

## Response to Editor

*Main concerns 1)  
inclusion and propagation of uncertainties*

We have expanded the discussion on measurement uncertainties as follows:

“The typical 5-min measurement precision is 0.3 ‰ for  $\delta^{13}\text{C}$  and 0.05  $\mu\text{mol mol}^{-1}$  for  $\text{CO}_2$  mole fraction according to the instrument manufacturer. Our own Allan variance analysis revealed a precision of 0.05 ‰ for  $\delta^{13}\text{C}$  and 0.07 ppm for  $\text{CO}_2$  mole fraction at the hourly averaging interval. The precision of the ambient measurement was lower than this due to error propagated through the calibration procedure. According to a laboratory test on an analyzer of the same model and using the same calibration procedure as ours, the hourly  $\delta^{13}\text{C}$  precision is about 0.4 ‰ (Wen et al., 2013). The calibration gases have much lower  $\delta^{13}\text{C}$  than the ambient delta values. The mismatch in the delta value between the calibration standard and the ambient air is common in applications of the IRIS technique ( $^{13}\text{C}$  in methane, Röckmann et al. 2016;  $^{18}\text{O}$  in water vapor, Lee et al. 2005;  $^{13}\text{C}$  in  $\text{CO}_2$ , Bowling et al. 2003). But because the system measures the concentrations of the major and the minor isotopologue independently, it is not critical that the calibration standard and the measurement target have matching isotope ratios (Bowling et al. 2005).”

*Main concerns 2)  
regression via Miller-Tans and Keeling methods*

Thank you for your recommendations on the papers relevant to this topic. After careful reading of these papers, we now realize that the geometric mean regression method we used originally is not a good choice. We now use the ordinary least squares regression. This point is noted in the Methods section:

“For the purpose of comparing with the Miller-Tans results, we also estimated the source isotope ratio using the Keeling mixing line method. According to Wehr and Saleska (2017), at the measurement uncertainties of our instrument system, the ordinary least squares procedure has much lower bias errors of parameter estimation than the geometric mean regression. In the following, the results of both the Miller-Tans method and the Keeling mixing line method were based on the ordinary least squares regression.”

The change in the regression method caused the mean  $\delta\text{s}$  to decrease by an average amount of 0.9 ‰. As a result, the partitioned biological flux in the YRD changed from being a weak sink to a weak source.

*Main concerns 3)  
Please include uncertainties in all source isotope delta end members ( $\delta_F$ ,  $\delta_C$ ,  $\delta_P$ ) and fluxes ( $F_F$  and  $F_C$ ) propagate errors formally, stating the values and uncertainties of each term. A Monte Carlo analysis is not justified because the underlying mass-balance equations are explicit. Disregarding the fact that it is not needed for the propagation of errors, the MC analysis (in l. 459-467) omits the uncertainties in the fluxes  $F_F$  and  $F_C$  and the isotope deltas  $\delta_S$  and  $\delta_C$ . Most importantly, the uncertainty treatment should not be dealt with "as an afterthought", but should be carried out consistently throughout the manuscript.*

We have now included uncertainties in all isotope delta end members and the fluxes (Tables 2 and 3; Figures 6 - 8).

The Monte Carlo procedure is used frequently by the hydrological community to quantify uncertainties associated with the partitioning of hydrological inputs to lake systems from isotope end members. There, the partitioning equations are also explicit expressions of the mass conservation principle. We used the same procedure to quantify how uncertainties in the isotope end members and the anthropogenic fluxes propagate through the partitioning equations. (In this updated analysis, we have considered uncertainties in all the input terms. Thank you for your suggestion.)

We now treat the issue of uncertainty throughout the paper. We have added the following description to the Methods section:

“The partitioning equations are explicit expression of the mass balance principle. But uncertainties in the isotope delta end members and the anthropogenic fluxes can propagate through these equations, causing uncertainties in the estimation of  $F_S$  and  $F_P$ . Here we used a Monte Carlo analysis to quantify these uncertainties. The same analysis has been applied to the partitioning of lake water budgets from isotope end members (Jasechko et al. 2014). The procedure employed a Gaussian distribution for errors in the input variables and an ensemble of 10,000 realizations for each month. Errors in  $F_S$  and  $F_P$  were computed as one standard deviation of these realizations after excluding the top and bottom 50 extreme ensemble members.”

