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Interactive comment on "In situ measurement of CO_2 and CH_4 from aircraft over northeast China and comparison with OCO-2 data" *by* Xiaoyu Sun et al.

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Response to Referee comment 2

The authors thank all reviewers for their constructive comments and suggestions, which have helped us to improve the quality of this paper both in sciences and writing. All comments are carefully considered and responded.

The manuscript by X. Sun et al. describes aircraft in-situ observations of CO2 and CH4 taken over Jiansanjiang, Northeastern China, between August 7 and 10, 2018. The authors used a turboprop aircraft which was limited to 0.6-7 km flight altitude. Therefore, the profiles only covered the upper part of the planetary boundary layer (PBL) and only part of the free troposphere. In general, I greatly appreciate the efforts of taking aircraft in situ observations of CO2 and CH4 and I understand their usefulness and limitations well. However, I think the focus of the manuscript is not balanced. Due to the limited altitude coverage, the results would be most useful for validating the performance of Tan-Tracker, Carbontracker, CAMS or any other profile-based greenhouse gas data set. However, this is done only very briefly for Tan-Tracker and without much discussion about the obvious shortcomings of the model in the specific situation (active vegetation uptake of CO2 and CH4 emissions from rice fields) - especially near the surface. Instead, they spend most of the analysis and discussion on the comparison with the column-averaged OCO-2 XCO2 product - even though they correctly state that the largest error in this comparison comes from the unmeasured (extrapolated) part of their profiles. My suggestion would be to rewrite sections 5.2 and 5.3 and put more emphasis on the profile comparison. This should include a more detailed analysis how biases near the surface influence the column-averaged XCO2 and XCH4 values. Major issues: - concerning the profile to column comparison, the authors should also have a look at 1) J. Messerschmidt et al.: Calibration of TCCON column-averaged CO2 : the first aircraft campaign over European TCCON sites. Atmos. Chem. Phys., 11(21):10765– 10777, 2011. doi:10.5194/acp-11-10765-2011. 2) M. C. Geibel et al.: Calibration of column-averaged CH4 over European TCCON FTS sites with airborne in-situ measurements. Atmos. Chem. Phys., 12(18):8763– 8775, 2012. doi:10.5194/acp-12-8763-2012. Especially Geibel et al. discuss the effect of limited flight altitude on the column uncertainty due to extrapolation of the observed profiles in more detail than Wunch et al., 2010. - in Section 5.3, the authors should use the OCO-2 prior profile for extrapolating to the bottom and top of the atmosphere. The use of any other profile will create additional biases when comparing to OCO-2 data.

Thank you very much for the suggestions. We added analysis on the section 5.2 for profile comparison in our revised manuscript, Line 244, Page 8: Response: “GHGs profiles have been rarely observed before near the experiment site, or over Northeast of China as far as we know. The model simulations are based on data of regional emission inventory. So the accuracy of simulated profiles and concentration near surface over the experiment site still remains unknown. The continuous and regular observation of the GHGs profiles are necessary to better understand the regional emission amounts and the variation of the GHGs."

We refer to the articles mentioned above, and revised extrapolation method of CO2 profile in the revised manuscript. One more method is used and estimated, in the additional method, CO2 concentration at altitude with no data is replaced by OCO-2 a priori profile directly.

We corrected the sentences in section 5.3 in the revised manuscript, Line 262, Page 9: "We used two extrapolation methods to extend the profile of the aircraft measurements and then estimates the XCO2 value of the in-situ measurement respectively. 1) The unknown part of the aircraft profile was directly from the OCO-2 a prior profile. 2) A well-mixed and constant mixing ratio of CO2 is assumed from the surface to the lower limit of flight, and from the upper limit of flight to the tropopause. The CO2 concentrations above the tropopause were calculated with an empirical model (Toon and Wunch, 2014) which considers tropopause height as well as realistic latitude and time dependencies through curve fitting of data from high-altitude balloons, AirCore, Observations of the Middle Stratosphere balloon, and aircraft. In general, the mole fraction of CO2 decreased exponentially with height from the tropopause to upper stratosphere, and the tropopause height was obtained from NCEP reanalysis data with a $2.5^\circ \times 2.5^\circ$ resolution, which was linearly interpolated to the geographic coordinates of Jiansanjiang. Figure 7 (in the revised manuscript) shows the extrapolated CO2 profiles using method (2)."

