Review of: Anthropogenic and natural controls on atmospheric d13C-CO2 variations in the Yangtze River Delta: Insights from a carbon isotope modeling framework
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This paper describes a study of CO2 emissions in the megacity region of the Yangtze River Delta of China, which include several major cities in eastern China. The novel contribution of this study is the WRF-STILT modeling of the emissions making extensive use of the stable isotopic composition of carbon in CO2 (d13C-CO2). The simulation agrees well with the CO2 observations. The modeling of d13C-CO2 allows investigation of the contributions of various anthropogenic and biogenic sources. The topic of this study falls well within the scope of Atmospheric Chemistry and Physics. Therefore, this paper should be published after minor revision.

We thank the Reviewer for their thoughtful comments and detailed suggestions. All points have been addressed below (review query in Italic; author response in blue). Changes to the text in the manuscript have been marked in bold text.

My concerns include the need for clarification and further discussion of several points and the need for quantification of uncertainties in calculations resulting from the modeling runs. Particular instances of these are given below.
Done as suggested.

Specific comments:
Line 114: Replace "be used" with "been used."
Done as suggested.

Line 188ff: Move Figure 6 here, since you are describing it here. You should refer to it here, changing the number to 2, and therefore adjusting the figure numbers for the old 2-5 to 3-6, both in the text and in the figure captions.
Thanks for pointing this out. The description presented here is mainly for the method sections and has not yet referred to the analysis of background data. We believe it is best to keep Figure 6 as is.

Lines 205-206: The lowest quintile is the lowest 20%, not the lowest 5%. You can just say "the bottom 5%" to describe the data used in this approach to background.
Done as suggested.

Lines 216-218: Give the parameters you used in the CCGCRV curve fitting calculations.
Done as suggested. We added "(a digital filtering curving fitting program developed by the Carbon Cycle Group, NOAA, USA)" following CCGCRV, and also added "we derived CCGCRV curving fitting lines by using 11 regressed parameters, which were based on hourly the time series of observations/simulations".

Line 231: Add (Figure 2 (perhaps changed to Figure 3)) after "YRD."
Done as suggested, we added (Figure 2) after YRD.

Line 243: Replace "East China" with "Eastern China."
Done as suggested.

Line 247: Insert "backwards" after "locations."
Done as suggested.
Line 250: Replace "at the end of" with "for."
Done as suggested.

Lines 263-264 and elsewhere: Replace "EDGAR v432" with "EDGAR v4.3.2."
Thanks for pointing these out. We have made these corresponding revisions throughout this manuscript.

Line 276: Please clarify "enhancement." Is this proportion of emissions due to source i? "Enhancement" sounds like it is the amount of CO2 in excess background.
Here the enhancement is CO_2 mixing ratios contributed by different CO_2 emission sources as described on lines 180-182 "Note that $\boldsymbol{\Delta}CO_2$ is the sum of all simulated sources/sinks $[\boldsymbol{\Delta}CO_{2\_sim}]_i$ and represents the total simulated CO_2 enhancement. We use $\boldsymbol{\Delta}CO_{2\_obs}$ as the observed CO_2 total enhancement, which can be calculated by using the CO_2 observation minus the CO_2 background values.". The enhancement proportion indicates the proportions of a specific enhancement to total CO_2 enhancement. We added "where δ_i is the $\boldsymbol{\delta}^{13}C\text{-}CO_2$ value from source category *i*, and p_i is the corresponding enhancement proportion (i.e. proportions of a specific enhancement *i* to total CO_2 enhancement). We define $\boldsymbol{\delta}_{s\_sim}$ as the simulated carbon isotope ratio of all sources to differentiate it from the observed $\boldsymbol{\delta}_{s\_obs}$." on lines 301-303.

Line 286: When considering the biosphere in cities, people are starting to include the effects of human respiration and excretion (Turnbull et al., 2015; Miller et al., 2020, just published in PNAS), using information from Prairie and Duarte (2007). You might want to comment on how this would affect your analysis. The d13C of human respiration should reflect that of the average diet. Also, is any bioethanol used in the gasoline? You should confirm this, since this is common is some cities.

Thank you for raising this concern. As mentioned in previous studies (Turnbull et al., 2015; Miller et al., 2020), both biofuel combustion and human respiration will emit CO_2. For the biofuel combustion related CO_2 emissions, there are bioethanol in the gasoline and other fuels, which have been attributed to organic emissions in the EDGAR inventory and considered in our simulations on lines 313-314 as "biofuel combustion and biological emissions (−28.20‰ ± 1.00‰)".