*Main concerns 4)*

*The fact that your Miller-Tans regressions (Fig. 3 and Fig. 4) show non-zero intercepts provides the insight that they are not a suitable method to analyse the data on the annual timescale. You need to provide evidence that the method is suitable on shorter time-scales, in particular, the monthly timescale you are envisaging. A suitable timescale would be one that results in a non-zero intercept. You might find that a monthly timescale is not appropriate either, in which case an even more finely-grained analyses would be required, e.g. diel or even hourly, with subsequent aggregation of the derived source isotope deltas into daily/monthly/annual means. This was already pointed out by Referee #1. Please include a set of figures (Miller-Tans plots) that shows the data aggregated to subannual time-scales (monthly, possibly diel) and state the resulting source isotope deltas and their uncertainties.*

That the Miller-Tans intercept is not zero is indeed a cause of concern. At the monthly time scale, the original Miller-Tans method has a non-zero intercept in the range of 300 to 900 %  $\mu\text{mol mol}^{-1}$ . (A monthly example is now provided in Supplementary Figure S2.)

Following your suggestion, we have tried the 5-hour moving-window technique described by Vardag et al. (2016) for January and July 2014. Only 4 % of the data (all obtained at nighttime) satisfies their data screening criteria. The mean regression equation is  $y = -26.41 (\pm 4.03) x + 428.84 (\pm 211.12)$  for January and  $y = -25.64 (\pm 6.39) x + 687.83 (\pm 264.67)$  for July, where  $x$  is  $(C_a - C_b)$  and  $y$  is  $\delta_a C_a$ .

That the intercept does not go to zero even at such a short-time scale leads us to hypothesize that the concentration and isotope measurements at the WMO baseline background site

(WLG) do not represent the true background for this highly urbanized region. We have also applied the method to other isotope data published for urban areas, and found that the problem seems common for urban studies. Most of these urban datasets yield nonzero intercepts (Supplementary Figure S3 and Table S1).

In the end, we decided to replace the WLG measurement with the tropospheric CO<sub>2</sub> concentration calculated by CarbonTracker over the YRD region as the background. To overcome the problem that CarbonTracker does not calculate tropospheric  $\delta^{13}\text{C}$ , we rearranged the original Miller-Tans equation to a form that allows an intercept but without the need for a known background  $\delta^{13}\text{C}$ . The changes of the background and the regression equation cause  $\delta_s$  to decrease slightly by a mean value of 0.1 ‰, in comparison to the results obtained with the WLG as the background and the original Miller-Tans equation.

Please refer to Section 2.2 (which is completely rewritten) for additional explanation.

*Main concerns 5)*

*I've tried to replicate the slope of -12 ‰ (instead of -24 ‰), which Referee #2 reported in their review posted on 3 October 2016, but have not been able to.*

Thank you for checking on this. A surface source isotope ratio of -12 ‰ would seem too high to us.

*Detailed Comments:*

*(1) l. 33: Why do you restrict "daytime" to 1000-1600 and "nighttime" to 2200-0600, omitting the hours in between?*

We have added the following explanation:

“Morning and evening transitional periods were omitted to avoid the confounding effects of sign change of the biological flux and sudden changes in the atmospheric stability regime.”

*(2) l. 162: Please describe how the standard gases were calibrated against NBS 19 (no hyphen) and NBS 20 (with space) and report the individual results of these measurements before conversion to the VPDB scale. What  $\delta(13\text{C})$  values (w.r.t. VPDB) did you use for NBS 20? The standard gases are a long way removed from the atmospheric value and virtually identical. This introduced significant scale uncertainties, see paper by Coplen et al. New guidelines for  $13\text{C}$  measurements. Anal. Chem. 78, 2439-2441.*

We apologize for a mixed-up here. NBS 19 and NBS 20 were used in an earlier study (Wen et al. 2013). In the present study, the reference materials were actually IAEA-CO-8 and IAEA-CO-9. We have added these details:

“... their  $\delta^{13}\text{C}$  values were measured with a mass spectrometer (MAT-253, Finnigan) using IAEA reference materials IAEA-CO-8 (-5.76 ‰ VPDB) and IAEA-CO-9 (-47.32 ‰ VPDB). The mass spectrometer measurements of these reference materials were  $(-5.81 \pm 0.04) \text{‰}$  and  $(-46.64 \pm 0.08) \text{‰}$  before normalization to the VPDB scale.”