And we added sentence in our revised manuscript, Line 277, Page 9: "For method (1), since the value of CO2 mole faction of unknown part is the same as that of OCO-2 a-priori profile, as eq. (5) shows, no extra uncertainty would be introduced by extrapolation."

And the following sentences were added in our revised manuscript, Line 286, Page 10: “Because of the lack of observation data near the surface, the missing measurement data was directly replaced by the data at the lowest altitude measured by the aircraft. The error caused by this practice is shown in table 3, with an average of 0.79 ppm for XCO2.This is also the impact of the lack of near-surface observations on the overall XCO2 estimates. Therefore, observations from near the surface to about 1 km from other method, such as in-situ GHG measurement by tethered balloon and high tower, is necessary for accurate estimation of XCO2.”

Minor issues: 1. p. 2, l. 41-42: it is not true that passive satellite observations of GHGs can provide all-weather, all-day global coverage.

We corrected the sentences in our revised manuscript, Line 41, Page 2: “. . . ,which can provide global coverage of the column-averaged dry-air mole fraction of CO2 (XCO2).”

2. p. 2, l. 51: the quantity X_gas as provided by TCCON as well as the satellite instruments is column-averaged dry-air mole fraction, not volume mixing ratio. Please check the definition of mole fraction vs. volume mixing ratio and replace "volume mixing ratio" throughout the text.

Thanks, We have checked the article and replaced the “mole fraction” and “volume mixing ration” to “column-averaged dry-air mole fraction” or “X_gas”.

3. p. 3, l. 75: if possible, please provide references for all 3 satellites mentioned here.

The references are added in the article. The following 4 references for, respectively, TanSAT, GMI/GF5 and GAS/FY3D are added in the article.

We corrected the sentences in our manuscript, Line 75, Page 3: “Three satellites designed for CO2 measurement, TanSAT (Yang et al., 2018; Yang et al., 2020), GMI/GF-5 (Li et al., 2016), and GAS/FY-3D (Qi et al., 2020),. . .”.

4. p. 4, l. 100: can these standard gases be referenced to the WMO GHG scale? And could you tell the nominal concentrations of CO2 and CH4 in these standards? Is

isotopic composition of the standards an issue for the aircraft measurements?

Yes, the standard gases can be tracede back to the WMO greenhouse GHG scale, which has been tested in some experiments. The concentration of the CO_2 is 400.13 ppm and CH_4 is 1.867 ppm of the standard. The standard gas we use has been measured in the laboratory for the proportion of δ13C in CO_2. The range of the proportion is -8.0‰ to -8.2‰ close to the natural content, so it will not cause significant isotopic effect on the measurement of CO_2 by optical method and meet the requirements of standard gas (Yao et al., 2013).

We added the details of the standard gas in our manuscript, Line 121, Page 4: "The standard gas we used is. . .."

5. p. 4, l. 105: should be: "Aircraft measurements were carried out ..."

We corrected this sentences in the revised manuscript as, Line 147, Page 5: "Aircraft measurement were carried out from August 7 to 10 over Jiansanjiang (47.11°N, 132.66°E, 61 m above sea level), which is located in Heilongjiang province, Northeast China. Figure 2 shows the geolocation of the Jiansanjiang aircraft and the fight path."

6. p. 4, l. 109: are the mentioned times local or UTC? Please also provide the year for the dates!

Yes, the time mentioned here is local time, GMT+8.

We corrected the sentence in our revised manuscript, Line 152, Page 5, to "Three profiles were obtained between around 08:00 and 11:00 in local time (GMT+8) on August 7, 9, and 10, 2018."

7. p. 4, l. 118: be consistent in the use of mixing ratio vs. mole fraction.

We replaced all the words "mixing ratio" to "mole fraction" to keep consistent.

8. p. 5, Eq. 3: all numbers should have units in this equation!

Thanks very much, we added the units to all the numbers in the Eq.3: Lv = 2.500×106 J Kg-1, Mw is the molecular weight of water equals to 18.016, R = 8.3145 J K-1mol-1, and es (in hPa) at temperature T (in K). We corrected the sentences in our revised manuscript, Line 177, Page 6: "Where Lv = 2.500×106 J Kg-1, Mw is the molecular weight of water equals to 18.016, R = 8.3145 J K-1mol-1, and es (in hPa) at temperature T (in K)."

9. p. 5, l. 138-151: can you derive the planetary boundary layer height from your meteorological data, e.g by calculating the Bulk-Richardson number or some other indicator?

Sorry, the meteorological obtained aircraft is limited and we did not have the actual wind speed value of the atmosphere required for calculation of the Bulk-Richardson number.

