

Dear Drs Gromov and Brenninkmeijer

many thanks for sending your revised manuscript. Unfortunately, I am not able to accept it for publication because various unclear aspects of the manuscript that I pointed out before, but which have still not been resolved. These are minor revisions, so I will oversee them myself.

Best regards

Jan Kaiser

Corrections (referring back to file acp-2014-598-author_response-version5.pdf)

unambiguous: As noted before the term "unambiguous(ly)" implies it is not open to other interpretations, which goes against the grain of scientific principles. If you wanted to make the statement stronger, you could say it's "very likely", maybe even "virtually certain". As you acknowledge yourself in l. 142 "Such sampling-induced mixing renders an unambiguous determination of the artifact source isotope signature rather difficult because neither mixing nor isotope ratios of the admixed air portions are known sufficiently well". And on line 354, you strive "more unambiguous verification". Unambiguous implies 100 % certainty, so it can't be more unambiguous. Would you please revise your manuscript to reflect the actual level of (un)certainty, along the lines suggested above?

tautology: If, as you say the observations hold for all observations below 400 nmol/mol, i.e. including tropospheric conditions, than this implies the tautological statement "tropospheric CO mixing ratios are largely affected by varying tropospheric [CO]". I agree that OH concentrations in addition to CO emissions affect to emissions affect [CO], so you might want to include both in your revised manuscript. In line with your comment on bins, this should probably be 390 nmol/mol as you don't resolve individual values between 390 and 410 nmol/mol (in Figure 1b, which is the one being discussed in this paragraph).

O₃ mole fractions: Please change ">" to "≥" in the suggested correction. In line with your comment on bins, this should probably be 390 or 410 nmol/mol, though.

580-600 nmol/mol bin: Please amend to 570-590 nmol/mol bin, or whichever bin you refer to.

l. 160-162: I accept that you would like to adopt a level of redundancy, but it is important that the statements are clear and the second sentence is not. If you would like to retain this sentence, please clarify which C1 and WAS data (presumably [CO] and $\delta^{18}\text{O}(\text{CO})$?) you refer to and what "of the same nature" refers to.

Keeling plot (l. 223 and 348): As you can see in Keeling (1958), he only measured the CO₂ mole fraction and $\delta^{13}\text{C}(\text{CO}_2)$, not the mole fraction of added CO₂. Could you please change the sentence to reflect the use of the added selection criterion for the range of samples you apply the Keeling plot to? I also noted an additional error that appeared between versions 4 and 5 of the manuscript: $[\text{CO}]_c$ in the numerator of Eq. (4) should instead be $[\text{CO}]_t$.

Keeling (1958) The concentration and isotopic abundances of atmospheric carbon dioxide in rural areas. *Geochim. Cosmochim. Acta* 13, 322-334

Notation (l. 253, Fig. 6 and elsewhere): I suggested a notation that avoids the appearance of C_c , C_a , O_t , O_c , e.g. $\delta^{13}\text{C}(\text{CO}, c)$. The notation should be consistent, not distinguishable because it is confusing to use different samples for the same quantity. If you need to distinguish further between measure and modeled isotope deltas, you could use additional labels, e.g. $\delta^{13}\text{C}(\text{CO}, m)$. However, I don't think this is necessary because you actually use the measured concentrations in the mixing calculations.