Please refer to our response to Main point 1 regarding the mismatch between the calibration gas isotope ratio and the ambient isotope ratio.

(3)l. 169: *What do you mean by removing "40 3-minute data points"? Data over 3-minute intervals on 40 different occasions? Did this apply to all calibration gas runs? Were there only 40 calibration gas runs in the whole dataset? Perhaps your response could include a schematic or figure to illustrate what you have done.*

This sentence has been changed to:

“We removed the first 3 min of the data after switching to the ambient sampling mode from the calibration mode.”

(4)l. 170: *Why were these data removed?*

We remove the first 2-min data because the analyzer needed time to adjust to the step change after valve switching (Supplementary Figure S1).

(5)l. 204: *Where were the Mount Waliguan data obtained? Please give a reference to the data depository and state how the data were measured, by whom and which instrument was used.*

An online reference is added

(6) *The 2015 WLG data are now available, so the analysis should be performed with the actual data rather than some extrapolated version.*

We have updated this analysis using the actual observations at WLG.

(7) *What efforts have been made to ensure that the new data are compatible (i.e. on the same scale) as the WLG data. What is the uncertainty of the delta(13C) of the a) WLG data and b) the new data with respect to VPDB?*

The measurements at WLG are traceable to standards supplied by NOAA-ERL. The CO<sub>2</sub> concentration of our calibration gases was calibrated against three primary standards (with known mole fraction but unknown  $\delta^{13}\text{C}$ ) obtained from NOAA-ERL. One of us (Wen) has recently acquired  $\delta^{13}\text{C}$  gas standards from NOAA-ERL, but unfortunately our working standards had already been depleted by then.

We have added the following text to the Methods section:

“Table 1 lists the concentrations and their isotopic compositions of the standard gases used in this study. These gases were balanced in air. Their CO<sub>2</sub> mole fractions were measured with an IRIS analyzer (model G1101-i, Picarro Inc.) and calibrated against three primary standards obtained from NOAA-ERL which were traceable to the WMO 2007 scale reported by the Central Calibration Laboratory of the World Meteorological Organization, and their  $\delta^{13}\text{C}$  values were measured with a mass spectrometer (MAT-253, Finnigan) using IAEA reference materials IAEA-CO-8 (-5.76 ‰ VPDB) and IAEA-CO-9 (-47.32 ‰ VPDB).”

*Technical corrections:*

(1) *There should always be a space between value and unit, incl. per cent and per mill symbols, e.g. "-8.48 ‰", "13 ‰", "15 °C". There is no space before the degree sign if it refers to a geographic coordinate, but there is a space before the compass direction: "33° N". Please correct these errors throughout the manuscript.*

Changed throughout the whole paper.

(2) Please use SI units for mole fractions, i.e. " $\mu\text{mol mol}^{-1}$ ", not ppm, which is ambiguous.

Changed.

(3) Units and chemical symbols must not be mixed. The name of the quantity should identify the species of interest, i.e. "the CO<sub>2</sub> mass flux", and the unit should simply be " $\text{mg m}^{-2} \text{s}^{-1}$ " (e.g. l. 254).

Corrected

(4) Please note that in line with international conventions, indices should be enclosed in brackets after the quantity symbol, including  $\delta$  values, i.e. the correct notation is " $\delta(13\text{C})$ ". Unfortunately, in the past, the isotope geochemistry community has ignored this convention, but this is not a good reason to continue this poor practice. Alternatively, you might want to omit the index and just write " $\delta$ " because - at least in the context of the present manuscript - only <sup>13</sup>C/<sup>12</sup>C isotope ratios are considered. This makes also for a more concise terminology when you discuss the isotope delta of sources (Eq. 5), which would then be " $\delta_S$ " instead of " $\delta_S(13\text{C})$ ", see comment on line 97 below. As you are not qualifying the mole fraction symbol further in Eq. 1, I suggest you should omit the index "(13C)" entirely.

