General Response: We greatly appreciate the referee for his/her time and efforts devoted to the review of our submission. The major comment was the manuscript didn't provide details about the model configuration and tower measurement. The reviewer also questioned the arrangement and completeness of the manuscript, for instance, readers do not know the locations of the Lin'an tower station and Hangzhou thus they cannot catch up the further discussion. We realize that most of the comments are due to the missing of necessary details regarding the modeling method, the observational method, and the study domain. We will present these details in this document as shown in the following responses. The reviewer suggested that the 20km grid resolution simulation may not be directly compared with local-scale flux measurement, thus we conduct a new set of simulation at 4km grid resolution over a smaller domain covering the tower measurement site, and revise the manuscript accordingly. In addition, some of the sub-figures in the original submission have been rearranged and drawn separately, we will show these figures as well.

Specific comments and responses:

Comment#1: Authors never reported model configurations, specifically for meteorology, although one of the key works for this study is a numerical simulation. For the publication of modeling work, the basic model configuration for both CO_2 and meteorology is essential for the scientific community's numerical experiments' reproducibility

Response: The model configuration in this study mostly follow the work of Hu et al. (2020), except that Hu et al. (2020) simulated North America but our simulation is over East Asia. Hu et al. (2020) is frequently cited in our manuscript but we forget to mention about the configuration. We apologize for this careless mistake. As a coupled weather-biosphere model, the WRF-VPRM simulation contained two parts of configuration for WRF and VPRM respectively. The configuration on the WRF side is presented in the following table (Table S1 in revised manuscript).

Table: WRF-VPRM Model Configuration

Attribute	Configuration	Reference
Short wave radiation	Duhia algorithm	Dudhia (1989)
Long wave radiation	Rapid radiative transfer model (RRTM)	Mlawer et al. (1997)
Boundary layer	Yonsei University (YSU) scheme	-
Microphysics	Morrison scheme	Morrison et al. (2009)
Cumulus	Grell-3 scheme	Grell and Devenyi (2002)
Land surface model	Noah land-surface scheme	Chen and Dudhia (2001)
Vertical levels	47	-
Horizontal resolution	20 km × 20 km with 234 (south-north) × 285 (west-east) grid points; 4km × 4km with 215 (south-north) × 280 (west-east) grid points	-

Time step	60s	-
Meteorological initial and lateral boundary conditions	NCEP/DOE Reanalysis 2 (R2)	-
Interior nudging	Spectral nudging	-
Nudging variables	horizontal wind components, temperature, and geopotential height	-
Nudging coefficient	3×10^{-5} s^{-1}	-
Nudging height	above PBL	-
Wave number	5 and 3 in the zonal and meridional directions, respectively	-

The configuration on the VPRM side refers to emission inputs, initial and boundary conditions, and the parameterization (for $PAR_0, \alpha, \beta, \lambda$). We have described emission inputs and initial and boundary conditions in the manuscript at line#91-97. Physical parameterization followed the default configuration as mentioned at line#112. The values of the default parameterization are presented in the following table (Table S2 in revised manuscript).

Table: VPRM Parameter Values Used in This Study

	evergreen forest	Deciduous forest	Mixed forest	Shrub	Savanna	Crop	Grass
PAR_0 (μmol PAR·m^{-2}·s^{-1})	745.306	514.13	419.5	590.7	600	1074.9	717.1
λ (μmol CO_2·$m^{-2}s^{-1}$/μmol PAR·m^{-2}·s^{-1})	0.13	0.1	0.1	0.18	0.18	0.085	0.115
α (μmol CO_2·m^{-2}·s^{-1}·°C^{-1})	0.1247	0.092	0.2	0.0634	0.2	0.13	0.0515
β (μmol CO_2·$m^{-2}s^{-1}$)	0.2496	0.843	0.27248	0.2684	0.3376	0.542	-0.0986

The above tables are included in the “supplement information” of the revised manuscript. We also add necessary description in the main text of the revised manuscript.

We conduct a new set of simulation with 4km grid resolution with exactly the same configuration over a smaller domain, as shown in the following figure (Figure 1(b) and (d) in revised manuscript). The new 4km-grid simulation has a domain size as 215 (south-north) × 280 (west-east) grid points over a significantly smaller domain than the 20km-grid simulation domain. The 4km-grid simulation showed very similar result to the 20km-grid simulation. Thus the 20km-grid simulation was used to characterize the spatiotemporal distributions of CO_2 over China, and the 4km-grid simulation was only used to compare with tower data collected at Lin’an tower. Detailed comparison will be shown in the response for comment#3. Our major conclusion was not changed, thus we do not attempt to rerun the whole China domain simulation with 4km grid resolution due to limited computational resource.