For the CO_2 emissions related to human respiration, our previous study found it only accounted for 3.7% of anthropogenic emissions in the YRD area (Xu et al., 2017), which was a relatively smaller role and most of the local human food diet is dominated by C_3 grains, having the same $\boldsymbol{\delta}^{13}C\text{-}CO_2$ value as biological CO_2 flux of −28.20‰. Also note the biological CO_2 flux (used in this study) from Carbon Tracker assimilation system considered anthropogenic is fixed and attributed the rest of CO_2 changes to biological CO_2 flux (Peters et al., 2007). Therefore we believe the uncertainty of the biological CO_2 flux will contain the small proportion of human respiration. We have added more description to clarify it on lines 316-323 "Since CO_2 emissions associated with human respiration (Prairie and Duarte, 2017; Turnbull et al., 2015; Miller et al., 2020), is relatively small (3.7% of anthropogenic emissions in the

YRD area, Xu et al., 2017), and given that the local food diet is dominated by C_3 grains that have a similar $\boldsymbol{\delta}^{13}$C-CO_2 value as the biological CO_2 flux of −28.20‰, we assume it has the same isotope signals as local C_3 plants and ecosystem respiration. Further, the biological CO_2 flux from the Carbon Tracker assimilation system considered anthropogenic as fixed and attributed the remainder to the biological CO_2 flux (Peters et al., 2007). Consequently, we believe the uncertainty in the biological CO_2 flux will include the small proportion of human respiration."

Line 322: Replace "blue" with "blue-red."
Done as suggested.

Lines 333-334: Replace "below" with "to."
Done as suggested.

Line 335: What hours did you use for daytime? Most modelers stress that mid-day to midafternoon hours work best, when the planetary boundary layer height (PBLH) is best simulated.
Here we displayed (1) all hours (2) nighttime and (3) daytime, respectively in Figure 3, and hours between 10:00 to 16:00 represent daytime, hours between 22:00-6:00 are for nighttime, we added them on line 371 for clarification, which was also list in the label of Figure 3. We agree that generally daytime hours work better for the PBLH variations because of strong vertical development in the daytime, meanwhile some recent studies also found WRF-STILT will underestimated the PBLH compared with Lidar observations (Sargent et al., 2018), which indicate the daytime PBLH performance is not as good as nighttime. Another reason for choosing all-day simulations is that both biological and anthropogenic CO_2 flux have strong diurnal variations (i.e. much higher in daytime and lower in nighttime for anthropogenic emissions), so if only use daytime observations, the derived scaling factors will reflect bias in both the *a priori* diurnal scaling factors and daily averages of CO_2 emissions, so even the scaling factors is larger than 1, it does not only indicate the anthropogenic CO_2 is underestimated, it can also be caused by underestimation of diurnal scaling factors in daytime not the daily averages. Considering above factors, we compared model-observation for daytime, nighttime and whole day in Figure 3b-d, and finally choose the whole day averages to scale monthly CO_2 emissions.

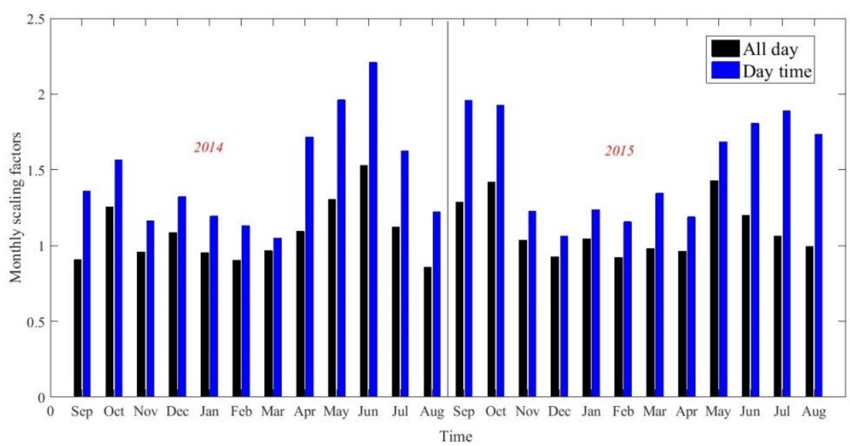

Figure R1. Derived monthly scaling factors for all day and only day time.

