Response to Referee #2' Comments

MS No.: amt-2021-92
MS type: Research article
Title: Atmospheric Carbon Dioxide Measurement from Aircraft and Comparison with OCO-2 and Carbon Tracker Model Data

Dear Professor,

Thank you very much for spending your valuable time in reviewing our research paper and providing the list of comments/suggestions. You have put forward detailed and specific modification suggestions for the article, including the layout of the article, the citation of references, the interpretation of formula parameters, etc. We have responded to your modification suggestions one by one and made corresponding modifications to the manuscript. The amendments are mentioned below. Your valuable comments play a very important role in improving the article. Thank you again for your valuable comments

Report #2:

General

The paper introduces aircraft based measurements of column weighted CO2 mixing ratio using a lidar at the Chinese coast including comparison with satellite data. The paper is within the scope of the journal but especially section 3.2 requires revision because of misleading sentences. Here the authors should use the papers of the other groups applying similar methods (Refaat, Amediek in introduction). Sections 3.4 and 3.5 are too short.

Response: We are very thankful to you for your kind words and positive feedback about our article. We have followed your suggestions carefully and revised the manuscript accordingly. We have modified misleading sentences, Eq. 7 and Eq. 8 have been explained in detail. Besides, we expanded Sections 3.4 and 3.5 to explain the experimental results in more detail. The corrections have been made in the revised manuscript. Thank you for your suggestions.

Line 197: In our experiment, the random noise followed Gaussian distribution. When the points on the pulse are superposed, the sum continues following the Gaussian distribution of $N(\rho^l, (\varepsilon^l)^2)$, where the mean and the standard deviation are

$$\rho^l = \frac{1}{N}\sum_{k=1}^{N} \alpha_k^l \, , \tag{7}$$

$$(\varepsilon^l)^2 = \frac{1}{N^2}\sum_{k=1}^{N} \left(\sigma_k^l\right)^2 \, , \tag{8}$$

Where, N is the point number of the pulse, ρ^l and ε^l represent the mean and standard deviation. α_k^l is the value of each point on the pulse, and σ_k^l is the standard deviation of each point. Hence, the SNR of the sum can be written as

$$SNR_{PIM}^{l} = \frac{\rho^{l}}{\varepsilon^{l}} = \frac{\sum_{k=1}^{N} \alpha_k^{l}}{\sqrt{\sum_{k=1}^{N} (\sigma_k^{l})^2}}, \tag{9}$$

Therefore, we can choose the number of points on the pulse to improve the SNR of each pulse.

Line 271:

3.4 OCO-2 Measurement Results

During this flight experiment, the OCO-2 passed over the flight area on March 16 and the observations over the study area are shown in Figure 18. The solid red line in figure 18(a) is the flight path of the aircraft. The yellow mark point is the position of the suborbital point of the OCO-2 trajectory in the flight area. Figure 18(b) shows the XCO_2 results detected by OCO-2. Figure 18(c) shows the corresponding standard deviation production of OCO-2. As can be seen from Figure 18(a), OCO-2 observations covered both ocean and land surfaces. Due to the fast flight speed of the satellite, the data time period falling in the study area was from 12:57:25 to 12:57:38 UTC. A quality flag was applied to the satellite dataset and the cloud-contaminated retrievals were removed. In the flight area, there is little difference between the values of XCO_2 measured by OCO-2 over land and ocean areas. The average value of XCO_2 over land area is 414.28 ± 0.81 ppm and that over ocean area is 414.23 ± 0.55 ppm. However, due to the uneven distribution of CO_2 volume mixing ratio in the land area, the standard deviation of XCO_2 products over the land area is larger than that over the ocean. The XCO_2 measured by OCO-2 varied from 401.66 ppm to 418.80 ppm, with an average of 414.25 ± 0.62 ppm.