We now use the symbols you recommended, such as  $\delta_S$ ,  $\delta_F$ ,  $\delta_C$ , and  $\delta_a$ .

(5) The language used at a number of places in the manuscript is not as clear as it should be. Please make the following changes to rectify this. Terms like "<sup>13</sup>C isotope composition", "<sup>13</sup>C isotopic signature", "highly enriched <sup>13</sup>C signal", etc. do not make sense because they must be quantified by reference to the other stable carbon isotope, <sup>12</sup>C.

Corrected.

(6) l. 24: This is grammatically wrong. Please change to "Observations of mole fraction and carbon stable isotope composition (expressed as  $\delta(13\text{C})$ ) of CO<sub>2</sub> in urban ...".

Corrected

(7) l. 25: Add "and sinks" after sources.

Added.

(8) l. 26: Replace "C" with "carbon". Please make the same change everywhere else in the manuscript as well. Unnecessary abbreviations should be avoided for clarity and easier reading.

Replaced.

(9) l. 29 & 30: Please also give the standard deviations of mean mole fraction and  $\delta(13\text{C})$ .

Done.

(10)l. 31: Change word order to "highly  $^{13}\text{C}$ -enriched". Delete "the" before "Mount Waliguan".

Corrected.

(11)l. 34: Change to "we showed that the  $\delta(^{13}\text{C})$  of  $\text{CO}_2$  sources"

Changed

(12)l. 43: Change to "unique  $^{13}\text{C}/^{12}\text{C}$  ratios" or "unique  $\delta(^{13}\text{C})$  values" or "unique carbon stable isotope signatures".

Changed.

(13)l. 48: Replace "isotopic signature" with " $\delta(^{13}\text{C})$ ".

Replaced

(14)l. 49: Replace "heavier" with "higher".

Replaced.

(15)l. 51: Replace " $^{13}\text{C}$  composition" with " $\delta(^{13}\text{C})$ ".

Replaced

(16)l. 55: Replace " $\delta^{13}\text{C}$ " with "carbon stable isotope".

Replaced

(17)l. 56: " $^{13}\text{C}$  signature" - see l. 43.

Replaced

(18)l. 74, 87, 96, 123, 145: Delete "its" (mole fractions don't have  $\delta(^{13}\text{C})$  values).

Deleted

(19)l. 97 & elsewhere: Wrong order of indices - this should be  $\delta_{\text{S}}(^{13}\text{C})$ .

Corrected

(20)l. 114: Either add the index " $^{13}\text{C}$ " to the delta symbols on the left hand side of the equation or omit it from the right hand side.

Changed.

(21)l. 115: "at a background site"

Edited

(22)l. 116: Please rephrase. Mole fractions don't have isotopic compositions, and there is no such thing as a "<sup>13</sup>C isotopic composition", also see comments above.

Changed to “mole fraction and δ<sup>13</sup>C value of CO<sub>2</sub>” throughout the paper.

(23)l. 125: "built-up area"

Edited.

(24)l. 127: Change to "190 million" and add a space before the compass direction (e.g. "33.41° N").

Edited

(25) Replace hyphens with to, e.g. "29.04° to 33.41° N".

Replaced

(26)l. 129: Rephrase to "The main vegetation types are all C3 species".

Rephrased

(27)l. 150: I find it hard to believe that there was no other anthropogenic CO<sub>2</sub> source within a 3 km radius. Are there no houses? No heating system? No humans? No cars?

This sentence has been changed to “There was no large industrial CO<sub>2</sub> source in the 3 km radius...”

(28)l. 152: Please delete the "~" (tilde) sign and give an accurate distance instead (rounded and/or with uncertainties, if necessary). In any case, the "~" sign is not the correct symbol to designate approximations (for which "≈" should be used).

Corrected

(29)l. 162: Please describe

Please refer to detailed comments 2 and 7.

(30)l. 173: Do you mean "high bias" or "high error"? And if bias, do you mean "biased high" (i.e. too large) or "highly biased" (i.e. biased a lot, but unclear in which direction)? How much is the bias (or error)?