We added the following details: "The monthly scaling factors derived from using daytime and all-day observations are also shown in Figure S4. These factors vary seasonally with higher values observed in summer. When using daytime values only, the scaling factors were much larger than the all-day values. This can be seen in Figure 3 by comparing the simulated and observed CO_2 mixing ratios. We should note here that the larger scaling factors based on the daytime data could be caused by bias in the *a priori* daily scaling factors used to generate the hourly CO_2 emissions (Hu et al., 2018b); the monthly anthropogenic averages; and bias in negative biological CO_2 enhancement. Since our study is mainly focused on the seasonality of all-day observations, the monthly scaling factors derived from the all-day approach will be used for the following analyses." on lines 455-464 for clarification.

Your Figure 3 suggests that the monthly average of nighttime modeling results matches the observation best.
Yes. The results indicate that nighttime modeling has less bias than daytime. It's one of the reasons why we not choose all-day instead of daytime observations to do the constraint at monthly scales.

Line 345: What are the two months that fall far below the trend in Figure 4a? Do you have an explanation for these?
These two months are March and August. It indicates that PBLH variations and other meteorological factors (i.e. monthly changed footprints sources) also play a role in affecting CO_2 variations. We added on lines 383-385 "We also note that there were two months (March and August) that fall far below this trend, implying that changes in the monthly footprints (source area) can also play an important role."

Line 359: Neither Figure 5a nor 7b is consistent with a negative average summer NEE. Indeed, Figure 5a suggests the opposite since all 2014 summer months are positive in NEE/biological contribution to the CO2 enhancement, as are June and July 2015.
Yes, as displayed in Figure 7b, the daytime NEE are generally negative and nighttime NEE are positive in summers, which will lead to negative CO_2 enhancement in daytime and positive CO_2 enhancement in nighttime, respectively. The Figure 5a displayed monthly averaged biological CO_2 enhancement which will smooth the diurnal variations. Another reason is that daytime PBLH were generally much higher than nighttime, which leads to much lower absolute CO_2 enhancement in daytime than in nighttime, and the averages of daily or monthly CO_2 enhancement will appeared as CO_2 positive values.

Line 363: Replace "4b-c" with "4b-d."
Done as suggested.

Lines 375-376: What are the uncertainties in the observed anthropogenic CO2 enhancements? In general, please give uncertainties.
Here, uncertainty of the observed anthropogenic CO_2 enhancement mainly came from CO_2 background and simulated biological enhancement. Where both CO_2 background and biological NEE were derived from Carbon Tracker systems as described in Methods Section. To quantify the uncertainty of Carbon Tracker CO_2 background data, we first calculated the annual averages at Mauna Loa background site (https://scrippsco2.ucsd.edu/data/atmospheric_co2/mlo.html, the red dot as list below). The averages were 398.04 ppm and 400.08 ppm, and the background values derived from Carbon Tracker system were 400.43 ppm and 402.21 ppm for 2013 and 2014, respectively. The comparisons between the Mauna Loa

site and Carbon Tracker system were consistent with only a 2 ppm difference. We believe the actual bias in the Carbon Tracker system above China is likely smaller than 2 ppm because the atmosphere should be slightly enhanced by local emissions compared to Mauna Loa site. Based on the above analyses, we attribute a 2 ppm uncertainty to the background estimate. For the uncertainty derived from simulated biological enhancement, we attribute a larger 50% relative bias based on our previous study (Hu et al., 2018b), which used eddy covariance flux measurement to evaluate biological CO_2 flux in Carbon Tracker systems. Based on the above calculations, we updated the results on lines 413-416 as:

"were 38.36±3.32 ppm and 37.89±2.80 ppm for 2014 and 2015, respectively. Here, the uncertainty of the observed anthropogenic CO_2 enhancements was calculated by prescribing a 2 ppm potential bias for the Carbon Tracker CO_2 fields and 50% to the simulated biological CO_2 enhancement (Hu et al., 2018b)."

