We would like to thank Referee #2 for his/her thoughtful comments and detailed suggestions to our manuscript. In the following, we answer to the reviewer's comments and indicate the changes in the manuscript that were implemented as a consequence of the recommendations. The comments are in black and italic. Our answers are in blue and plain text.

Anonymous Referee #2

Received and published: 6 August 2019

This paper describes urban measurements of CO_2 by in-situ and by the novel open-path laser system "GreenLITE" with multiple reflectors and transceivers deployed in the Paris area. Observations are compared to high-resolution WRF-Chem simulations with a representation of CO_2 fluxes from anthropogenic emissions and biosphere atmosphere exchange. The paper is well written, and I recommend publishing after the following minor comments are addressed.

We thank the reviewer for his/her work and suggestions.

General comments:

For the WRF-Chem modeling CO_2 emissions at annual and national scale for scaling the high spatial resolution emissions to the year of interest have been taken from the Global Carbon Atlas (GCA), however it is unclear what these data are based on (e.g. UNFCCC reporting, BP statistical reports, or other sources). The Global Carbon Atlas has some missing links in the "Data contributors" section making traceability of the emissions impossible. This needs to be clarified.

We recognize that the sentence was insufficiently detailed. We have now modified the following statements with a supplement table (Table S1) to address the data sources and the corresponding references.

"This is accomplished by rescaling the maps with the ratio of the annual budgets of national CO₂ emissions for the countries within the domain between the base year 2005 for IER and 2010 for AirParif and the year of simulation (2015/2016), taken from Le Qu ér éet al. (2018) (<u>https://www.icos-cp.eu/GCP/2018</u>. See also Table S1 in the supplement for details about original data sources)."

Table S1. National CO₂ emissions from fossil-fuel combustion and cement production for the countries within the WRF-Chem domain used in this study (unit: MtCO₂/yr). The data in the following table are taken from Le Qu é éet al. (2018), available at <u>https://www.icos-cp.eu/GCP/2018</u>, last access: August 2019. (The use of data is conditional on citing the original data sources: data in black are from the CDIAC inventory (Boden et al., 2017), data in red are from the UNFCCC national inventory reports (UNFCCC, 2018), data in purple are from the BP Statistical Review of World Energy (BP, 2018). Cement emissions are updated from Andrews (2018))

	Austria	Belgium	France (including Monaco)	Germany	Italy (including San Marino)	Luxembourg	Netherlands	Spain	Switzerland	United Kingdom
2005	79.37	125.64	432.64	867.22	495.23	12.05	177.53	368.96	45.78	570.00
2010	72.38	113.58	397.90	833.68	424.87	11.15	182.18	283.88	45.05	512.21
2015	66.70	100.23	348.16	797.08	355.48	9.26	165.03	271.73	38.74	422.66
2016	67.40	100.24	350.10	801.75	350.32	9.00	165.52	260.99	39.20	398.55

References:

Le Qu é é C., Andrew, R. M., Friedlingstein, P., Sitch, S., Hauck, J., Pongratz, J., Pickers, P. A., Korsbakken, J. I., Peters, G. P., Canadell, J. G., Arneth, A., Arora, V. K., Barbero, L., Bastos, A., Bopp, L., Chevallier, F., Chini, L. P., Ciais, P., Doney, S. C., Gkritzalis, T., Goll, D. S., Harris, I., Haverd, V., Hoffman, F. M., Hoppema, M., Houghton, R. A., Hurtt, G., Ilyina, T., Jain, A. K., Johannessen, T., Jones, C. D., Kato, E., Keeling, R. F., Goldewijk, K. K., Landschützer, P., Lefèvre, N., Lienert, S., Liu, Z., Lombardozzi, D., Metzl, N., Munro, D. R., Nabel, J. E. M. S., Nakaoka, S., Neill, C., Olsen, A., Ono, T., Patra, P., Peregon, A., Peters, W., Peylin, P., Pfeil, B., Pierrot, D., Poulter, B., Rehder, G., Resplandy, L., Robertson, E., Rocher, M., Rödenbeck, C., Schuster, U., Schwinger, J., S d érian, R., Skjelvan, I., Steinhoff, T., Sutton, A., Tans, P. P., Tian, H., Tilbrook, B., Tubiello, F. N., van der Laan-Luijkx, I. T., van der Werf, G. R., Viovy, N., Walker, A. P., Wiltshire, A. J., Wright, R., Zaehle, S., and Zheng, B.: Global Carbon Budget 2018, Earth System Science Data, 10, 2141-2194, 2018.

Andrew, R. M.: Global CO₂ emissions from cement production, Earth System Science Data, 10, 195-217, https://doi.org/10.5194/essd-10-195-2018, 2018.

Boden, T. A., Marland, G., and Andres, R. J.: Global, Regional, and National Fossil-Fuel CO₂ Emissions, available at: http://cdiac.ornl.gov/trends/emis/overview_2014.html (last access: July 2017), Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn., USA, 2017.

BP: BP Statistical Review of World Energy June 2018, available at: https://www.bp.com/content/dam/bp/en/corporate/pdf/energy-economics/statistical-review/bp-stats-review-2018-full-report.pdf, last access: June 2018.

UNFCCC, 2018. National Inventory Submissions 2018. United Nations Framework Convention on Climate Change. Available at: http://unfccc.int/process/transparency-and-reporting/reporting-and-review-under-the-convention/greenhouse-gas-inventories-annex-i-parties/national-inventory-submissions-2018; accessed June 2018.

