We would like to thank Referee #3 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 #3**

Lian et al., present long open-path spectroscopy measurements for the City of Paris from December 2015 to November 2016 in conjunction with in-situ observations from towers in and around Paris as well as WRF-Chem simulated observations from two different urban canopy schemes. It is assumed that the authors are using the GreenLITE measurements along with the in-situ tower observations to discern which WRF-Chem urban canopy scheme can best represent vertical mixing and transport in urban areas.

My main concern with this article is that the specific objective/conclusions of the paper are unclear. The authors have conducted a lot of work analyzing data from many components, but it is uncertain as to whether they have drawn any solid conclusions.

The objective, which I assume is using GreenLITE and the in-situ observations to evaluate WRF-Chem urban canopy configurations, should be more clearly stated in the Introduction and the title of the paper should be changed.

Without a clear narrative, the paper mainly comes across as a presentation of data which is difficult to evaluate as a reader. I do not recommend that this work be accepted for publication in ACP without substantial revisions to clarify scientific objectives/conclusions.

We thank the reviewer for his/her work and comments on our paper. To answer this criticism, we have added a few sentences in the introduction and conclusion sections that make clearer the objectives of the paper and the conclusions that were derived.

Urban areas are significant sources of fossil fuel  $CO_2$  emissions.  $CO_2$  measurements in urban areas are used in conjunction with atmospheric transport models and statistical inversion techniques to estimate city  $CO_2$ emissions. The novel GreenLITE<sup>TM</sup> laser imaging system deployed in Paris provides a much wider spatial coverage of atmospheric  $CO_2$  concentrations over the complex urban environment, which makes it possible to provide new insights into the  $CO_2$  characteristics compared to the highly accurate in-situ measurements that can only be made at point locations, and can be influenced by local sources to a poorly known extent. In this paper, we analyze the measurements provided by this novel system, together with the more classical in-situ sampling and high-resolution modeling and we focus on the temporal and spatial variability of atmospheric  $CO_2$  concentrations. The main purpose of the paper is therefore an evaluation of the new system capabilities to provide information on both the emission and the atmospheric transport, typically whether the new system can falsify the emission inventory used or point out to transport modeling deficiencies (the two hypotheses formulated in the paper).

We have added the following paragraph in the introduction to better clarify the objectives of this work:

"The detailed objectives of this paper are:

- To analyze in detail the information content of the GreenLITE<sup>™</sup> data in addition to conventional in-situ CO<sub>2</sub> measurements in order to better understand the temporal and spatial variations of near-surface CO<sub>2</sub> concentrations over Paris and its vicinity.

- To evaluate the performance of the high-resolution WRF-Chem model coupled with two urban canopy schemes (UCM, BEP) for the transport of CO<sub>2</sub> over the Paris megacity area based on the two types of CO<sub>2</sub> measurements.
- To discuss the potential implications of assimilating the GreenLITE<sup>™</sup> data into the CO<sub>2</sub> atmospheric inversion systems with the ultimate goal of increasing the robustness of the quantification of city emissions and constraining the spatial distribution of the emissions within the urban area."

The main conclusions of this study are:

- Two urban canopy schemes (UCM, BEP) as part of the WRF-Chem model are capable of reproducing the seasonal cycle and most of the synoptic variations in the atmospheric CO<sub>2</sub> in-situ measurements over the suburban areas, as well as the general corresponding spatial differences in CO<sub>2</sub> concentration between pairs of in-situ stations that span the urban area.
- The GreenLITE<sup>TM</sup> measurements are less sensitive to local unresolved sources than the in-situ point measurements, and are then better suited for the comparison to km-scale modeling. In our analysis, the GreenLITE<sup>TM</sup> data have been used to show a deficiency of the UCM scheme during the winter, linked to underestimated vertical mixing. Conversely, the model-GreenLITE<sup>TM</sup> discrepancy that is observed during the summer is not yet fully understood. Several evidences suggest an increase of measurement noise and bias in some of the GreenLITE<sup>TM</sup> chords during the summer season, that must be resolved or reduced before assimilating the whole dataset into the CO<sub>2</sub> atmospheric inversion system that aims at retrieving urban fluxes.
- Within the city, the misfit between the observed and simulated CO<sub>2</sub> concentrations is found to be highly sensitive to the WRF-Chem configuration for the urban canopy scheme, which affects the atmospheric vertical mixing. We also show that the CO<sub>2</sub> concentrations are impacted by the spatial distribution of the emission and the presence of local sources that are poorly resolved in the inventory. This study stresses the difficulty in reproducing precisely the atmospheric CO<sub>2</sub> concentration within the city because of our inability to represent the detailed spatial structure of the emission and because of the sensitivity of the concentration to the strength of vertical mixing. From the model results analysis, we infer that the uncertainty on the vertical mixing is much larger than the uncertainty on the emissions so that atmospheric concentration measurements within the city can hardly be used to constrain the emission inventories.