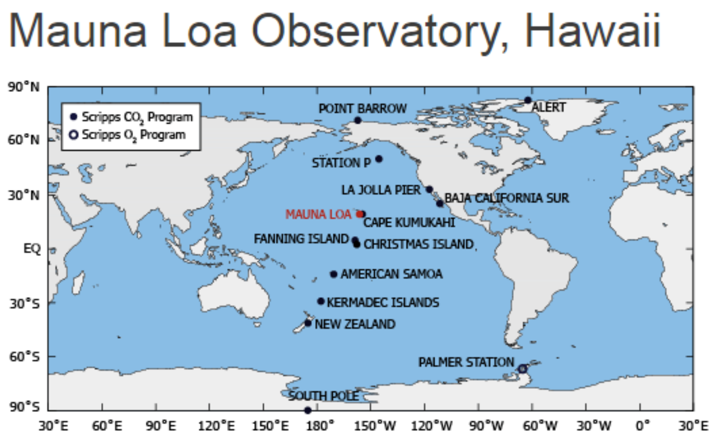

Figure R2. Locations of Mauna Loa background site (red color).

Lines 388-390: The absolute enhancements depend on many things, including the meteorology and the magnitude of the emissions. You can't conclude that the YRD has more emissions simply because the enhancements are higher. Modeling is critical for coming to that conclusion.

Thank you for pointing this out. We agree that CO_2 enhancements are influenced by meteorology and the magnitude of the emissions. We deleted "indicating greater anthropogenic CO_2 emission".

Line 392: Explicitly explain where these percentages come from.

These percentages were calculated by dividing the simulated CO_2 enhancement from each province by simulated total CO_2 enhancement for the whole domain. The CO_2 enhancement from each province was simulated by multiplying CO_2 emissions in each province with the corresponding footprint. For clarification, we added "The CO_2 enhancements from each of the 5 zones were simulated by multiplying CO_2 emissions in each province with the corresponding footprint." on lines 257-259.

Line 395: Where do you show that the maximum source contribution exceeded 50% on 19 September 2013?

The reason to mention this extreme situation to illustrate the large influence of long-distance transport at some special periods. We added "not shown" for clarification on line 435.

Lines 396-399: Please explain how the "anthropogenic enhancement" is different from the *"anthropogenic emissions."*

The anthropogenic emissions represent anthropogenic CO_2 emissions (or flux), and anthropogenic enhancement represents anthropogenic CO_2 enhancement (or concentration) simulated by using CO_2 emissions in atmospheric transport model. Theoretically, if CO_2 emissions for different categories were homogeneously distributed, the two proportions of "anthropogenic enhancement" and "anthropogenic emissions" for the same category should be the same, while in the real situations both meteorological factors and CO_2 emission's spatial distributions will bring inconsistence between the "anthropogenic enhancement" and "anthropogenic emissions" for each CO_2 category. The comparisons between them is to illustrate whether enhancement proportions between each CO_2 category can represent corresponding emission proportions. We added "The comparisons between the proportions of simulated enhancement and proportions of corresponding CO_2 emissions can illustrate whether CO_2 enhancement partitions is a good tracer for emissions in a complex urban area." on lines 439-441.

Line 408: Replace "2014" with "2014-2015."

Done as suggested.

*Lines 408 and 415: Express the emissions as *1011 kg, the same units as in Table 1, for consistency.*

Done as suggested. We also changed the units from 10^{12} kg to 10^{11} kg on line 451.

Line 417: Replace "is" with "are."

Done as suggested.

Lines 430-431: A positive biological CO2 signal during winter is consistent with a negligible role for photosynthesis, but it could be that photosynthesis is still important, just not as important as respiration. Will human respiration affect this?

Yes. We agree that it can be also explained by the fact that photosynthesis is still important, just not as important as respiration. These changes have been applied to lines 487-488 as "which implies a positive biological CO_2 signal where ecosystem respiration is more important than photosynthesis"