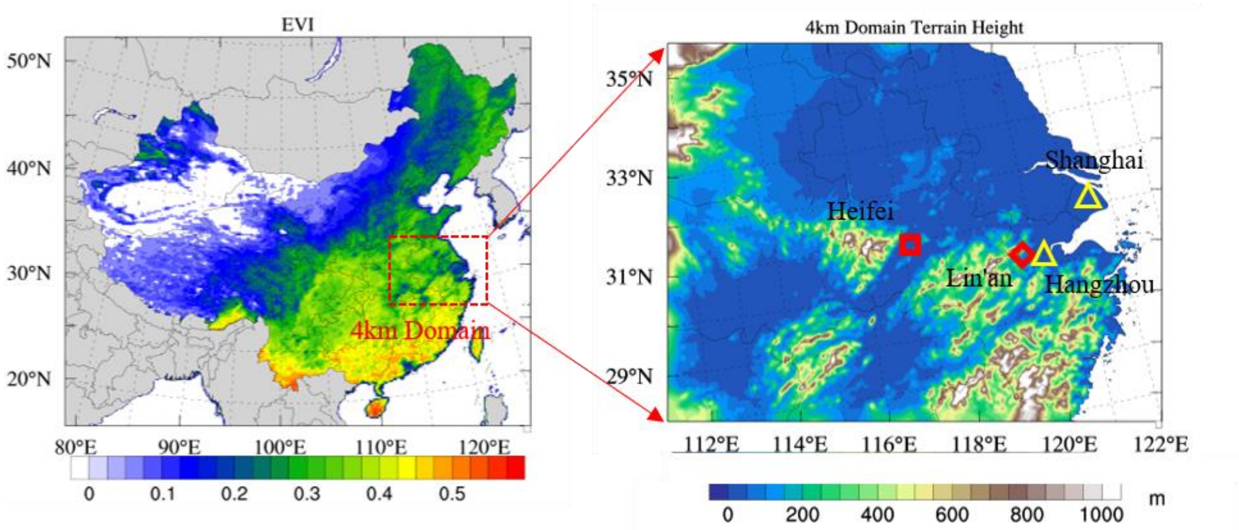

Figure: 4km-grid simulation domain over Yangtze River Delta (YRD).

Reference:

Chen, F., & Dudhia, J. (2001). Coupling an advanced land surface-hydrology model with the Penn State - NCAR MM5 modeling system. Part I: Model implementation and sensitivity. Monthly Weather Review, 129(4), 569–585.

Dudhia, J. (1989). Numerical study of convection observed during the Winter Monsoon Experiment using a mesoscale two-dimensional model. Journal of the Atmospheric Sciences, 46(20), 3077–3107.

Grell, G. A., & Devenyi, D. (2002). A generalized approach to parameterizing convection combining ensemble and data assimilation techniques. Geophysical Research Letters, 29(14), 1693. https://doi.org/10.1029/2002gl015311

Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., & Clough, S. A. (1997). Radiative transfer for inhomogeneous atmospheres: RRTM, a validated correlated-k model for the longwave. Journal of Geophysical Research - Atmospheres, 102(D14), 16663–16682. https://doi.org/10.1029/97jd00237

Morrison, H., Thompson, G., & Tatarskii, V. (2009). Impact of cloud microphysics on the development of trailing stratiform precipitation in a simulated squall line: Comparison of one- and two-moment schemes. Monthly Weather Review, 137(3), 991–1007.

Comment#2: The model configuration is also significant for understanding the analysis of CO_2 concentrations near the ground, interacting with the PBL and stable boundary layer (Section 3.3). Without prior knowledge about the model set-up, readers cannot be sure of the quality assurance of simulation results. For the analysis of CO_2 concentration near the ground, especially in nighttime, the PBL scheme's choice is significant. As the "first" WRF-VPRM simulation over the study domain, authors must conduct the PBL scheme sensitivity tests and find the best physics scheme combinations before progressing this manuscript.

Response: We totally agree with the comment that PBL scheme plays a very important role in model simulation. Selection of PBL schemes is critical for accurate simulation of lower tropospheric CO_2 vertical distribution as shown in previous studies (Ballav et al., 2016; Diaz-

Isaac et al., 2018). Our study applies the YSU scheme based on a thorough investigation of the YSU scheme (Hu et al., 2010; Hu et al., 2012; Hu et al., 2013; Hu et al., 2019) and a test comparison with the MYJ scheme. The YSU scheme has been shown to perform well for both daytime and nighttime at the 20 km grid spacings used in this study (Hu et al., 2012; Yang et al., 2019). The YSU scheme is a nonlocal scheme with explicit treatment of entrainment fluxes, which was shown to be critical to reproducing convective boundary layer structures (Hu et al., 2013) and achieve a better performance than some local schemes such as the Mellor–Yamada–Janjic (MYJ) scheme (Wang et al., 2016). For stable boundary layer, an update in stability function in 2013 led to a better YSU performance in terms of reproducing nighttime profiles of both meteorological and chemical variables, particularly over the Great Plains (Wang et al., 2016). YSU led to a better CO_2 simulation than MYJ in our earlier WRF-VPRM application over the U.S. domain (Hu et al., 2020), thus it is chosen in this study. The YSU scheme has been demonstrated to perform well as one of the best options over East Asia for both air quality modeling (Huang et al., 2014; Liu et al., 2016; Wang et al., 2020) and meteorology modeling studies (Cheng et al., 2014; Tao et al., 2011).