We have modified the conclusion and discussion section based on the main conclusions listed above.

I have the following suggestions regarding the technical components of the analysis.

## Main Comments:

(1) The observational network: can you provide some indication as to areas in which the observations are most sensitive especially CDS, JUS, and any others that are situated in or adjacent to major sources/sinks? I understand that the authors use WRFCHEM and not a Lagrangian approach so that footprints cannot be generated but having some understanding would help the comparison of GreenLITE and in-situ observations presented later in the paper since these two different types of measurements represent different spatial extents.

The footprint of the measurement station very much depends on the wind speed and direction, as well as the atmospheric stability. It is then difficult to interpret a mean footprint that aggregates a wide range of atmospheric situations. In response to the reviewer's concern about the major sources of emissions to the station, we carried out some sensitivity experiments for the one-month period of March 2016 with

anthropogenic and biogenic emissions limited to a given area within the simulation domain in order to quantify their respective contributions to the simulated CO<sub>2</sub> concentrations at a certain measurement site. This set of experiments includes the assignment of emissions to: 1) ONE: one grid cell that contains the insitu station, 2) GRP: all grid cells within the GReater Paris except the one where the station is located, 3) IDF: all grid cells within the IdF region except those of the Greater Paris, 4) OUT: all grid cells outside the IdF region, as shown in Figure S10 (a) (b) (c) (d) respectively. The contribution from sources outside the model domain is small enough so that its influence is negligible. Figure S11 shows the relative contributions (in percentages) of each component to the modeled total anthropogenic and biogenic CO<sub>2</sub> concentrations for one urban site JUS and one suburban site COU respectively. The simulated monthly mean concentrations of anthropogenic CO<sub>2</sub> are 11.0 ppm at JUS and 5.4 ppm at COU, which are much larger than those of biogenic CO<sub>2</sub> (0.6 ppm at JUS and 0.7 ppm at COU). In general, an urban station like JUS is under a strong influence of the anthropogenic emissions within the IdF region. The contributions of anthropogenic emissions in the vicinity of the station (ONE) and from the Greater Paris (GRP) areas to the simulated anthropogenic CO<sub>2</sub> concentrations are around 16% and 60% respectively, whereas the remote anthropogenic emissions account for less than 20%. For a suburban station like COU, the Parisian emissions (GRP) and the remote ones (OUT) have a comparable influence (~40%) on the simulated anthropogenic concentrations, with very large variations depending on the wind direction (downwind or upwind of the city). The biogenic  $CO_2$  concentrations mainly come from outside of the IdF region (~86%).

We have added the following statement in the main body of the revised manuscript and two figures (Figure S10 and S11) in the supplement based on the discussion above:

"Atmospheric transport simulations make it possible to assess the respective contributions of various areas/sectors to the measurements. Our preliminary sensitivity experiments (see Figure S10 and S11 for details) have shown that the anthropogenic emission from the Greater Paris area is the dominant contribution (~80%) to the anthropogenic CO<sub>2</sub> signal at the urban measurement stations. In order to get further insights into the characteristics of CO<sub>2</sub> spatial variations within the Paris city, it is therefore necessary to analyze the CO<sub>2</sub> differences with the consideration of the anthropogenic CO<sub>2</sub> emissions shown in Figure 2 and Figure 3."

For better clarity, we have also added the following analyses in the supplement together with Figure S10 and S11.

"In order to determine respective contributions of various areas/sectors to the simulated  $CO_2$  concentrations at a certain measurement site, we carried out a set of sensitivity experiments for the one-month period of March 2016 with anthropogenic and biogenic emissions limited to a given area. This set of experiments includes the assignment of emissions to: 1) ONE: one grid cell that contains an in-situ station, 2) GRP: all grid cells within the GReater Paris except the one where the station is located, 3) IDF: all grid cells within the IdF region except those of the Greater Paris, 4) OUT: all grid cells outside the IdF region, as shown in Figure S10 (a) (b) (c) (d) respectively.