Changed to “is biased high”

(31)l. 181: Do you mean relative humidity? In any case, the symbol "V" before "%" must be omitted.

Use SI units, i.e. 0.66 cmol/mol, or 0.66 x 10<sup>(-2)</sup> m<sup>3</sup>/m<sup>3</sup>, for mole fractions (or volume fractions).

Edited. This is now referred to as “water vapor mole fraction”

(32)l. 181: Please replace "<sup>13</sup>C composition" with "<sup>13</sup>C/<sup>12</sup>C ratio".

Edited

(33)l. 185: Which one is it? A volume fraction? Or a mole fraction? Do you make a correction for the non-ideality of the gases involved?

It is mole fraction. We did not correct for non-ideality.

(34)l. 188: This equation is wrong (or at least inconsistent with the text). The second term on the right hand side should most likely be 0.46 ‰ (H/% - 2.03).

Edited

(35)l. 189: Please delete "in percent". Physical quantities are not limited to one specific unit.

Deleted

(36)l. 190: Please change "isotopic composition" to "isotope delta" (two occurrences).

Changed

(37)l. 193: has -> have

Changed

(38)l. 253: Replace <sup>13</sup>C isotope composition with an adequate term - see comments above.

Replaced as “ $\delta^{13}\text{C}$  value”

(39)l. 255: Units and chemical symbols should not be mixed. If you want to specify the species the fluxes refer to, it should be enclosed in brackets, e.g.  $F_s(\text{CO}_2)$ , or you could explain in the text that "F" relates to CO<sub>2</sub> mass fluxes. The unit should be written as "mg m<sup>-2</sup> s<sup>-1</sup>".

Changed.

(40)l. 258: "unknowns to be solved for"

Edited

(41)l. 264: adapt -> adopt

Edited

(42)l. 264: State the value used for delta(<sup>13</sup>C) of CO<sub>2</sub> released during cement production, and its uncertainty. The uncertainty should be propagated through the flux calculation.



Done.

(43)l. 264: *You should calculate an uncertainty for  $\Delta P$  and propagate it to your flux calculation.*

Done.

(44)l. 271: *What is the uncertainty of this value?*

The 20 % value is based on an eddy covariance observation. The nighttime  $F_F$  is also assumed to have an uncertainty of 10 %.

(45)l. 288-290: *What are the standard deviations of these values?*

Information added.

(46)l. 292: *Replace "diurnal" with "diel". Diurnal means "during the day"; "diel" means "over 24 hours".*

Replaced.

(47)l. 293: *Define the winter season - which months?*

Defined (December to February).

(48)l. 301: *Value and uncertainty must be enclosed in brackets so that the unit symbol applies to both, i.e.  $(-24.75 \pm 0.26)$  ‰. Please revise this and similar occurrences throughout the manuscript.*

All checked

(49)l. 301: *bound -> limit*

Edited

(50)l. 307-321: *This whole section is missing uncertainties and their propagation through the calculation.*

Uncertainty ranges are now provided.

(51)l. 316: *The delta differences have been calculated incorrectly. These should be 1.53 ‰ and 0.53 ‰ because the delta scale is not linear.*

This comparison has been removed. (We have replaced Figures 4 and 5 with plots of the data obtained in January 2014.)

(52)l. 319: *What do you mean by "linear correlation"? Is that the slope? Please give the full result of the regression (slope and intercept) including uncertainties of slope and intercept and*

state the  $R^2$  value.

These are linear correlation coefficients. The detailed regression statistics are now added.

(53)Section 3.3: Again uncertainty estimates are missing as well as their propagation.

The uncertainty ranges are now included in Tables 2 and 3.

(54)l. 349, 449, etc.: There should be a space between "m-2" and "s-1".

Changed.

(55)l. 369: "consistent"

Edited.

(56)l. 379: What do you by "varied 70% to 80% in winter and about 50% in summer"? In the remainder of the paragraph you use fractional comparison between winter and summer.

Changed to "In Chicago, natural gas usage comprises 70 % to 80 % of total fossil fuel consumption in the winter and about 50 % in the summer (Moore and Jacobson 2015)"

(57)l. 421/2: "monthly mean" (x 2)

Edited.