Reference：

Ballav, S., Patra, P. K., Sawa, Y., Matsueda, H., Adachi, A., Onogi, S., De, U. K. (2016). Simulation of CO2concentrations at Tsukuba tall tower using WRF-CO2tracer transport model. Journal of Earth System Science, 125(1), 47-64. 10.1007/s12040-015-0653-y

Diaz-Isaac, L. I., Lauvaux, T., & Davis, K. J. (2018). Impact of physical parameterizations and initial conditions on simulated atmospheric transport and CO2 mole fractions in the US Midwest. Atmospheric Chemistry and Physics, 18(20), 14813-14835. 10.5194/acp-18-14813-2018

Hu, X.-M., Doughty, D. C., Sanchez, K. J., Joseph, E., & Fuentes, J. D. (2012). Ozone variability in the atmospheric boundary layer in Maryland and its implications for vertical transport model. Atmospheric Environment, 46, 354-364. DOI 10.1016/j.atmosenv.2011.09.054

Hu, X.-M., Klein, P. M., & Xue, M. (2013). Evaluation of the updated YSU planetary boundary layer scheme within WRF for wind resource and air quality assessments. Journal of Geophysical Research-Atmospheres, 118(18), 10490-10505. 10.1002/jgrd.50823

Hu, X.-M., Nielsen-Gammon, J. W., & Zhang, F. Q. (2010). Evaluation of Three Planetary Boundary Layer Schemes in the WRF Model. Journal of Applied Meteorology and Climatology, 49(9), 1831-1844. 10.1175/2010jamc2432.1

Hu, X.-M., Xue, M., & Li, X. (2019). The Use of High-Resolution Sounding Data to Evaluate and Optimize Nonlocal PBL Schemes for Simulating the Slightly Stable Upper Convective Boundary Layer. Monthly Weather Review, 147(10), 3825-3841. 10.1175/mwr-d-19-0085.1

Hu, X. M., Crowell, S., Wang, Q., Zhang, Y., Davis, K. J., Xue, M., . . . DiGangi, J. P. (2020). Dynamical Downscaling of CO2 in 2016 Over the Contiguous United States Using WRF-VPRM, a Weather-Biosphere-Online-Coupled Model. Journal of Advances in Modeling Earth Systems, 12(4), e2019MS001875. 10.1029/2019ms001875

Wang, W. G., Shen, X. Y., & Huang, W. Y. (2016). A Comparison of Boundary-Layer Characteristics Simulated Using Different Parametrization Schemes. Boundary-Layer Meteorology, 161(2), 375-403. 10.1007/s10546-016-0175-4

Yang, Y., Hu, X.-M., Gao, S., & Wang, Y. (2019). Sensitivity of WRF simulations with the YSU PBL scheme to the lowest model level height for a sea fog event over the Yellow Sea. Atmospheric Research, 215, 253-267. https://doi.org/10.1016/j.atmosres.2018.09.004

Comment#3: The fine resolution (20-km) is too coarse to capture Lin'an's footprint area, which would be roughly < 4 km at the height level under stable conditions. Therefore, it is hard and a bit unreasonable to directly compare with local-scale measured fluxes.

Response: We agree with the reviewer that grid resolution matters for air quality and meteorology simulations. We conduct a new set of simulation with 4km grid resolution over a smaller domain covering the TCCON-Hefei site and Lin'an tower site, and compare it with the 20km-grid simulation. In general, the 4km-grid simulation showed well consistent result with the 20km grid simulation over the same area, and no different conclusion could be drawn from the new set of simulation. The following figure (Figure S1 in revised manuscript) presents the spatial distributions of CO_2 and XCO_2 from the two sets of simulations. The 4km-grid simulation provided a more detailed presentation of the spatial distribution, but the levels of CO_2 and XCO_2 were quite close to the 20km-grid resolution, thus most of the discussions within the revised manuscript still use 20km-grid simulation data, only the discussion related with Lin'an tower data used the new 4km-grid simulation data.