Figure S11 shows the relative contributions (in percentages) of each component to the modeled total anthropogenic and biogenic  $CO_2$  concentrations for one urban site JUS and one suburban site COU respectively. The simulated monthly mean concentrations of anthropogenic  $CO_2$  are 11.0 ppm at JUS and 5.4 ppm at COU, which are much larger than those of biogenic  $CO_2$  (0.6 ppm at JUS and 0.7 ppm at COU). In general, an urban station like JUS is under a strong influence of the anthropogenic emissions within the IdF region. The contributions of anthropogenic emissions in the vicinity of the station (ONE) and from the Greater Paris (GRP) areas to the simulated anthropogenic  $CO_2$  concentrations are around 16% and 60% respectively, whereas the remote anthropogenic emissions account for less than 20%. For a suburban station

like COU, the Parisian emissions (GRP) and the remote ones (OUT) have a comparable influence (~40%) on the simulated anthropogenic concentrations, with very large variations depending on the wind direction (downwind or upwind of the city). Note that in these experiments, the emission inventory and the WRF-Chem modeling cannot describe the CO<sub>2</sub> patterns (both emission and concentration) at a scale finer than 1 km, and the simulation shows that the "local" contribution is significant. The unresolved spatial distribution of the emission can therefore be a significant contribution to the uncertainty. The biogenic CO<sub>2</sub> concentrations mainly come from outside of the IdF region (~86%)."



Figure S10. Four experiments are carried out for the JUS station with the assignment of emissions to: (a) ONE: one grid cell that contains an in-situ station; (b) GRP: all grid cells within the Greater Paris except the one where the station is located; (c) IDF: all grid cells within the IdF region except those of the Greater Paris; (d) OUT: all grid cells outside the IdF region. Another four experiments are carried out for the COU station.



Figure S11. Relative contributions (in percentages) of each component flux to the modeled total anthropogenic and biogenic CO<sub>2</sub> concentrations for (a) urban site JUS and (b) suburban site COU. Note that only the afternoon data (11-16 UTC) are used in the analysis.

# (2) GreenLITE campaign – can you provide more description than citing the Zaccheo paper as to how the GreenLITE observations are calibrated?

As mentioned by the reviewer, the calibration method is extensively described in Zaccheo et al. (2019). Nevertheless, we have followed the suggestion and now provide a better description (although very much summarized) of the procedure:

"These slowly time-varying differences were most likely due to a slight systematic long-term drift in both the on- and off-line wavelengths as a function of continuous operations. Such drift may induce some nonlinear impacts on the measured concentrations. It is therefore more appropriate to adjust the wavelengths rather than to apply a linear calibration to the retrieved concentrations. Unlike in-situ point measurement systems, there is no established method for calibration of long open-path systems to the WMO mole fraction scale used as an international standard for atmospheric  $CO_2$  monitoring (Tans et al., 2011). Therefore, a bias correction method was developed by AER (Zaccheo et al., 2019) for addressing observed slowly drifting biases between the GreenLITE<sup>TM</sup> prototype system and the two in-situ sensors (CDS and JUS) that are near the GreenLITE<sup>TM</sup> chords. This method computed a time-varying adjustment to the offline wavelength based on a non-linear optimization mechanism. This non-linear approach adjusts the GreenLITE<sup>TM</sup> offline wavelength considering not only the average values of hourly CO<sub>2</sub> concentrations at two in-situ stations, but also the corresponding average temperature, relative humidity, atmospheric pressure along the chord and an optimized online wavelength value during the measurement period. Finally, the median on- and off-line values over a 4-day window was used to recompute the GreenLITE<sup>™</sup> data from all chords using a radiative transfer based iterative retrieval scheme based on the LBLRTM model (Clough et al., 2005). Even though this approach is not ideal as the two in-situ stations and the GreenLITE<sup>TM</sup> system do not sample the exact same area, it does provide a well-defined mechanism that reduces the systematic long-term biases with no significant impact on the chord-to-chord variations."