10. p. 6, l. 160: the use of the word "accurate" here is misleading. If Tan-Tracker has been validated for accuracy, please provide a number. Or just drop "accurate". Besides, the aircraft observations show that the accuracy of Tan-Tracker here is limited.

Thank you, we remove the word "accurate" from the sentence.

11. p. 8, l. 219: why is one given in ppm and the other in percent?

Sorry for the sentence is misleading and the it is corrected as: ". . . , but the values of mole fraction of CO2 from Tan-Tracker and OCO-2 had negative bias estimates. The average bias between aircraft and OCO-2 is -4.68 ± 2.02 ppm (1.18 ± 0.11%)."

12. p. 8, data availability: it would be nice if at least the 3 profiles were provided as a supplement to the paper. The amount of data should be rather small.

Yes, all the 3 profiles and relative meteorological data such as profiles of temeratureare, pressure and water vapor are available from corresponding author upon request (dmz@mail.iap.ac.cn) .

13. C3- Fig. 1: the black-and-white map of China is not very appealing. Also, a close-up of the target region, potentially as a terrain map or satellite picture would be illustrative.

Fig.1 (figure 2 in the revised manuscript)was replotted and added the flight path over the google map, we zoom in the figure focusing on the area near the experiment site.

14. - Figs. 3-5: an indication of PBL height would be useful on all these figures.

Because of the height limitation of the data, the PBL height cannot be calculated from it. So we used PBL height from reanalysis product ERA-Interim (https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era-interim) with the spatial resolution of $0.125^\circ \times 0.125^\circ$, spatially and time averaged to the flight area and time. We revised the figure 5 (figure 8 in the revised manuscript).

15. - p. 15/16, Table 2: I assume that the numbers are for CO2 but it is not actually mentioned in the table captions. If so, a similar table for CH4 would be useful. I would also appreciate an estimate of the total resulting uncertainty.

Since OCO-2 only provide CO2 products, we do not provide the table of uncertainty of estimate XCH4. But we can provide the precision and accuracy of the CH4 measurement of the aircraft. The maximum and the average value of the difference between the standard gas and the measurement of the instrument of each day was given, and represent the accuracy of the aircraft data. For the precision, we calculated the one standard deviation of the data in each level flight, and the average and maximum value of 1-σ on each day is considered as the precision of the aircraft measurement.

The numbers on table 2 are for CO2, and we added the it to the introduction of the table: “ Table 2. Aircraft integration error budget of XCO2 estimation. . .”

We added the following references to our manuscript: Li Y. F., Zhang C. M., Liu D. D., Chen J., Rong P., Zhang X. Y., Wang S. P. CO2 retrieval model and analysis in short-wave infrared spectrum. Optik, 127, 4422-4425, doi:10.1016/j.ijleo.2016.01.144,

2016. Qi C., Wu C., Hu X., Xu H., Xu H., Lee L., Zhou F., Gu M., Yang T., Shao C., Yang Z. High spectral infrared atmospheric sounder (HIRAS): system overview and on-orbit performance assessment. IEEE Trans. Geosci .Remote Sens., PP, 1-18, doi:10.1109/TGRS.2019.2963085, 2020. Yao, B., Huang, J.Q., Zhou, L.X., Fang, S.X., Liu, L.X., Xia, L.J., Li, P.C., Wang, H.Y. Preparation of mixed standards for high accuracy CO2/CH4/CO measurements. Environ. Chem., 02:135-140, doi:10.7524/j.issn.0254-6108.2013.02. 019, 2013. Yang Z., Zhen Y., Yin Z., Lin C., Bi Y., Wu Liu., Wang Q., Wang L., Gu S., Tian L. Prelaunch Radiometric Calibration of the TanSat Atmospheric Carbon Dioxide Grating Spectrometer. IEEE Trans. Geosci. Remote Sens., 56, 4225-4233, doi:10.1109/TGRS.2018.2829224, 2018. Yang Z., Bi Y., Qian W., Liu C., Gu S., Zheng Y., Lin C., Yin Z., Tian L. Inflight Performance of the TanSat Atmospheric Carbon Dioxide Grating Spectrometer. IEEE Trans. Geosci .Remote Sens., PP, 1-13, doi:10.1109/TGRS.2020.2966113, 2020. Response to Referee comment 2

Please also note the supplement to this comment:
https://www.atmos-meas-tech-discuss.net/amt-2019-363/amt-2019-363-AC2-supplement.pdf

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