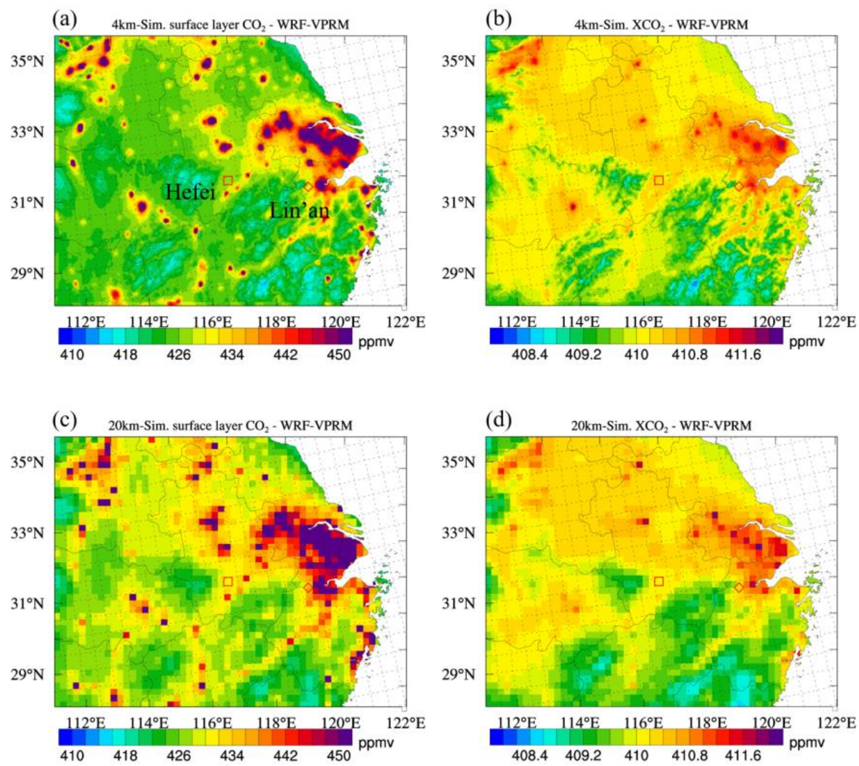

Figure: Annual averaged CO_2 (left column) and XCO_2 (right column) from WRF-VPRM 4km-grid simulation (top row) and 20km-grid simulation (bottom row). Locations of Hefei and Lin'an are presented with red rectangle and diamond.

For XCO_2 simulations, we find that the two sets of simulations differed by only 0.1 ppmv (<0.03%) at the TCCON-Hefei site. The following figure (Figure S2(a) in revised manuscript) presents the comparison of daily XCO_2 between 20km-grid and 4-km grid simulations at TCCON-Hefei site. The two simulations showed fairly close results.

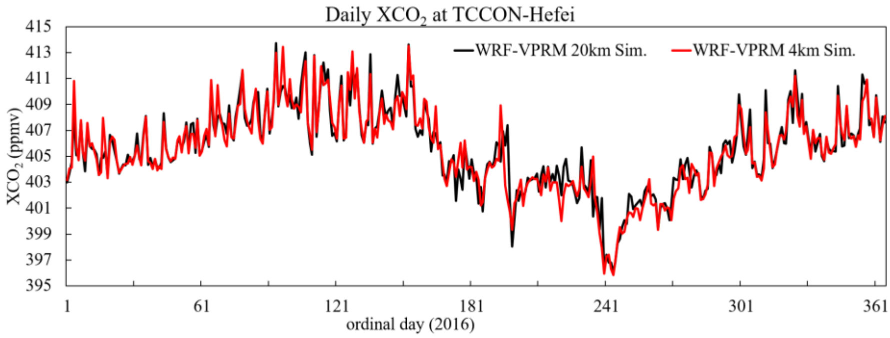

Figure: Comparison of 20km-grid and 4km-grid simulations at TCCON Hefei site.

For CO_2 simulations however, the 4km-grid simulation showed much smaller bias than the 20km-grid simulation at Lin'an tower for CO_2, thus we update within the manuscript to use 4km-grid simulation to compare with the Lin'an observation, as shown in the following figure (Figure 4(d) and (e) in revised manuscript).