3.5 Vertical Profile Comparison of CO_2 Concentration

The measurement results of the airborne greenhouse gas analyzer were compared with those of OCO-2 inversion and Carbon Tracker model, which is a global carbon cycle data assimilation system. The comparison results are shown in Figure 19. The CarbonTracker dataset was interpolated to the location of the experimental site. During the flight campaigns, the OCO-2 satellite passed over the flight area on March 16. Therefore, the data results of OCO-2 on March 16 were compared with those of CarbonTracker and in-situ data on March 14, March 16 and March 19, respectively. As can be seen from the detection results in Figure 19, the structural change of CO_2 concentration with height can be roughly divided into two parts. From the ground to the height of 4 km and above 4 km. Below 4 km, the detection results of OCO-2, airborne greenhouse gas analyzer and CarbonTracker model show a same decreasing of CO_2 concentration value with the increase of altitude but the values are different. The difference between the average values of CO_2 concentration obtained by the OCO-2 and the airborne greenhouse gas analyzer below 4 km on March 14, March 16 and March 19 were -1.3 ppm, 0.79 ppm, and 1.3 ppm, respectively. These three methods can well detect that the land in northeast China was the source of CO_2 in March. This change result of airborne greenhouse gas analyzer and Carbon Tracker is more obvious than OCO-2. On March 19, CO_2 concentration measured by the airborne greenhouse gas analyzer decreased from 430.3 ppm at 0.34 km to 413.09 ppm at 3.18 km. The computed results of CarbonTracker decrease from 429.75 ppm at 0.59 km to 415.7 ppm at 2.68 km. The CO_2 concentration result of OCO-2 decreased from 414.55 ppm on the ground to 412.39 ppm at 3.02 km. When the altitude is higher than 4 km, the CO_2 concentration is almost constant. This might be due to the stability of the atmosphere above.

Specific

Point 1:
Line 143ff: "two-way nested chemistry-transport model Tracer Model 5" (see also Peters et al, 2004). Improve sentence, it is not consistent with the provided references. The reference to CarbonTracker (Babenhauserheide et al, 2015) should be earlier. Unfortunately, the references use different full names for TM5 but not 'transfer model'.

Response 1: We are thankful to the reviewer for the valuable suggestion. We have revised the citation of references, and highlighted the revised/modified text with red font. Relevant changes have been made in the revised version of the manuscript at:

Line 154: CarbonTracker is an inverse model framework developed by (Peters et al., 2004). It combines the two-way nested transfer model 5 (TM5) with offline Atmospheric Tracer transfer model and updates the atmospheric CO_2 distribution and surface fluxes every year (Krol et al., 2005).

Line 413: Krol, M. C., S. Houweling, B. Bregman, M. van den Broek, A. Segers, P. van Velthoven, W. Peters, F. J. Dentener, and P. Bergamaschi (2005), The two-way nested global chemistry-transport zoom model TM5: Algorithm and applications, Atmos. Chem. Phys., 5, 417– 432.

Line 449: W. Peters, M. C. Krol, E. J. Dlugokencky, F. J. Dentener, P. Bergamaschi, G. Dutton, P. v. Velthoven, J. B. Miller, L. Bruhwiler, and P. P. Tan, Toward regional-scale modeling using the two-way nested global model TM5: Characterization of transport using SF6, J. Geophys. Res., 109, D19314, doi:10.1029/2004JD005020, 2004.

Point 2: I suppose the trace gas is CO_2 here, i.e., online means on a CO_2 line. If yes please say so.

Response 2: We are thankful to the reviewer for the valuable suggestion. We have replaced " trace gas " by "CO_2". Relevant changes have been made in the revised version of the manuscript at:

Line 162: The laser pulse of the online wavelength was strongly attenuated because it was absorbed by the CO_2 molecules while propagating through the atmosphere.

Point 3: "hard target": is that the surface or the cloudtop? Please be more precise here.

Response 3: We are thankful to the reviewer for the valuable suggestion. "hard target": is the surface, we have revised the mistake. Relevant changes have been made in the revised version of the manuscript at:

Line 170: R_G is the height of the surface above sea level.

Point 4: Please define all quantities in equations in the text. What is for example Pp? Use lower case for atmospheric pressure (p).

Response 4: We are thankful to the reviewer for the valuable suggestion. We have defined all quantities in equations. And we used lower case for atmospheric pressure in all equations and text. we highlighted the revised/modified text with red font. Relevant changes have been made in the revised version of the manuscript at:

Line 180:

$$V = P_P * \Re_v , \tag{3}$$

where $\Re_v$(V/W) represents the voltage response rate of the APD detector, P_P is the power of echo signal, V is the voltage.