A general response to comments (3) (4) and (5):

We fully agree with the reviewer that the uncertainties associated with atmospheric transport, anthropogenic emissions, biogenic fluxes and background conditions could all have a more or less impact on the model performance. Over the years, many studies have been carried out at different scales and regions on the analysis, derivation and quantification of critical sources of uncertainties that lead to the model-observation misfits. Nevertheless, even with a state-of-art atmospheric transport model and an inventory at a high spatio-temporal resolution, the uncertainties associated with the modeling are inevitable and cannot be completely eliminated. The ensemble-based sensitivity study and a full analysis on the uncertainties derived from different anthropogenic inventories, biogenic fluxes, atmospheric transports or background conditions are out of the scope of this study that focuses on the potential contribution of GreenLITE<sup>TM</sup> observing system, but will be specifically addressed within another dedicated study.

(3) WRF-Chem – Is this paper an analysis of WRF-Chem urban canopy models for cities like Paris (evaluated using GreenLITE and in-situ observations)? If so, please substantiate/provide reference for the claim that "previous sensitivity tests indicate that different physical schemes in the WRF-Chem model lead to mean differences of 2-3ppm on the simulated  $CO_2$  concentrations over Paris, whereas the various urban canopy schemes lead to much larger differences." This seems like the motivation for much of the work presented within the paper, but I am not sure that this claim, if substantiated, holds true across most urban areas.

As our answers to the general comment, our main objective is not fully to test the modeled CO<sub>2</sub> sensitivity to the use of different physical schemes and to discern which urban canopy scheme could reach best results when comparing the model to observations. Several studies have demonstrated that the city-scale physical and dynamical processes in the atmospheric modeling system remain a challenge. In order to select an adequate WRF-Chem model configuration for Paris, we did perform some preliminary sensitivity experiments to test the impact of different physical schemes (5 PBL schemes + UCM, 2 PBL schemes + BEP) on the simulated CO<sub>2</sub> concentrations. The simulations were carried out for two months, including one winter month (January 2016) and one summer month (July 2016). These preliminary sensitivity results indicate that different PBL schemes in the WRF-Chem model lead to monthly average differences of 2-3ppm on the simulated CO<sub>2</sub> concentrations over Paris, whereas the urban canopy schemes lead to much larger differences of 8-10 ppm. We thus carried out the 1-year simulation with two different urban canopy schemes as they are sufficient to address the paper main question regarding the ability of a configuration of the WRF-Chem model to simulate the CO<sub>2</sub> atmospheric transport in an urban environment, but also to provide an estimate of the modeling uncertainty.

We have added the following sentence in the manuscript to account for the reviewer's comment:

"In order to select an adequate model physical configuration for Paris, we carried out some preliminary sensitivity experiments to test the impact of different physical schemes on the simulated  $CO_2$  concentrations. These tests use up to five different PBL schemes and two urban canopy schemes. The simulations were carried out for two months, including one winter month (January 2016) and one summer month (July 2016). These preliminary sensitivity results indicate that different PBL schemes in the WRF-Chem model lead to monthly average differences of 2-3 ppm on the simulated  $CO_2$  concentrations over Paris, whereas the two different urban canopy schemes lead to much larger differences of 8-10 ppm. Thus in this study, we carried out the 1-year simulation with two different urban canopy schemes as they are sufficient to address the paper main question regarding the ability of a configuration of the WRF-Chem model to simulate the  $CO_2$  atmospheric transport in an urban environment, but also to provide an estimate of the modeling uncertainty. All of the other physics options remained the same for the two experiments (Table 2)."

a. If this is not the focus, and the purpose is to use the meteorology to understand the variability of the measurements, then I believe the authors should pick a model and use it throughout the rest of the analysis. It seems (from Figure 5) that the BEP model is largely better. The rest of the analysis using UCM could be put in the supplementary information. As an aside, I do think that the authors could use the ensembles in a way that would help them draw some robust conclusions. Their ensembles provide some measure of the atmospheric transport and dispersion uncertainty which can be used to contextualize their comparison between GreenLITE and the in-situ observations (S1 and S4).

The main purpose of the study is to assess the potential contribution of the GreenLITE<sup>TM</sup> system, in addition to the more classical in-situ sampling, for a better understanding of the temporal and spatial variations of near-surface CO<sub>2</sub> concentrations over Paris and its vicinity (due to emissions / atmospheric transport). Even though the two urban canopy schemes do not represent the full range of uncertainty in the atmospheric transport, in some extent they do provide an insight into the critical impact of the atmospheric transport on simulated atmospheric CO<sub>2</sub> concentrations. It thus appears to us that it is necessary to keep the UCM analysis within the main body of the paper. We also tested different PBL schemes for the impact of the atmospheric transport, but decided to show results for the two urban canopy schemes because they caused the largest differences in simulated CO<sub>2</sub> concentrations.