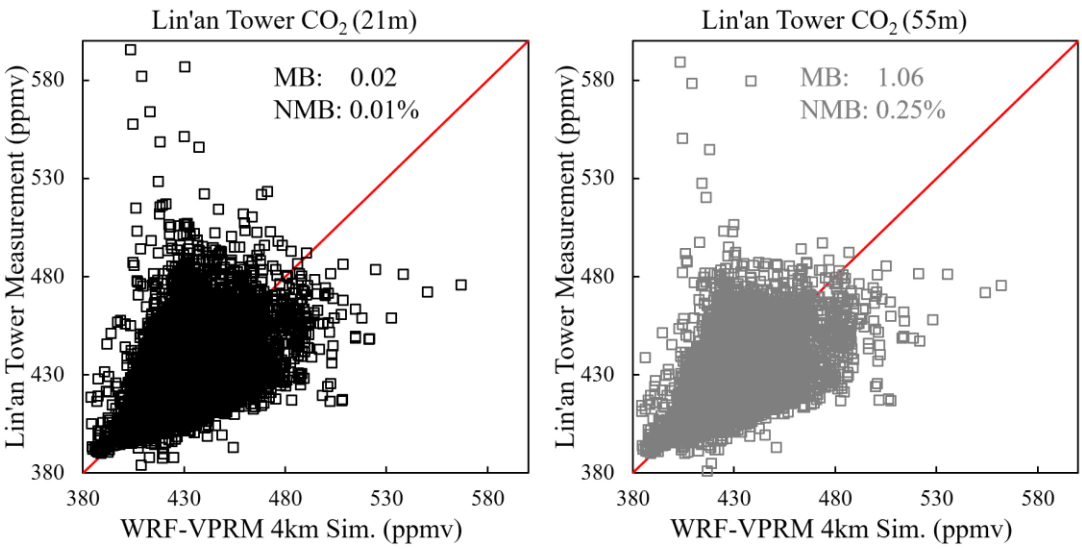

Figure: WRF-VPRM 4km-grid simulation evaluated against Lin'an tower observations at 21m (left) and 55m (right).

Comment#4: This manuscript's key sites or regions are Lin'an and Hangzhou, but their locations and site descriptions are missing. No mark on maps or description sub-section. This is very important for readers' understanding. In Line 295, for example, the authors tried to describe the transport of CO_2 plume from Hangzhou. However, readers do not know their spatial location, so they cannot catch up the further discussion. How far are the two locations? How much is Hangzhou close to efficiently affect to Lin'an? The authors explained in Line 240 that the Lin'an site could be affected by regional anthropogenic emissions. However, readers would not understand which regions or directions could be the main culprit. Therefore, wind direction analysis should be needed in Figure 6, where only wind speeds are displayed. Besides the location of Lin'an, its LULC features should be described in a sub-section.

Response: We agree with the referee that more details are necessary to demonstrate the locations of Lin'an and Hangzhou, especially for those unfamiliar with China. Lin'an is a district of Hangzhou city. The Lin'an Regional Atmospheric Background Station is about 60km west to the downtown center of Hangzhou. To show these details, we add the description of the location in the revised manuscript at line#129-130. We also include the following figure (Figure 2(c) in revised manuscript) to demonstrate the locations of Lin'an and downtown center of Hangzhou, and also demonstrate the prevailing wind at Lin'an. To demonstrate the wind speed as well as the wind direction, the wind rose map was derived from hourly observations of 10m and 55m wind speed and wind direction at Lin'an for 2016-2018. It shows the prevailing wind directions at Lin'an are northeast and southwest.

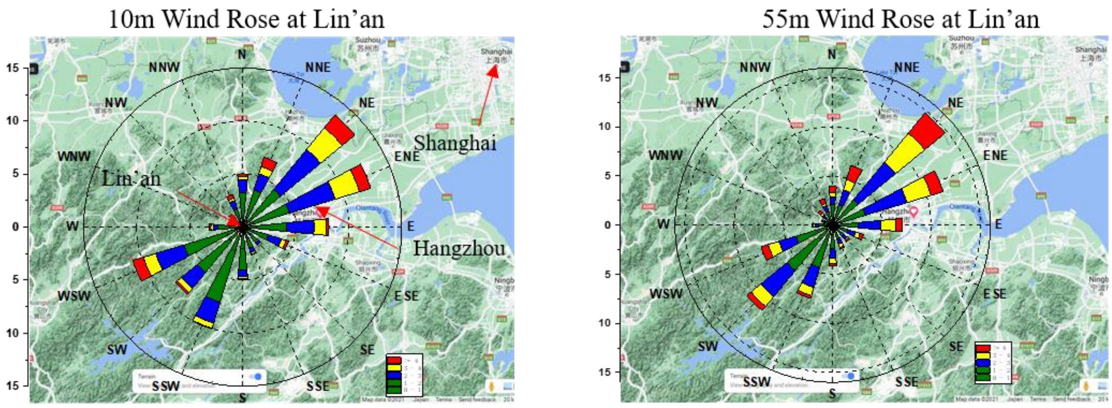

Figure: Wind rose map derived from Lin'an tower hourly observations of 10m (left) and 55m (right) wind speed and wind directions for 2016-2018.

Comment#5: A native English speaker should edit this paper, especially for tense. Usually, past tense is supposed to be used in the method and the results and discussion sections, especially for the action and experiment have done already.

Response: As recommended by the editor during our initial submission, the manuscript has been carefully edited by a native speaker, and a lot of grammar typos have been corrected before the open discussion. We have changed to past tense for the descriptions of modeling method. Most of the discussions have also been changed to past tense. Full version of revised manuscript is not allowed to be submitted during the open discussion, thus we list some some examples as below:

At line#88: "Both simulations were configured with 47 vertical layers with model tops at 10hPa."