As replied above for the CO_2 emissions related to human respiration, our previous study found it only accounted for 3.7% of anthropogenic emissions in the YRD area (Xu et al., 2017), which was a relatively smaller role and most of the local human food diet is dominated by C_3 grains, having the same $\boldsymbol{\delta}^{13}C\text{-}CO_2$ value as biological CO_2 flux of −28.20‰. Also note the biological CO_2 flux (used in this study) from Carbon Tracker assimilation system considered anthropogenic is fixed and attributed the rest of CO_2 changes to biological CO_2 flux (Peters et al., 2007). Therefore we believe the uncertainty of the biological CO_2 flux will contain the small proportion of human respiration. We have added more description to clarify it on lines 316-323 "Since CO_2 emissions associated with human respiration (Prairie and Duarte, 2017; Turnbull et al., 2015; Miller et al., 2020), is relatively small (3.7% of anthropogenic emissions in the YRD area, Xu et al., 2017), and given that the local food diet is dominated by C_3 grains that have a similar $\boldsymbol{\delta}^{13}C\text{-}CO_2$ value as the biological CO_2 flux of −28.20‰, we assume it has the same isotope signals as local C_3 plants and ecosystem respiration. Further, the biological CO_2 flux from the Carbon Tracker assimilation system considered anthropogenic as fixed and attributed the remainder to the biological CO_2 flux (Peters et al., 2007). Consequently, we believe the uncertainty in the biological CO_2 flux will include the small proportion of human respiration."

Line 434: Replace "domain" with "background."
Done as suggested.

Line 435: Add "(Figure 6)" after "respectively." This may become Figure 2.
Done as suggested.

Line 454: Replace "Figures 3 and 7" with "Figures 3a and 8", but the vertical scale in Figure 8 is too compressed to be seen clearly.
Done as suggested.

Lines 465-469: Please give uncertainties. Are the seasonal increases significant? Could PBLH simulation issue explain the large discrepancies, especially since the model diurnal variations are greater than those in the observations.
The main uncertainties associated with the simulation of hourly CO_2 are uncertainty in the meteorological fields, transport model, and *a priori* CO_2 flux. As shown in Figure R3, linear relationship between hourly CO_2 and $\delta^{13}C$-CO_2 bias were observed. This suggests the hourly $\delta^{13}C$-CO_2 simulations have similar bias as the sources. At the annual scale, the main uncertainty for both CO_2 and $\delta^{13}C$-CO_2 is attributed to the PBLH simulations and *a priori* anthropogenic CO_2 emissions. Here the bias for *a priori* anthropogenic CO_2 emissions were < 6% as calculated in this study, and the bias caused by PBLH uncertainty was usually <13% (Hu et al., 2018a; 2018b). Therefore, we attribute an uncertainty of 20% for the simulated CO_2 and $\delta^{13}C$-CO_2 at the annual scale. We have added "The main uncertainties associated with the simulation of hourly CO_2 and $\delta^{13}C$-CO_2 are uncertainty in meteorological fields, transport model (i.e. number of released particles), and *a priori* CO_2 fluxes. At the annual scale the main uncertainty is attributed to the PBLH simulations and *a priori* anthropogenic CO_2 emissions. The anthropogenic CO_2 emissions biases were < 6% as described above, and the bias associated with PBLH uncertainty was typically <13% (Hu et al., 2018a; 2018b). There, we attribute a 20% uncertainty to the simulated CO_2 and $\delta^{13}C$-CO_2 signals on an annual time scale." See lines 469-475 for clarification.

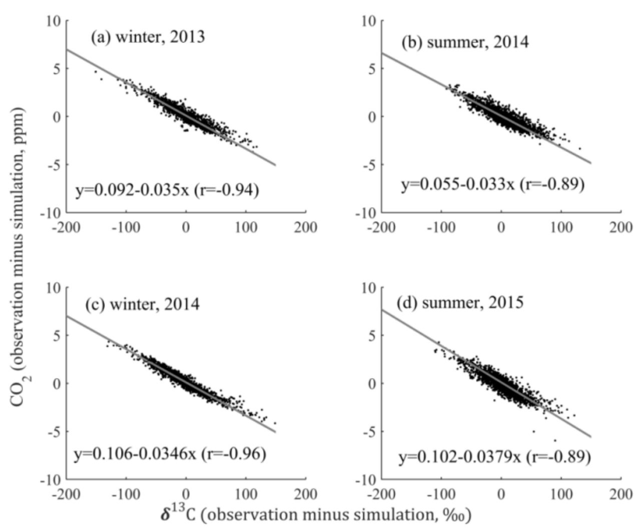

Figure R3. Relationship of observation minus simulation residual between CO_2 and $^{13}CO_2$ for (a) winter in 2013, (b) summer in 2014, (c) winter in 2014, and (d) summer in 2015.

We changed "significantly" with "obviously" for clarification. We also agree that the large discrepancies of hourly $\boldsymbol{\delta}^{13}$C-CO_2 variations were mainly caused by CO_2 simulations, which was basically caused by simulations of atmospheric transport process and PBLH simulations can have large influence. We added "(potentially caused by PBLH simulation issue during these periods)" on line 526-527.