(58)l. 422: "WLG value"

Edited.

(59)l. 459-467: This MC analysis omits the uncertainties in the fluxes  $F_F$  and  $F_C$  and the isotope deltas  $\delta_S$  and  $\delta_C$  and should be improved by including them.

This analysis has been updated with the uncertainties in FF and Fc. The uncertainties in the partitioned fluxes are now shown (Figures 7 and 8).

(60)l. 493: The correlation coefficient is not a meaningful figure of merit to compare the "goodness of fit" by a Keeling plot regression and by a Miller-Tans regression because they plot different variables on the axes. It would be more useful to state the resulting uncertainties of the source  $\delta$  values,  $\delta_S(13C)$ .

We have deleted this portion of the text. Thank you.

(61)l. 853: "diel variation"

Edited

(62)Figure 1: x-axis label should be  $x(H_2O)/\%$  or  $y(H_2O)/\%$ .

We kept the original x-axis label but changed the independent variable in Equation 2 to “H<sub>2</sub>O” to be consistent with this data plot.

*(63)Figure 5: x-axis label.*

Edited

*(64)Figs. 7 & 8: The lack of variability in  $F_F$  seems to be implausible. What variability would you expect? Please indicate this on the plot and include it in the uncertainty propagation.*

All the variables are now given an uncertainty range.

## Response to Referee 2

*This paper uses continuous measurements of the stable isotopic composition of carbon ( $\delta^{13}\text{C}$ ) in  $\text{CO}_2$  to investigate  $\text{CO}_2$  emissions in the city of Nanjing and the surrounding areas, taking advantage of differing footprints for night (Nanjing) and day (surrounding area) times of day. The most important observation is that some of the  $\delta^{13}\text{C}$  values are higher than background. The only mechanism to produce this is by incorporating emissions from cement production, which is characterized by very high  $\delta^{13}\text{C}$  values of +0.20 ‰.*

### *Main concerns 1)*

*The major concern in this paper is that the authors are using the Miller-Tans (2003) approach to determine the average monthly  $\delta^{13}\text{C}$  of the sources for daytime and nighttime. In that formulation, the background values of  $\text{CO}_2$  mole fractions and  $\delta^{13}\text{C}$  are included in the linear equation, with the slope being the  $\delta^{13}\text{C}$  of the high $\text{CO}_2$  endmember. The equation for this analysis is:*

$$\delta_{\text{obs}} - \delta_{\text{bg}} = \delta_s (\text{C}_{\text{obs}} - \text{C}_{\text{bg}})$$

*where  $\delta_{\text{obs}} - \delta_{\text{bg}}$  is plotted against  $(\text{C}_{\text{obs}} - \text{C}_{\text{bg}})$ . There should be no intercept determined during the analysis. However, the plots shown in Figures 4 and 5 show significant non-zero intercepts. I have looked at the data from the supplement, together with the NOAA data from WLG, which is being used as background in this new version, and found that the intercepts for the nighttime data for 2013-2014 should be  $\sim -12\text{‰}$  with a zero intercept, instead of  $-24\text{‰}$  as shown in Figure 5. My analysis was approximate, in that I used a smoothing spline for interpolating the WLG data, instead of the Thoning et al. (1989) smoothing code. However, these are very different values and should affect the fluxes being calculated. At minimum, the meaning of the intercept values should be discussed, as well as why it is calculated.*

[Please refer to our response to the editor](#)

### *Specific comments:*

*(1) Line 35 and often throughout: Give uncertainties or standard deviations of ranges.*

[Added](#)

*(2) Line 120: After “(Miller et al. 2003)” add “and was used in an urban environment by Newman et al. (2016).”*

[Added](#)

*(3) Line 121: Insert “continuous measurements in” between “to” and “an urban environment.”*

[Added](#)

*(4) Line 138: You have justified using the Miller-Tans method earlier in the Introduction. There is no reason to discuss the performance of the Keeling plot method. Also, you do not show any Keeling plots. This discussion is left over from the previous version of the manuscript.*

[We now include the Keeling plot results \(Figure 6; Supplementary Table S2\).](#)

(5)Lines 160–162: *The carbon isotopic compositions of the two standards (Table 1) are very similar and very far from the values of the ambient measurements. Can you show that there are no extrapolation uncertainties?*

Please refer to our response to the editor.

