

Review of revision of “Interpreting the  $^{13}\text{C}/^{12}\text{C}$  ratio of carbon dioxide in an urban airshed in the Yangtze River Delta, China

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In this latest revision, the authors have used a modification of the Miller-Tans (2003) approach to interpret the stable carbon isotopic composition of  $\text{CO}_2$  emissions from Nanjing and the greater Yangtze River Delta in China. In this modification, they use background  $\text{CO}_2$  mole fractions from CarbonTracker for each data point, but they do not assume any  $\delta^{13}\text{C}$  value, which becomes embedded in the intercept of the correlation line. The slope gives the composition of the high- $\text{CO}_2$  mixing end member, as for the original Miller-Tans approach. The authors compare results between this calculation and the Keeling Plot method in which neither the mole fraction nor the  $\delta^{13}\text{C}$  of the background are known but are assumed constant over the duration of the sampling period. It is unclear what their conclusion is from the comparison.

My major concern at this time is the standards used to calibrate the Picarro  $\text{CO}_2$  Isotopic Analyzer (G1101-i) for  $\delta^{13}\text{C}$ . Two standards with differing  $\text{CO}_2$  mole fractions are run, but the  $\delta^{13}\text{C}$  values are within uncertainty and are very different from the ambient measurements. Citing Bowling et al. (2005), the authors say “because the system measures the concentrations of the major and minor isotopologue independently, it is not critical that the calibration standard and the measurement target have matching isotope ratios.” However, in the Bowling et al. paper the standard values are much closer to those being analyzed, and Bowling et al. (2005) say that it is “not critical that the calibration gases differed in isotope ratio”, not that the calibration standards differ from the target air. The method used here, by Xu et al., assumes linearity over a very wide range. Perhaps the authors could borrow a couple of standard tanks to determine the calibration line for the instrument over this wide range of compositions and then use the temporal variations of their standards to adjust for drift of the instrument, as was done by Verhulst et al. (2016) for  $\text{CO}_2$  mole fraction.

Verhulst, K. R. et al., 2016, Carbon Dioxide and Methane Measurements from the Los Angeles Megacity Carbon Project: 1. Calibration, Urban Enhancements, and Uncertainty Estimates: Atmospheric Chemistry and Physics Discussions, p. 1–61, doi:10.5194/acp-2016-850-SC1.

Specific comments:

Line 65: “configuration” should be plural “configurations”

Line 83: Add “isotope ratio” before “mass-spectrometry” and change “MS” to “IRMS”

Line 96: The Verdag et al. (2016) study is for simulated data only, not measurements.

Line 138: “method” should be plural “methods”

Line 167: Are you sure that these values are “before” normalization? They are probably on the VPDB scale. If not, please convert to VPDB.

Line 186: Add “the average” after “if”.

Line 188 and elsewhere: There are spurious “!” symbols. Please remove.

Lines 214-216: What results do you get if you force the correlations through zero?

Lines 230-232: Newman et al. (2016) gives all monthly Miller-Tans plots for 2011, forced through zero and no intercepts are obvious. No intercepts are required for the monthly plots over 8 years of data.

Line 239: Since you have  $C_b$  from CarbonTracker, you can calculate  $\delta_b$ . Are the values reasonable? If not, then there is a problem with assumptions.

Line 243: At the end of the paragraph, insert a statement such as: “We call this a modified Miller-Tans analysis, which requires knowledge of  $CO_2$  mole fraction, but not  $\delta_b$ .”

Line 263: Replace “at” with “for”.

Line 291: Is  $F_P$  the net biological flux? If so, it might be better to use “ $B_{IO}$ ”, since “ $P$ ” suggests photosynthesis, but this is photosynthesis + respiration.

Line 303: Insert “value for” before “ $\delta_F$ ” and “the” after “was”.

Line 307: “an U-shaped” should be “a U-shaped”

Line 322: “expression” should be plural “expressions”

Lines 322-330: How do the Monte Carlo analysis errors compare to an average explicit error propagation calculation?

Lines 343-344: Are these standard errors or standard deviations? Standard errors are probably more appropriate here.

Line 352: “Contrary” should be “In contrast”.

Line 357: Insert “modified” before “Miller-Tans”.

Line 367: Delete “)” after “1.79”.

Line 383: Insert “combustion” after “fossil fuel”.

Line 384: Insert “manufacturing” after “pig iron”.

Lines 434-442: Comparison with Paris (Widory and Javoy, 2003), Los Angeles (Newman et al., 2016)?

Line 450: Add “of” after “difference”.

Line 454: Add “of” after “difference”.

Lines 508-510: What is the  $R^2$ ?

Line 533 “method” should be “methods”

Lines 536-538: Where does the value of 0.21 ‰ come from? Is this for all sources, both anthropogenic and biogenic? And is this difference significant?

Line 539: “Table 2” should be “Table 3”.

Line 549: What is your interpretation of the bias between the two methods (Table S2)? What is your conclusion?

Line 553: Replace “If” with “When”.

Line 557: The value of 0.21 ‰ seems much lower than the monthly differences observed in Fig. 6b.

Line 571: Insert “modified” before “Miller-Tans”.

Line 929: How does the background you used compare to the WLG data for  $CO_2$  mixing ratio?

Line 937: Insert “modified” before “Miller-Tans”.

Lines 966-967, Table 1: Are these measurements from IRMS and G1301?

Line 976: “uncertain range” should be “uncertainty”