Line 505: Replace "than" with "in." The baseline simulation in Figure 10b (red) is more enriched in the heavy isotope, as evidenced by its less negative values between April and October.

Done as suggested. We revised the typo by replacing "than" with "in". Yes. The baseline simulations of $\boldsymbol{\delta}^{13}$C-CO_2 in Figure 10b (red line, containing photosynthesis) is more enriched in heavy $\boldsymbol{\delta}^{13}$C-CO_2 than blue line (excluding photosynthesis) between April and October, which was caused by discrimination associated with ecosystem photosynthesis as previously explained on lines 485-487.

Lines 509ff: Are the differences of 0.08-0.20‰ significant? Please give uncertainties. Similar comment for the next paragraph.

The difference of 0.08-0.20‰ only accounted for 5%~13% of observed/simulated $^{13}CO_2$ seasonality ~1.5‰. Since there are only 2 numbers, the statistics cannot be calculated to report significance. We revised this sentence as "Generally, both ecosystem photosynthesis and respiration played minor roles in controlling the atmospheric $\boldsymbol{\delta}^{13}$C-CO_2 seasonality within this urban area".

*Line 530: Add the definition of the regional source term "(ds*DCO2)."*
In general, be consistent with "ds"

The $\boldsymbol{\delta}$s×$\boldsymbol{\Delta}CO_2$ can be treated as the regional source term. For additional clarification we have added "The product on the right-hand side of equation 3 is the simulated regional source term that is added to the background value and contains both enhancement and $\boldsymbol{\delta}^{13}$C-CO_2 signals contributed by different CO_2 sources/sinks. This product can also be treated as an observed term when using the derived $\boldsymbol{\delta}_{s\_obs}$ and observed $\boldsymbol{\Delta}CO_{2\_obs}$ values" on lines 192-195.

Line 544: If you use nighttime simulations, you still have respiration.

Yes. We agree that nighttime observations will still include respiration. The choice of choosing nighttime data is to minimize the influence of respiration and to mainly focus on anthropogenic CO_2 sources. We have added "mainly" before "focus on the anthropogenic CO_2 sources" on line 614.

Lines 597-598: Do you need to show both 13a and b? They are almost identical. You could just show one and generally state the results for the second in the text.

Since both nighttime and all-day were analyzed (see response to Reviewer 2) we prefer to retain these analyses.

Lines 600-601: Replace "relatively similar with" with "similar to."

Done as suggested.

Lines 608-609: Insert "absolute" after "1.57%." Are the uncertainties in your calculations small enough that "a 0.013‰ – 0.038‰ change" is significant?

Done as suggested. The uncertainty of the calculated sensitivity of change in atmospheric δ_{13}C-CO_2 to cement proportions should be much smaller than "0.013‰ – 0.038‰" because the uncertainty is a relative value not an absolute value.

Lines 621-622: Add "calculated from the simulations" before "was shown" and "From the EDGAR v4.3.2 inventory" after "proportion."
Done as suggested.

Figure 1: More information is needed in the caption – significance of the different-colored boundaries. Is the red triangle in (b) the same as the blue dot in (a) (Nanjing UIST)?
Done as suggested. We revised the caption as "Figure 1. (a) Weather Research and Forecasting Model simulation domains and the location of WLG site , the different region colors represent three domains, (b) cement production distribution in YRD and Eastern China. Both green dot in (a) and red star in (b) are NUIST observation site."

Figure 2: What is the base map in the middle of (a) – city lights?
The base map is annual total anthropogenic CO_2 emissions in our study domains, and it is explained in the caption.

Figure 4: Explain the Ds – what quantities are subtracted? Is the PBLH from the model? Have you compared the simulated PBLH with data? Are the data plotted in (a) averages for all hours of the day?
The ΔPBL height is the difference of simulated PBL heights in the same month for different years. Since there is a lack of PBLH observations, it has not been compared to field observations. The data in (a) are for all hours. We also added "these data points represent the difference of monthly averages in two different years for all hours." in the caption of Figure 4.

Figure 5: I think the captions for (b) and (c) are switched.
Done as suggested.

Figure 7: More explanation is needed in the caption. What is the origin of the background in (a)? What are the vertical lines in (b)? The latter question can be avoided by using the same shading in both panels.
Done as suggested.