Line 176:

$$\tau_{CO_2} = \int_{R_G}^{R_A} \Delta\sigma_{CO_2}(p(r), T(r)) N_{CO_2}(r) dr = \frac{1}{2} \cdot ln\left(\frac{P(\lambda_{off}, R) \cdot P_0(\lambda_{on})}{P(\lambda_{on}, R) \cdot P_0(\lambda_{off})}\right) , \tag{2}$$

where $\Delta\sigma_{CO_2}$ is the differential absorption cross section of the online and offline wavelengths, N_{CO_2} is the molecular density of the CO_2. p and T are pressure and temperature profiles.

Line 189:

$$IWF = \int_{R_G}^{R_A} \frac{N_A \cdot p(r) \cdot \Delta\sigma_{CO_2}(p(r), T(r))}{RT(r)\left(1 + X_{H_2O}(r)\right)} dr , \tag{6}$$

where N_A is the Avogadro's constant, R is the gas constant, $p(r)$ and $T(r)$ are the pressure and temperature profiles, respectively. X_{H_2O} is the dry-air ratio of water vapor, IWF represents the integral weight function.

Point 5: Section 3.1: Please improve text for the non-expert reader.

Response 5: We are thankful to the reviewer for the valuable suggestion. We have improved text about section 3.1. We highlighted the revised/modified text with red font. Relevant changes have been made in the revised version of the manuscript at:

Line 209: The performance of the ACDL system was evaluated by comparing the original echo signals over three different surface types, including the ocean, the mountain, and the urban residential surface types. The original signals of the ACDL over the ocean, urban residential, and mountainous areas are shown in Figures 4, 5, and 6, respectively. Including local amplification of each signal. The amplification signals from left to right are online monitor signal, online echo signal, offline monitor signal and offline echo signal. In each group of original echo signals, the online and offline monitor signals are fixed at the same position but the echo signals appear in different positions due to the different heights of the ground surface. The original signals were filtered before using, and the signals whose pulse peak values were not in the linear region of APD were discarded. The echo signals in the ocean area were significantly smaller than those over the residential and the mountain areas. This might be due to the low reflectivity of the ocean, which leads to the reduction of the signal noise ratio (SNR) over the ocean.

Point 6: Section 3.2: I suppose Eq. 7 describes the signal and Eq.8 the noise, if yes please write that (see also reviewer #1). Please correct misleading sentences. What is on the abscissa of panel a of Figs. 7-9 (with units)?

Response 6: We are thankful to the reviewer for the valuable suggestion. Eq. 7 and Eq.

8 have been explained in detail, and the abscissa of Figs. 7-9 is marked. We highlighted the revised/modified text with red font. Relevant changes have been made in the revised version of the manuscript at:

Line 197: In this study, the PIM uses the integrated value of the points on the pulse to calculate DAOD. In our experiment, the random noise followed Gaussian distribution. When the points on the pulse are superposed, the sum continues following the Gaussian distribution of $N(\rho^l,(\varepsilon^l)^2)$, where the mean and the variance are(Zhu et al., 2020; Yoann et al., 2018)

$$\rho^l = \frac{1}{N}\sum_{k=1}^{N} \alpha_k^l \, , \tag{7}$$

$$(\varepsilon^l)^2 = \frac{1}{N^2}\sum_{k=1}^{N} \left(\sigma_k^l\right)^2 , \tag{8}$$

Where, N is the point number of the pulse, ρ^l and ε^l represent the mean and standard deviation. α_k^l is the value of each point on the pulse, and σ_k^l is the standard deviation of each point. Hence, the empirical estimate of the SNR of the equivalent measurement on the whole averaging window can be written

$$SNR_{PIM}^l = \frac{\rho^l}{\varepsilon^l} = \frac{\sum_{k=1}^{N} \alpha_k^l}{\sqrt{\sum_{k=1}^{N} \left(\sigma_k^l\right)^2}} , \tag{9}$$

Therefore, we can choose the number of points on the pulse to improve the SNR of each pulse.