(4) Anthropogenic Fluxes – The use of the anthropogenic fluxes in the analysis should be reconsidered or better explained. For example, does IER have any temporal variability? If so, please explain. If not, the authors could consider scaling using published methods. The authors could use other emission products that have temporal variability if needed. The loss of spatial scale (e.g. going from 5-10km) seems less important than preserving some temporal structure in emissions. Other products are also more recent and thus more represented of ex-urban fluxes which constitute a large portion of  $CO_2$  inflow to Paris.

Yes, the IER inventory used in this study has a detailed country-specific temporal profiles (monthly, daily and hourly) at spatiotemporal resolutions of 5 km and 1 h. Given the fact that it has a higher spatial resolution than some other emission products and it has been rescaled to account for annual changes in emission between the base year and simulation timeframe, this inventory is sufficient to be used in this study.

We have added the information about the IER temporal variability in the revised manuscript:

"CO<sub>2</sub> emissions from fossil fuel CO<sub>2</sub> sources outside the IdF region are taken from the inventory of the European greenhouse gas emissions, together with country-specific temporal profiles (monthly, daily and hourly) at a spatial resolution of 5 km (updated in October 2005). This inventory was developed by the Institute of Economics and the Rational Use of Energy (IER), University of Stuttgart, under the CarboEurope-IP project (<u>http://www.carboeurope.org/</u>)."

Could the authors also further explain "we interpolate the emissions to the WRF-Chem grids following the principle of mass conservation?" This is unclear in both its meaning and why it is important.

The total magnitude of anthropogenic emissions should be consistent before and after the interpolation to model grid cells. We have modified the statement to make it clearer:

"Finally, we interpolate the emissions onto the WRF-Chem grids, making sure to conserve the total budget of emission in the process, as done in previous studies (e.g. Ahmadov et al., 2007)."

As with the WRF-Chem comments, the authors could use an ensemble of anthropogenic emission products (those outside of Paris) to help contextualize the GreenLITE and in-situ observations in terms of emission uncertainty (refer to Martin et al., 2018).

Please see the analysis above. The contribution of anthropogenic emissions outside the IdF region to the simulated anthropogenic  $CO_2$  concentrations over urban areas is relatively small (~20%), and our present simulations are capable of reproducing the seasonal cycle and most of the synoptic variations in the atmospheric  $CO_2$  point measurements over the suburban areas. In addition, this distant contribution is much smoother, both in temporal and spatial scales, than the impact of more local emissions. We then do not expect a significant impact of these distant emissions on the  $CO_2$  signatures that are analyzed in the paper. We then do not feel that an ensemble of anthropogenic emission products outside the IdF region will bring critical insights on the main conclusions at two urban in-situ stations (JUS & CDS) and the GreenLITE<sup>TM</sup> measurements. A deeper analysis of the impact of uncertainties in the anthropogenic emissions outside the IdF region so utside the IdF region so utside the IdF region so utside the IdF region so the main conclusions at two urban in-situ stations (JUS & CDS) and the GreenLITE<sup>TM</sup> measurements. A deeper analysis of the impact of uncertainties in the anthropogenic emissions outside the IdF region so utside the IdF region is out of the scope of the paper.

(5) Biogenic Fluxes – The use of VPRM to represent the urban biosphere is an active area of research and there are lots of questions as to how well a biospheric model captures the urban biogenic emissions. When VPRM was optimized using flux data, were urban towers used to help parameterize the "urban" areas of Paris? The paper mentions that the western portion of Paris has much green space and thus biogenic sources might be important in this area of the city and impact the analysis. How was Paris-VPRM (or VPRM) validated, e.g. comparison to in-situ data from towers outside of the city that are surrounded by vegetation (maybe OVS)? Has it been used in other studies? How does it vary as a function of time in comparison to the anthropogenic fluxes like what is shown in Figure 3?