At line#126: "Hourly measurements of CO2 concentrations were collected at the Lin'an Regional Atmospheric Background Station …"

At line#188: "Evaluation at the Lin'an station was performed with the 4km-grid simulation"

At line#251: "WRF-VPRM reproduced the trends in good agreement with ground and satellite

observations."
At line#269: "We find that both models prominently overestimated during nighttime, which shall be attributed to the bias in simulating NEE"

Comment#6: The main title is not proper for summarizing the whole content. Specifically, the first part of the sentence (before 'and') indicated only tower data, although the authors used integrated various measurement data. In the later part, after 'and', the sentence sounds like the WRF-VPRM model analysis, which is odd because we do not usually analyze the model itself.
Response: We agree with the referee that the original title emphasized too much on the tower measurement. We have revised the title as: "Analysis of CO_2 spatiotemporal variations in China using a weather-biosphere-online-coupled model"

Comment#7: Figures are a bit chaotically mixed, so readers cannot smoothly follow the writing flow. Please explain figure by figure in the body for the consistency of the flow of paragraphs. In Figure 1, for example, the spatial distribution (upper panel) and the photo of the Lin'an site (bottom panel) should be drawn on two different figures. In Figure 6, some sub-figures should also be separated.
Response: We agree with the referee that some of the figures contain too many sub-figures which may not belong to the same category. According to this comment, we have separated Figure 1 into two different figures (Figure 1 and Figure 2 in revised manuscript) to show the simulation domains and photos of Lin'an station separately. We also rearranged Figure 6 (Figure 7 in revised manuscript) as recommended by the reviewer as shown in response for comment#13.

Comment#8: Line 92: Add the version of WRF.
Response: We use WRF Version 3.9.1.1 for the WRF-VPRM model simulation. We have included this information at line#86 in the revised manuscript.

Comment#9: Line 283: What is ΔH?
Response: ΔH stands for the above ground height difference between the two levels being investigated. For the Lin'an CO_2 concentration observations, ΔH stands for the height difference between the 55m and 21m monitors, thus ΔH is 34m. We appreciate the referee for point out this issue, and we have included this information at line#292 in the revised manuscript as: "Fig.8(b) presents the correlation between air temperature gradient ($\Delta T/\Delta H$) and CO_2 concentration gradient ($\Delta CO_2/\Delta H$) calculated with annual averaged diurnal tower observations, where ΔT, ΔCO_2, and ΔH represents the difference of air temperature, CO_2 concentration, and height between the two tower levels. The temperature gradient and CO_2 concentration gradient clearly demonstrate the influence of boundary layer stability on the CO_2 vertical profile." We would also like to mention that in the original version of Fig.7(b) (now is Fig.8(b) in the revised manuscript), we used annual averaged diurnal data for each year to calculate the gradients, thus there were 24 data points for each year and there were 72 data points in the figure. But we just realize that it was not consistent with Fig.7(a) which showed diurnal profiles averaged for all three years. So, we calculate the gradients from diurnal data

averaged for all three years thus there are only 24 data points in revised manuscript, and the correlation is calculated as -0.98.

Comment#10: Line 300: Footprints at each level of the flux site should be quantified
Response: We agree with the referee that showing the footprints would be a straightforward demonstration to support the discussion regarding transport impact, but unfortunately there was no wind speed and wind direction measurement at 21m of the Lin'an tower. We only have wind observations at 10m and 55m. We apply the method proposed by Hsieh et al. (2000) to calculate footprints at these two levels. The scalar flux (F) and the footprint (f)are related by (equation 1 in Hsieh et al. (2000)):

$$F(x, z_m) = \int_{-\infty}^{x} S(x) f(x, z_m)\, dx$$

where $S\ (unit{:}\ g\ m^{-2}\ s^{-1})$ is the source strength, z_m is the measurement height, and the mean wind direction is along the horizontal coordinate, x. Based on this method, we calculated the peak location of the footprint ($x_{f=f_{peak}}$, equation 19 in Hsieh et al. (2000)) and the location where the fetch-to-height ratio equals 90% ($x_{F/S_0=0.9}$, equation 20 in Hsieh et al. (2000)) at 10m and 55m respectively as shown in the following figure. We applied the CALMET model (Scire et al., 1998) to calculate related variables such as friction velocity and sensible heat.

