.edu/Main/TCCONSites, 1/8/2021).

We clarify the definition of "the atmosphere over the open ocean" as follows:

Line 71: However, TCCON **sites are land based and** very limited number of sites observ**e the atmosphere over** open oceans**, which are defined as the ocean area outside the coastal region**.

We corrected some typos and a few values caused by a mistake in the calculation script as follows:

Line 173: *In short, the MIROC-4 ACTM uses a hybrid vertical coordinate to resolve gravity wave propagation into the stratosphere.*

Line 179: *A high accuracy of the MIROC-4 ACTM is indicated by the agreement of simulated "age of air",..*

Line 302: *In all latitudes, in situ and satellite XCO_2 show an overall significant positive correlation (R^2: NIES = 0.84 ± 0.02, ACOS = 0.74 ± 0.0**8**, OCO-2 = 0.8**2** ± 0.0**5**) (Table 2).*

Line 307*: … and decreases by 40% (0.5**6** ppm) on average between the northernmost and southernmost regions (Table 2).*

Line 318:

	R^2			RMSE		
Latitude	**NIES**	**ACOS**	**OCO-2**	**NIES**	**ACOS**	**OCO-2**
20° N–30° N	0.86	0.64	0.81	1.06	1.70	1.26
0° N–10° N	0.81	0.76	0.76	1.02	1.17	1.23
20° S–10° S	0.8**5**	0.8**2**	0.8**8**	0.8**4**	0.**79**	0.7**0**

Line 321:

	difference in situ XCO_2 – satellite XCO_2					
Latitude	**Avg. NIES**	**Std.**	**Avg. ACOS**	**Std.**	**Avg. OCO-2**	**Std.**
20° N–30° N	0.61	0.87	1.60	0.59	1.14	0.52
0° N–10° N	0.51	0.87	1.00	0.60	1.12	0.52
20° **S**–10° S	0.2**0**	0.8**1**	0.4**8**	0.6**3**	0.3**1**	0.6**3**

Line 410: *An initial comparison of the in situ XCO_2 dataset with ACOS v9r revealed a decrease of the negative bias by 50%* ***on average*** *at northern midlatitudes as compared to ACOS v7.3 (Fig. A1)****, and by more than 90% of the average values of OCO-2 v10 as compared to OCO-2 v9.***

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