As for the urban biogenic emissions mentioned by the reviewer, we certainly agree that it might be important for the simulated  $CO_2$  concentrations and it is an active area of research. To which extent the biogenic fluxes affect the simulated  $CO_2$  concentrations in the Paris urban areas remains an open question. Whereas there is no eddy covariance measurement in the Paris urban area that is available for the biospheric flux optimization and we are not able to make an evaluation of the Paris-VPRM model in this study. Nevertheless, we have performed some further analyses and validations of the VPRM model at a suburban station at SAC in a dedicated study mentioned above. Since these analyses at SAC do not reflect the model performance of the biosphere mode in the urban area, it is out of the scope of this study. Mean diurnal cycles of  $CO_2$  biogenic flux (NEE) for 12 calendar months and for 8 vegetation classes used in VPRM over Domain 03 are shown in Figure 4 and the related texts are in Section 3.2.2.

(6) Results – (4.1) There are a lot of moving pieces in this analysis and it is hard to ascertain the main conclusions from the statistical analysis. Do you think that the uncertainties associated with the other components (e.g. anthropogenic emissions and vprm sources and sinks) would have changed some of these results especially during the growing seasons or per your analysis of the seasonality of the sectors? From the Table, it is unclear that BEP outperforms UCM for much of the year. As with 4.1, I am not sure what the main takeaway is from this analysis.

In fact, each paragraph in Section 4.1 relates to a certain aspect regarding the statistics for observed and modeled CO<sub>2</sub> concentrations for periods of the day (all hourly data, hourly afternoon data) and two urban canopy schemes (UCM, BEP). In general, the model performance is better during the afternoon than it is for the full day. UCM and BEP have different performances for four seasons and for urban/suburban areas (see answers below, this is also the main takeaway). The statistics further confirm the fact that the GreenLITE<sup>TM</sup> measurements represent an average over a wide area, and are then less sensitive to local unresolved sources than the in-situ measurements.

We do not know to what extent the uncertainties associated with the other components (e.g. anthropogenic emissions and VPRM sources and sinks) would have changed some of these results since we have not made the relevant sensitivity experiments. A full analysis of these uncertainties would be a paper by itself.

In the third paragraph of Section 4.1, we have already discussed the different performances of UCM and BEP for four seasons and for urban/suburban areas with the following statements: 1) The statistics for BEP compared to the observations within the urban areas are significantly better than UCM during autumn and winter; 2)  $CO_2$  concentrations are better reproduced by both UCM and BEP in the spring; 3) Both models show lower correlations during summer; 4) the UCM and BEP also have comparable performances at periurban areas while the BEP is slightly better at some suburban sites as shown by the statistics.

(4.2.1) Why did you use the wind per ECMWF versus wind measurements at the upwind tower(s)? I am sure, on average, the ECMWF winds are similar to what is measured at the towers but since you are comparing hourly measurements, this may make a difference.

The ECMWF wind product is used here for 2 reasons: Firstly, our previous study has shown that the wind speeds provided by the ECMWF high-resolution operational forecasts (HRES) are, in general, closer to the observations than those provided by WRF (Lian et al., 2018). Secondly, the WRF model was run with two configurations (UCM and BEP urban canopy schemes) in this study. If we make use of the modeled winds, the UCM and BEP modeled  $CO_2$  spatial differences should be analyzed using their corresponding modeled wind fields, and the observed winds are then needed for the analysis of the observed  $CO_2$  spatial differences. However, given the small-scale wind variations reproduced by the model, it is hard to determine that the wind data at which station should be used in the analysis. For the purpose of a fair and uniform comparison, we thus use an independent wind product. The HRES with a horizontal resolution of about 16 km could provide a synoptic wind pattern as a proxy for all stations located within the IdF region. We have added the following sentence in the manuscript to account for the reviewer's comment:

"The HRES wind product is used here for two reasons: Firstly, our previous study has shown that the wind speeds provided by HRES are, in general, closer to the observations than those provided by WRF (Lian et al., 2018). Secondly, the WRF-Chem model was run with two configurations (UCM and BEP urban canopy schemes) in this study. If we make use of the modeled winds, the UCM and BEP modeled  $CO_2$  spatial differences should be analyzed using their corresponding modeled wind fields, and the observed winds are then needed for the analysis of the observed  $CO_2$  spatial differences. However, given the small-scale wind variations reproduced by the model, it is hard to determine that the wind data at which station should be used in the analysis. For the purpose of a fair and uniform comparison, we thus use an independent wind product."

# Also, how much time does it take to traverse some of the towers that are farther apart (e.g. COU and SAC)? Did you compare observations from similar times or did you account for a lag in the measurements via travel time?