Line 526:

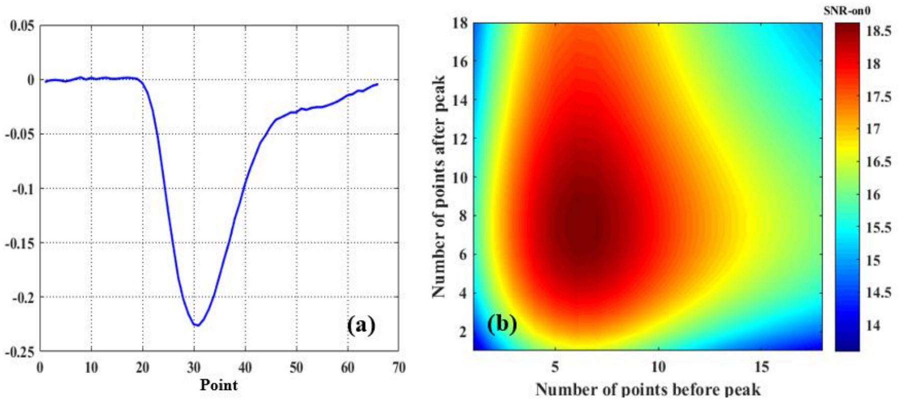

Figure 1: (a) Online wavelength monitoring pulse signal. (b) The change of pulse signal SNR with the number of selected pulse points.

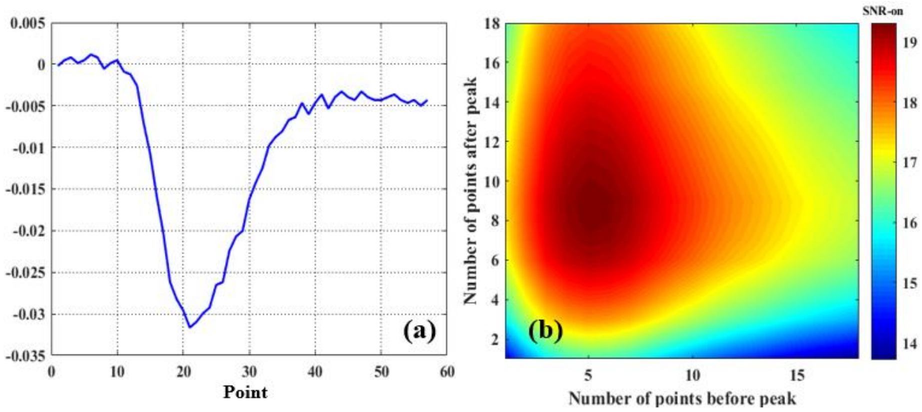

Figure 2: (a) Online wavelength echo pulse signal in land area. (b) The change of the SNR of the echo pulse signal in the land area with the number of selected pulse points.

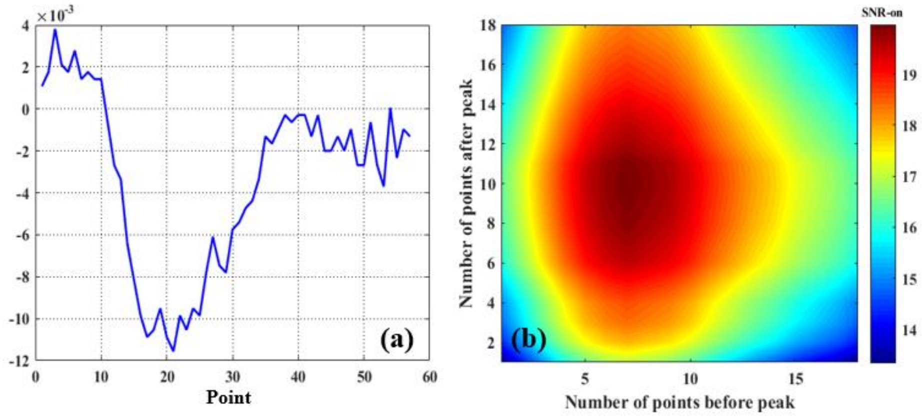

Figure 3: (a) Online wavelength echo pulse signal in ocean area. (b) The change of the SNR of the echo pulse signal in the ocean area with the number of selected pulse points.

Point 7: I would suggest to rearrange section 3.3 to have every result for the 14 March flight together, i.e. exchange the paragraph beginning with line 245 with the part from line 226 to line 244.