(6)Lines 287–289: *What does it mean to say that  $\delta^{13}\text{C}$  values display a larger seasonal cycle than do  $\text{CO}_2$  mole fraction values? The latter is  $\sim 20$  ppm, whereas the former  $\sim 2\%$ . Are you thinking about a relative fraction?*

This sentence is changed to “The  $^{13}\text{C}/^{12}\text{C}$  ratio of atmospheric  $\text{CO}_2$  displayed a more clear seasonal cycle than the mole fraction.”

(7)Lines 299–321: *The Miller-Tans (2003) method assumes a zero intercept. What is the meaning of your intercept, and how does it affect the interpretation of the slope?*

Please refer to our response to the editor.

(8)Lines 311 –321: *What is the significance of these observations? Are they important enough that you should show figure(s) with the correlations?*

We have added this sentence:

“These correlation patterns suggest that atmospheric  $\delta^{13}\text{C}$  was influenced more by surface sources at the regional scale than at the local (city) scale.”

(9)Lines 348–355: *A lot of values are given – what are the uncertainties?*

The uncertainties of these values are now given.

(10)Lines 369–370: *Replace with: “to 2013, which was consistent with the seasonal variation of background and transport of emissions from fossil fuel combustion (Newman et al. 2016).”*

Replaced

(11)Line 418: *“Park Falls”*

Edited

(12)Line 420: *“fossil fuel combustion which has low  $^{13}\text{C}$  contents.” This depends on the nature of the fossil fuel component. As shown in Table 2, gasoline combustion produces  $\text{CO}_2$  with  $\delta^{13}\text{C}$  very similar to the biosphere, and combustion of coal produces  $\text{CO}_2$  with values higher than the biosphere.*

This sentence has been changed to “...the plant photosynthetic enhancement of  $^{13}\text{C}$  is offset by the  $\text{CO}_2$  from fossil fuel combustion which has lower  $^{13}\text{C}/^{12}\text{C}$  ratios than the atmosphere.”

(13)Lines 477–504: *Remove this section. It is left over from the previous version and no longer necessary*

We now present the Keeling results.

### Response to Referee 3

*The authors made an obvious effort to improve the paper, and addressed most of my concerns reasonably well. There are still some weak points, and I realize that the work needed to fix some of them may be not possible anymore. I list my last concerns below and I leave it to the editor to decide if any of them is important enough to delay the publication.*

#### *Main concerns 1)*

*The accuracy is still not addressed in the answer or in the paper (but I do realize that this may be difficult with the information available). The precision for 1-hr intervals is specified; however, the calibration is done using 2-min averaged data, which will have lower precision; in this case the dataset could show 3-hourly steps due to the calibration.*

[Please refer to our response to the editor.](#)

#### *Main concerns 2)*

*The final NOAA data for 2015 for WLG are available. Did the authors at least check whether their extrapolated data for 2015 are somewhat similar to the real data from NOAA? (this should be easy)*

[Done. Thank you.](#)

#### *Main concerns 3)*

*the two sets of calibration gases listed in Table 1 have identical isotope values – please check if this is correct*

[This is because the two sets came from the same mixtures of CO<sub>2</sub> and air.](#)

#### *Main concerns 4)*

*I still find the estimation of uncertainties of the main results poor, and also the answers to the reviewers questions related to this. When comparing average mole fractions or isotope values (for examples Nanjing with WLG), the uncertainty should be given, in order to decide if the values are significantly different or not. For the final flux results, the Monte Carlo simulation estimates the influence of a 1‰ error in  $\delta^{13}C_p$  and  $\delta^{13}C_f$ , but does not account for the other variables that contribute to the overall uncertainty ( $F_f$ ,  $F_c$ ,  $\delta^{13}C_s$ ,  $\delta^{13}C_c$ ). Also, the results are only discussed in text as overall averages; when looking at Figs 7 and 8, it is not clear without errors shown which of the values are different from zero and if any of the variability is significant.*

[Please refer to our response to the editor.](#)