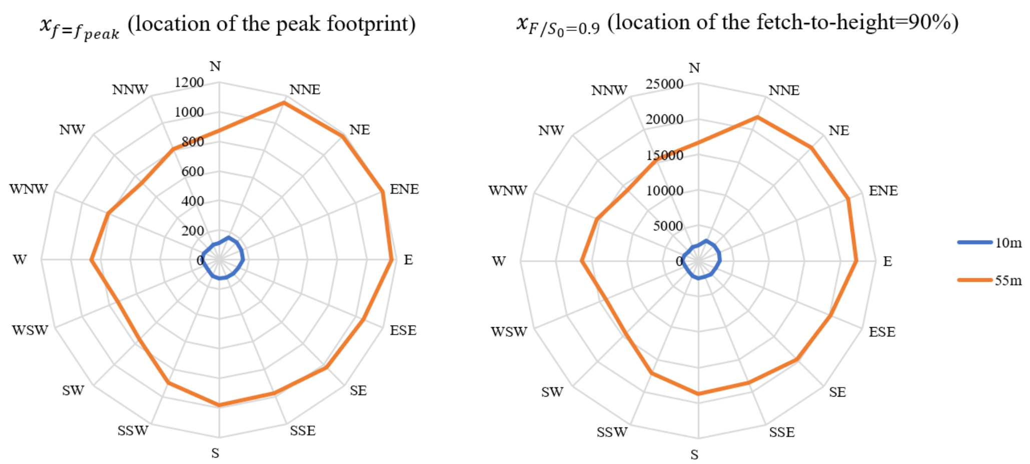

Figure: Locations where footprint reaches peak value (left); Locations where the fetch-to-height ratio equals 90%. Both units are meters.

The above figure demonstrates that upper air (55m) received influences from prominently longer distances than lower air (10m). Considering the dominant upwind directions are northwest at Lin'an tower (figure in response to comment#4), it's likely that 55m at Lin'an had larger footprints than 21m from Hangzhou. Footprint was mentioned at line#296 and line#300 in the original manuscript. In that paragraph, we attempted to demonstrate that the boundary layer stability was closely correlated with the CO_2 concentration gradient. Footprint was mentioned to further the discussion by demonstrating that 55m received more influence from Hangzhou than 21m. We assumed that upper air usually has larger footprint than lower air. However, this comment reminds us that we are not able to solidly demonstrate it because no wind speed and wind direction measurement was available at 21m. Thus we decide to remove the discussion regarding footprint (line#295-302 in original manuscript) in this revision, and our main conclusion in this paragraph remains unchanged..

Reference:

Hsieh, C.I., Katul, G., Chi, T.W: An approximate analytical model for footprint estimation of scalar fluxes in thermally stratified atmospheric flows, Advances in Water Resources, 23, 765-772, 2000.

Scire, J.S., Robe, F.R., Fernau, M.E., Yamartino R.J.: A user's Guide for the CALMET Meteorological Model (Version 5) Earth Tech Inc, Concord, MA (1998)

Comment#11: Figure 1: Figure 1(f) is missing, although Line 131 referred to it.
Response: We apologize for this careless typo. We have split the original Figure 1 into two figures as suggested by comment#7. We have revised the description as: "Flask samplings of CO_2 surface with monthly intervals are collected through the National Oceanic and Atmospheric Administration's (NOAA's) Earth System Research Laboratory (ESRL) at four sites (shown in Fig. 1(a)) within our modeling domain."

Comment#12: Figure 4: The graphic resolution is poor for (e). Readers cannot identify or separate the difference between the shaded area and others.
Response: We apologize for this careless mistake. The original figure has been automatically compressed in the .docx document. We have turned off the "automatic compress" option in Microsoft-Word software, and updated all figures with high resolution in the revised manuscript.

Comment#13 Figure 6. The scale of the y-axis must be matched for a clear comparison.
Response: The y-axis in Figure 6 has been adjusted accordingly as shown in the following figure (Figure 7 in revised manuscript). As recommended in comment#7, we reorganize the figure by removing the wind speed figure (c) as shown below. We also use wider distance between the CO_2 concentration figures (a-f) and NEE figures (g and h).