Response 7: We are thankful to the reviewer for the valuable suggestion. We have rearranged section 3.3 and highlighted the revised/modified text with red font. Relevant changes have been made in the revised version of the manuscript at:

Line 244: Figure 14 shows the comparison of the XCO_2 calculated from the ACDL measurements with the dry-air mole fraction of CO_2 measured using the UGGA. Both of the datasets show a good agreement by exhibiting a similar variation trend. The results from the two datasets also show that the volume mixing ratio of the atmospheric CO_2 is highest over the residential area and the lowest over ocean surface. The average value of XCO_2 obtained by the ACDL calculations was 426.27 ppm, and the average value of CO_2 mole fraction obtained by the UGGA measurements was 413.91 ppm. Moreover, the standard deviation of the UGGA observations was smaller than that of the ACDL measurements, and this might be due to the different working principles of the two instruments. The ACDL measures the weighted average concentrations at different altitudes. However, the UGGA measures the CO_2 value at the aircraft location.

In this study, the in-situ observations measured using the UGGA were also analysed for several days. The vertical profiles of the atmospheric CO_2 were measured using the UGGA during spiral and the descent of the aircraft and the results are shown in figure 15. The data recorded below 0.5 km were discarded because of sudden spikes due to slowing down of the aircraft and the associated sudden pressure changes. Figure 15 shows that the atmospheric CO_2 volume mixing ratio is largest near the ground, and it decreases gradually with the progression in the altitude. This might be due to the weak photosynthesis as the plants are in dormant stage during winter in northeast China (Mustafa et al., 2021). Moreover, northeast China is also a source of carbon due to heating and industrial activities, which also contributes significantly to the atmospheric CO_2 (Shan et al., 1997). In addition, the CO_2 concentration at different altitudes were the highest on 18 March. This could be caused by the weather conditions and pollution levels. Table 3 shows the weather report released by the Qinhuangdao meteorological station on each day of the flight.

Table 1: The weather report released by the Qinhuangdao Meteorological Department on each flight day.

Date Day Month	Weather	Temperature Highest / lowest	Wind direction/ Wind scale	AQI	PM2.5 ($\mu g/m^3$)	XCO2 (ppm)
11 March	sunny	16°C/ -3°C	Northeast/5	80	48	416.23±2.68
14 March	sunny	14°C/ -1°C	Northeast/3	60	28	414.43±1.19
16 March	cloudy	11°C/ -1°C	North/breeze	58	30	412.82±2.14
18 March	cloudy	10°C/ 4°C	Southwest/ breeze	175	131	422.59±6.39
19 March	cloudy	15°C/ 7°C	Southeast/1	139	105	415.02±3.79

The AOD values measured using various instruments on each flight day are shown Figure 16, and the results show that the AOD was the largest on 18 March. The highest CO_2 concentration on March 18 was likely caused by the higher pollution levels. A ground station was arranged in the flight area to verify the airborne results. A Micro Pulse Lidar (MPL) was installed at the Funing ground station to monitor the change of local pollutants and the boundary layer. The change of pollutants and the boundary layer in Funing ground station during the flight test on March 18 is shown in Figure 17. The dry-air mole fraction of CO_2 reaches its maximum value at about 1.4 km on March 18 (figure 15). This might be due to the fact that the height of the boundary layer was about 1.5 km on March 18 (figure 17), and the pollutants and the greenhouse gases cannot escape through the boundary layer.

Point 8: Sections 3.4 and 3.5: Please say more to Fig. 18, including the shown standard deviations. Please include the flight data in panel 18b in the same color scale, or maybe a slightly shifted scale, to consider that the satellite data must have a systematic low bias because of the influence of the altitude region with lower CO2 above the flight track (upper troposphere and the stratosphere). This bias should be mentioned in the text, here and also in section 3.5 as justification of the use of CarbonTracker (please spell out in caption of Fig.19). The last sentence of section 3.5 has to be replaced; I don't think you refer to the stratosphere here when in the figure is only the troposphere. It might be useful to indicate the flight

altitude in Fig.19.