We ignore the time lag needed to transport information from upwind to downwind sites spanning the city by computing spatial gradients between concentrations at a given time. This is mainly due to the fact that the consideration such a time lag might be somewhat meaningless given the wind shear in the PBL during the afternoon when the mixing layer is usually well developed. Typical wind speed over Paris at 700 m above ground level is 7 m/s (25 km/h) and the distance between COU and SAC is approximately 38 km so that air masses take, on average, less than 2 hours to travel between the two sites at this height. Conversely, the wind speed at ground level is much smaller so that there is not a single time-lag that can be used. We thus assume that the analysis that is based on  $CO_2$  concentration differences measured during the same 1hour window is a minor issue. Note that, when comparing observation and models, the time lags are consistent.

### Minor Comments:

Be specific as to what model you are using. I think in most cases you are referring to WRF-Chem models but there are others too such as VPRM, etc.

Following the reviewer's recommendation, we have attempted to make it clear and specific.

Grammar should be checked in many places throughout the article to improve clarity. Examples include lines 34 through 36 (page 2),  $\sim 10$  (page 8).

As suggested, we have carefully done thorough English editing and corrected the grammatical mistakes in the revised manuscript.

Figures should be modified to improve clarity:

For example, Figure 1 should include a depiction of adjacent urban areas to show how remote AND, COU, OVS, and SAC are from ex-urban sources. This will help the reader know whether or not they sample "clean" air.

We feel that there is no need to add a depiction in Figure 1 for 3 reasons: 1) the distributions of the 1-km anthropogenic  $CO_2$  emissions together with all in-situ measurement stations are shown in Figure 2a. 2) the dominant land use categories together with all in-situ measurement stations are shown in Figure S5. These two figures could be sufficient to provide such a depiction. 3) As we have mentioned in Section 2.1, the insitu stations are installed on the rooftops or on towers to minimize the impact of local surface emissions. Moreover, the distance to the localized emissions was also taken into account as a necessary aspect in the design of this  $CO_2$  monitoring network to ensure they sample air that is not in the immediate proximity of large anthropogenic emissions.

Figure 2 should include roads and other infrastructure in the second panel especially since the authors have made spent time discussing sectorial emissions. Also note (a) and (b) on Figure 2.

This suggestion is well taken. We have added these infrastructures into a second panel in Figure 1 where it might be more appropriate.

As suggested, we have noted (a) and (b) on Figure 2.

For Figure 4, zoom into similar area as in Figure 2 to show if VPRM is capturing urban biospheric flux which can significantly impact the urban fluxes especially their variability.

Figure R1 a high-resolution zoom of Paris and shows the daytime (06-18 UTC) average of  $CO_2$  biogenic flux (NEE) in June 2016. Due to the 1-km SYNMAP land use data used for the VPRM model, the biogenic fluxes in Paris are almost zero except for a few grid cells containing two big parks that are located in the eastern and western Paris.

We thus feel that there is no need to make this high-resolution zoom of the GreenLITE<sup>TM</sup> covering areas since we have already mentioned it in Section 3.2.2 "The model simulates negative values of NEE (uptake of more than 5 gCO<sub>2</sub>/m<sup>2</sup>/day) over most of the region with the exception in urban areas where the values are assigned to zero."



Figure R1. Daytime (06-18 UTC) average of CO<sub>2</sub> biogenic flux (NEE) over Paris in June 2016

The authors should consider moving Figure 5 to the supplemental information. It doesn't provide much information, especially given the timeframe that makes it hard to see, expect to show that the UCM transport model yields extreme outliers in the winter.

Following the reviewer's recommendation, we have moved Figure 5 to the supplement as Figure S7.

For Table 3, explain by what criteria did you color code the Tables. It seems like the better models for the correlation coefficients have "red" shading where int the RMSE and MBE the colors are switched (aka blue is better while red is worse). I would remove "all hours" to make the table clearer - all hours not really needed.

We agree with the reviewer that the color scales in Table 3 can be misleading. The color only represents the values from minimum (blue) to maximum (red) in the cells instead of indicating the goodness of fit between model and observation. We have added the following text in the caption of Table 3 in order to clarify this issue:

"The color highlights the value in the cell with the minimum in blue, the median in white and the maximum in red. All other cells are colored proportionally."

Following the reviewer's recommendation, we have removed "all hours" to make Table 3 clearer and put it in the supplement as Table S2.