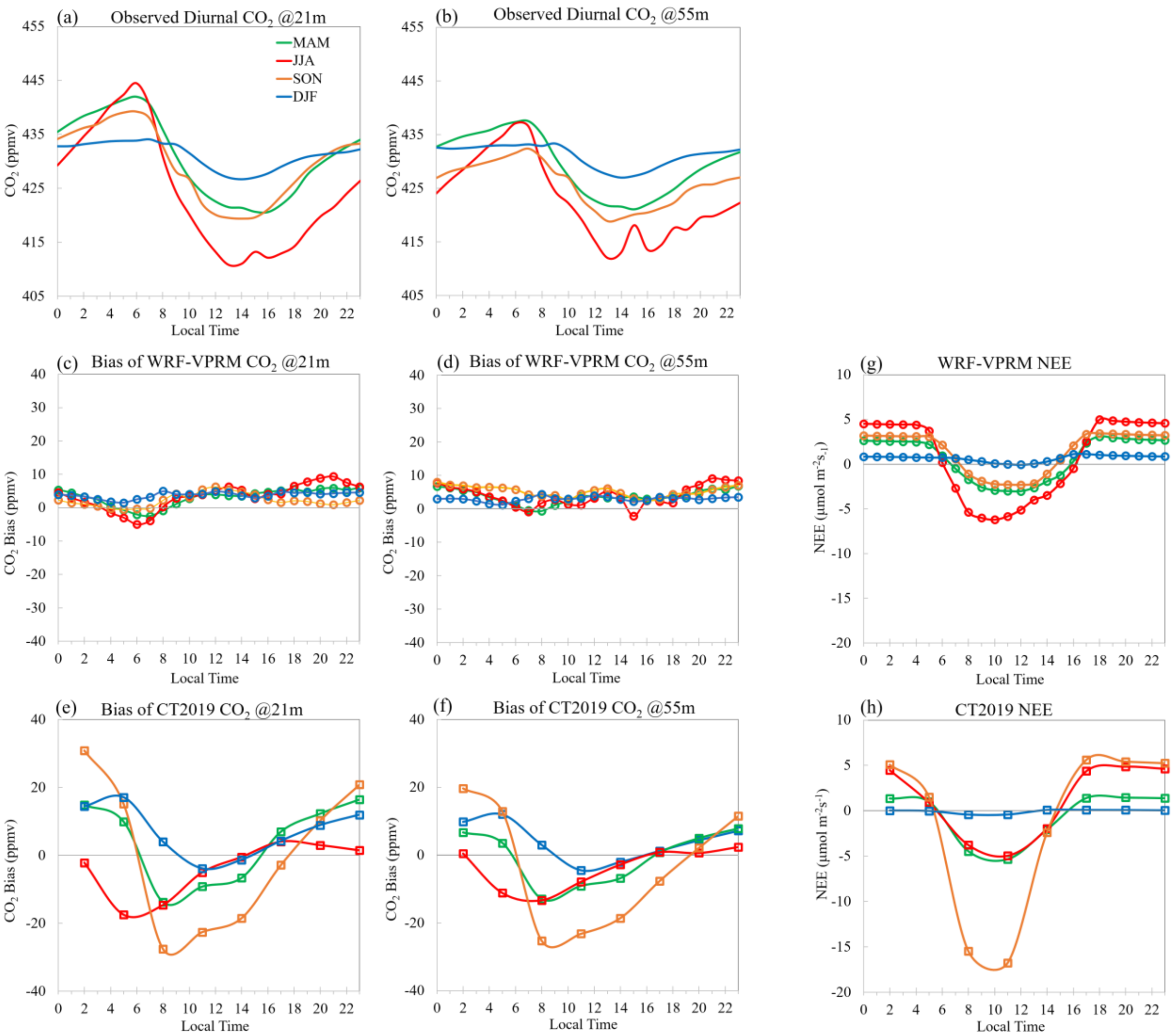

Figure 7: Seasonal mean diurnal variations of observed CO_2 at (a) 21m and (b) 55m; WRF-VPRM simulation biases of CO_2 at (c) 21m and (d) 55m; CT2019 simulated biases at (e) 21m and (f) 55m; Simulated NEE from (g) WRF-VPRM and (h) CT2019.

As the wind directions were already shown, we add a new figure (Figure S3 in the revised manuscript) to demonstrate the comparison of wind speed between 10m and 55m at Lin'an tower, as shown below.

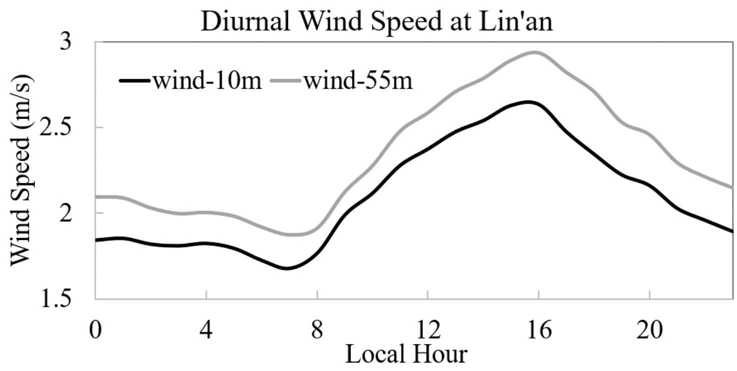

Figure: Observed diurnal profiles of wind speed at Lin’an.

Comment#14 Line 162: The full name of NMB is mentioned later, Line 166.

Response: We appreciate the reviewer for pointing this out. In the original submission we actually mentioned the full name twice at line#161 and line#166 respectively, we will remove the full name “normalized mean bias” at line#166 in the revised manuscript.