Response 8: We are thankful to the reviewer for the valuable suggestion. We have supplemented the contents of Section 3.4 and added the results of standard deviation, including the results of adding standard deviation in Figure 18 (b). Combined with the comment of reviewer #1, we revised the last sentence of section 3.5. We highlighted the revised/modified text with red font. Relevant changes have been made in the revised version of the manuscript at:

Line 272: During this flight experiment, the OCO-2 passed over the flight area on March 16 and the observations over the study area are shown in Figure 18. The solid red line in figure 18(a) is the flight path of the aircraft. The yellow mark point is the position of the suborbital point of the OCO-2 trajectory in the flight area. Figure 18(b) shows the XCO_2 results detected by OCO-2. Figure 18(c) shows the corresponding standard deviation production of OCO-2. As can be seen from Figure 18(a), OCO-2 observations covered both ocean and land surfaces. Due to the fast flight speed of the satellite, the data time period falling in the study area was from 12:57:25 to 12:57:38 UTC. A quality flag was applied to the satellite dataset and the cloud-contaminated retrievals were removed. In the flight area, there is little difference between the values of XCO_2 measured by OCO-2 over land and ocean areas. The average value of XCO_2 over land area is 414.28 ± 0.81 ppm and that over ocean area is 414.23 ± 0.55 ppm. However, due to the uneven distribution of CO_2 volume mixing ratio in the land area, the standard deviation of XCO_2 products over the land area is larger than that over the ocean. The XCO_2 measured by OCO-2 varied from 401.66 ppm to 418.80 ppm, with an average of 414.25 ± 0.62 ppm.