It is somewhat unclear how the statistics shown in Table 3 and Fig. S4 have been calculated for the GreenLITE vs. WRF-Chem measurements in section 4.1. Have the data from all chords related to e.g. T1 been combined and then the statistics is derived, or has each chord been treated independent and the resulting statistics shown in Table 3 and Fig. S4 reflect the average across all chords?

We agree with the reviewer that this point needs clarification. (PS: following the recommendation from Referee #3, we have split the previous Table 3 into Table 3 and Table S2. The previous Figure S4 is now Figure S6 in the revised manuscript)

We have added the following sentence to make it clearer:

"The statistics shown in Table 3, Table S2 and Figure S6 also indicate the ability of the models to reproduce the CO_2 at two urban in-situ stations (JUS & CDS) and the GreenLITETM measurements. As for the GreenLITETM data, we first compute the hourly averages of the observed and modeled CO_2 concentrations over all 15 chords for each transceiver (T1 and T2), and then calculate the respective statistics."

The discussion of the results in section 4.2.2 regarding the spatial gradients between different chords of the GreenLITE observations and the simulated counterparts, as well as the corresponding discrepancy between observations and model results should at least mention the potential impact of turbulent eddies and thermals. Those are likely to form in a convectively unstable atmosphere, i.e. during summer, and are unlikely to be represented properly in the MYJ PBL scheme (a local closure model) deployed in the WRF-

Chem simulations (c.f. Xiao-Ming et al., 2010). Ref.: Hu, Xiao-Ming, John W Nielsen-Gammon, and Fuqing Zhang. 2010. "Evaluation of Three Planetary Boundary Layer Schemes in the WRF Model." Journal of Applied Meteorology and Climatology 49 (9): 1831–44. doi:10.1175/2010JAMC2432.1

We greatly appreciate the reviewer's suggestion and fully agree that the vertical mixing associated with turbulent eddies and thermals plays an important role in the CO_2 transport and dispersion. The impact of insufficient vertical mixing, local eddy diffusion and entrainment flux under convective conditions reproduced by the local closure MYJ PBL scheme is a plausible explanation for the model-observation misfits. The revised manuscript has included the following discussion as suggested by the reviewer:

"Another potential source of measurement-model discrepancy is the atmospheric transport modeling as proposed in H2. According to previous studies (e.g. Hu et al., 2010), the turbulent eddies and thermals are unlikely to be reproduced properly by the local closure MYJ PBL scheme, which results in insufficient vertical mixing under convective (unstable) conditions, e.g. during summer. It may also indicate that the WRF-Chem model at a 1-km horizontal resolution cannot reproduce the fine-scale (sub-kilometer) CO₂ concentration features over a complex urban environment in Paris, as the analysis of JUS and CDS in-situ measurements has shown in Section 4.2.1."

Reference:

Hu, X. M., Nielsen-Gammon, J. W., and Zhang, F.: Evaluation of three planetary boundary layer schemes in the WRF model. Journal of Applied Meteorology and Climatology, 49(9), 1831-1844, 2010.

Specific comments:

P1 L36: I suggest replacing "have been used" with e.g. "have been or will be used" as you are referring also to future satellites.

Text changed as suggested.

P7 L21: please rephrase "low atmosphere", e.g. "lower part of the atmosphere"

Text changed as suggested. The modified text is as follows:

"It is well known that the lower part of the atmosphere is, on average, more stable in winter than in summer."

P7 L38: "in some respect superior" this should be formulated clearer. What I see from Table 3 is that RMSE with the BEP model is always better for T1 than for JUS, and better for T1 than for CDS with one exception.

To further clarify this point, the sentence has been refined as follows:

"The RMSE with the BEP scheme is within the range of 4.5 to 9.6 ppm for T1 which is substantially superior to those of JUS and CDS, with only one exception at CDS during summer when the value is slightly better for CDS than for T1."

P8 L21, P8 L37, and P10 L19: please rephrase "The std values", e.g. "Standard deviations"

Text changed as suggested.

P9 L38: What is the difference between the first two of the three hypotheses? Is H2 meant to refer to only transport model deficiencies, excluding inaccuracies in emissions? This should be made clearer. Also it should be made clear at the end of section 4.2.2 which hypothesis remains the most probable one.

Yes, Hypothesis 1 is about potential inaccuracies or uncertainties of the emission inventory for the Paris urban area, whereas Hypothesis 2 refers to the imperfect modeling of the atmospheric transport and dispersion of CO_2 over the complex urban area. We have modified the statement to make it clearer:

"• H2 The models fail in the description of CO₂ concentrations within the Paris city because of imperfect representations of atmospheric transport processes, excluding inaccuracies in emissions;"

Our analyses indicate that the model-GreenLITETM discrepancy during the summer is more likely the consequence of the measurement noise and bias in some of the chords, whereas it is hard to fully rule out the possibility of impacts of the emission spatial structure and the atmospheric transport that have been discussed in section 4.2.2. Therefore, we tend to be more cautious to make such an assessment based on our current knowledge. We have added the following sentence in the conclusion and discussion section to address this point:

"Although it is not yet fully understood, several evidences suggest an increase of measurement noise and bias in some of the GreenLITETM chords during the summer season, that must be resolved or reduced before assimilating the whole dataset into the CO_2 atmospheric inversion system that aims at retrieving urban fluxes."