Figure 8: This figure is good for showing model/observation comparison, but the vertical scales are too compressed to show long-term temporal variations or to compare between years.
Done as suggested, we have expanded the y-axis of CO_2 and δ^{13}C-CO_2.

Figure 9: (a) and (b) – the yellow color is very difficult to see. How do these plots look if you only use mid-day or early-mid afternoon results?
Done as suggested, we changed yellow to red color.

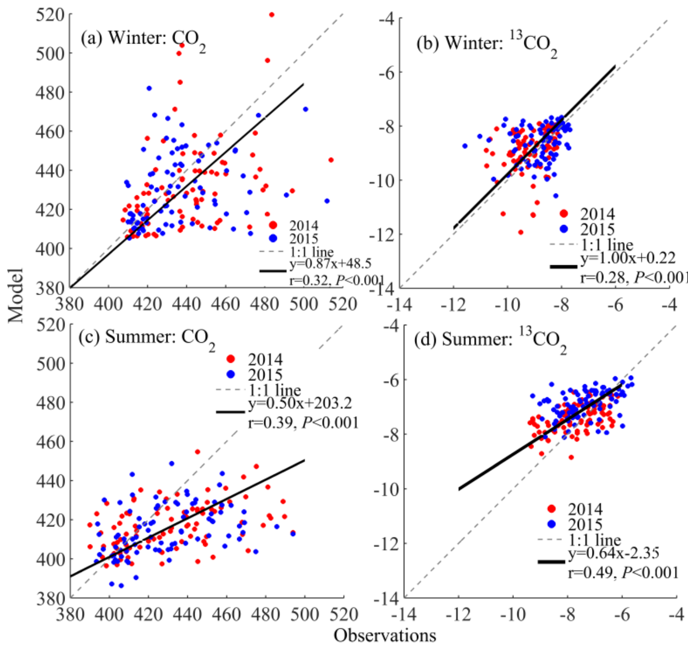

Figure R4. Scatter plots of observed versus modeled (a) winter time CO_2 mixing ratios, (b) winter time $\delta^{13}C$-CO_2, (c) summer time CO_2, and (d) summer time $\delta^{13}C$-CO_2 for both years, here these dots are day-time (10:00-16:00) averages.

We also did the comparisons by only choosing daytime observations. The results indicated that daytime CO_2 mixing ratio simulations in summer were slightly underestimated and that this causes $\delta^{13}C$-CO_2 to be overestimated. The simulations in winter can generally capture the trends for both CO_2 and $\delta^{13}C$-CO_2, during which the biological CO_2 enhancement played a relatively smaller role than anthropogenic emissions. We added this figure in supplemental file and discussed it in main text on lines 533-542.

Figure 10: "Observation" in the legend should be plural ("Observations"). What is the solid blue curve in (a) – probably the dashed blue line in the legend. This is difficult to see.

Done as suggested.

Figure 11. The 1:1 lines are not dashed in the figures, although the legends say they are. Please distinguish the 1:1 lines from the regression lines.

Done as suggested.

Figure 13. More explanation is needed in the caption. "Cement proportion" of what? Total anthropogenic CO2 emissions? EDGAR? "Cement increase ratios" – please explain what this is. Please be explicit as to what strategies 1 and 2 are, especially since some readers focus on the figures and not on the text.

Done as suggested. We revised this caption as "Sensitivity tests showing the influence of cement CO_2 emissions on δ_s for (a) nighttime, (b) all-day, and (c) the relation between cement CO_2 and $\delta^{13}C$ for simulation strategies 1 (There is no bias in the total anthropogenic CO_2 enhancement such that a proportional increase/decrease in the cement component does not change the relative anthropogenic contributions) and 2 (only the cement enhancement changes). Note that the numbers in brackets indicate changes in $\delta^{13}C$ with cement CO_2 enhancement proportion (the fraction of cement CO_2 enhancement to simulated total CO_2 enhancement) increase by 0.2 times. The x-axis values indicate changing cement enhancement proportions to 0.8 1.2, 1.4, 1.6, 1.8, and 2 times the original values." for clarification.

Table 1.: Explain "/"

Done as suggested, the "/" means not available.

Table 2.: Can you add rows for the average values for the model results and the observations for both CO2 mixing ratios and d13C-CO2 for each column?

Done as suggested.