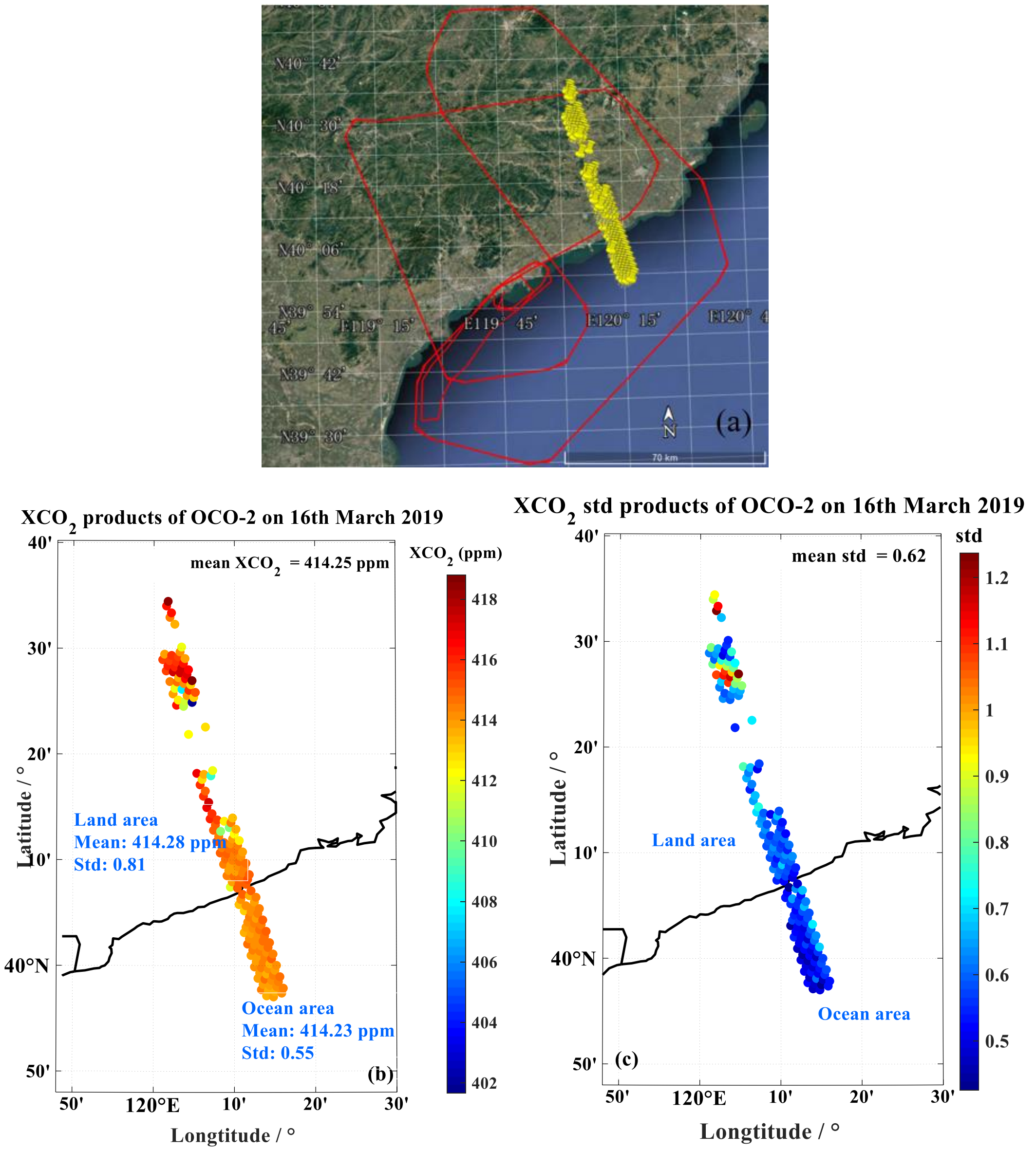

Figure 18: Orbit and detection results of OCO-2 satellite on March 16. The solid red line in figure (a) is the flight path of the aircraft. The yellow mark point is the position of the suborbital point of the OCO-2 trajectory in the flight area (© Google Earth Pro). Figure (b) shows the XCO_2 results detected by OCO-2. Figure (c) shows the corresponding standard deviation.

Line 299: This might be due to the stability of the atmosphere above.

Point 9: Line 376: Please replace the preprint by: Krol, M. C., S. Houweling, B. Bregman, M. van den Broek, A. Segers, P. van Velthoven, W. Peters, F. J. Dentener, and P. Bergamaschi (2005), The two-way nested global chemistry-transport zoom model TM5: Algorithm and applications, Atmos. Chem. Phys., 5, 417– 432.

Response 9: We are thankful to the reviewer for the valuable suggestion. We have replaced the preprint. Relevant changes have been made in the revised version of the manuscript at:

Line 412: Krol, M. C., S. Houweling, B. Bregman, M. van den Broek, A. Segers, P. van Velthoven, W. Peters, F. J. Dentener, and P. Bergamaschi (2005), The two-way nested global chemistry-transport zoom model TM5: Algorithm and applications, Atmos. Chem. Phys., 5, 417– 432.

Technical corrections

Additional to the remarks of reviewer #1 there are the following issues:

Point 10:
Line 64: Don't create fantasy names for existing institutes. The correct name is 'German Aerospace Center (DLR)'.
Line 143: Typo in citation.
Table 3: Is 'wind scale' 'wind strength in Beaufort'?
Line 266: Typo
References: Please remove control sequences (e.g. line 312) or blanks (e.g. line 449) and use subscripts instead.
Several times the name of the journal and the volume are missing, indicated by ',,', please insert it. In case of Yokota also the DOI is missing, meaning that it is impossible to find the paper. For books please provide publisher and city.
Use µ instead of mu, and CO2.

Response 10: We have revised the manuscript accordingly and we highlighted the revised/modified text with red font. Relevant changes have been made in the revised version of the manuscript at:

Line 65: In addition, the German Aerospace Center (DLR) developed a 1.57 µm double-pulse IPDA LIDAR instrument and measured the atmospheric CO_2 concentration with great accuracy during their airborne campaign in 2015 (Amediek et al., 2017).

Line 154: CarbonTracker is an inverse model framework developed by (Peters et al., 2004).

Line 449: W. Peters, M. C. Krol, E. J. Dlugokencky, F. J. Dentener, P. Bergamaschi, G. Dutton, P. v. Velthoven, J. B. Miller, L. Bruhwiler, and P. P. Tan, Toward regional-scale modeling using the two-way nested global model TM5: Characterization of transport using SF6, J. Geophys. Res., 109, D19314, doi:10.1029/2004JD005020, 2004.

Table 3: Yes professor, the 'wind scale' is 'wind strength in Beaufort'.

Line 299: This might be due to the stability of the atmosphere above.

Line 348: Kawa, S. R., Yang, M. Y. M. and DiGangi, J.: Airborne measurements of CO&amp column concentrations made with a pulsed IPDA lidar using a multiple-wavelength-locked laser and HgCdTe APD detector, Atmos. Meas. Tech., 11(4), 2001–2025, doi:10.5194/amt-11-2001-2018, 2018.

Line 485: Yokota, T., Yoshida, Y., Eguchi, N., Ota, Y., Tanaka, T., Watanabe, H. and Maksyutov, S.: Global Concentrations of CO_2 and CH 4 Retrieved from GOSAT : First Preliminary Results, , 5, 160–163, 2009.