

We want to thank John Miller for the review of our manuscript and his helpful suggestions for improving this study. Our replies are marked in blue.

Review of Maier et al, “Uncertainty of continuous  $\Delta\text{CO}$ -based  $\Delta\text{ffCO}_2$  estimates derived from  $^{14}\text{C}$  flask and bottom-up  $\Delta\text{CO}/\Delta\text{ffCO}_2$  ratios” by John Miller

## General comments

Overall, this is a very good study focused on developing high frequency proxies (here CO) for the estimation for the recently added fossil content of atmospheric  $\text{CO}_2$  measurements. The writing is generally very good and the figures are excellent, reflecting the analysis itself. The pdf has numerous comments but I will highlight two here:

1. Although this is clear in the title, use of “Delta(CO)-based DffCO<sub>2</sub> estimates”, when used without explanation, has the potential to be highly misleading because these estimates (the atmospheric data-based ones the paper shows to be trustworthy) are still based on D<sup>14</sup>C I’m not exactly sure of the solution, but perhaps you can employ nomenclature/notation that identifies such values as ‘calibrated’ by <sup>14</sup>C.

We agree, that this can lead to confusion. We tried to make this clearer in our manuscript, e.g. by using “proxy-based  $\Delta\text{ffCO}_2$ ” in the abstract, as you suggested. However, in the main text we would like to use “ $\Delta\text{CO}$ -based  $\Delta\text{ffCO}_2$ ” because this is also what we used in the companion paper (Maier et al., 2023). We also added some text in the introduction, to make it clear, that the “ $\Delta\text{CO}$ -based  $\Delta\text{ffCO}_2$ ” is calibrated with <sup>14</sup>C. (e.g., p. 3, l. 71-72; p. 3, l. 90; p. 5, l. 137)

2. I have a few questions about the TNO inventory that could benefit by a bit more investigation and explanation. First, it appears that TNO includes biofuels such as wood. But what about ethanol and biodiesel? Generally, can the fossil components of the TNO inventory be isolated for a more direct comparison with <sup>14</sup>C-based observations? Second, in investigation of the point source impacts for Heidelberg, can you transport the non-point-source sectors to see how much the match to data is improved – i.e., is the mismatch mainly due to the ratio of (dilute) point to area sources in TNO or mainly due to incorrect emission ratios for the area sources?

Thank you for your suggestions. The TNO inventory distinguishes between fossil fuel and biofuel  $\text{CO}_2$  and CO emissions. Thus, TNO includes emissions from wood-fired heating as well as biofuel emissions from the traffic sector. For the traffic sector this is based on national reporting of shares of ethanol in gasoline and/or biodiesel in diesel. Moreover, there is also data on the amount of biomass co-fired in coal-fired powerplants. This is also in the inventory as  $\text{CO}_2$  from biofuel.

We carried out both of the simulations you suggested (see Fig. 1 below). The magenta dots in Figure 1a show the contributions from the non-point-source sector only. They lead to an average ratio of about 6 ppb/ppm, which is well below the observed ratio (grey crosses). From that we conclude that TNO might underestimate the ratio of the area source emissions in the Rhine Valley. Furthermore, a potential wrong dilution ratio between point sources and area sources (both with correct emission ratios) can't explain the differences between observed and modelled  $\Delta\text{CO}/\Delta\text{ffCO}_2$  ratios, because both, the average modelled ratio from only-area sources (6.0 ppb/ppm) and the ratio from only-point sources (1.2 ppb/ppm) are below the observed ratio of 8.44 ppb/ppm.

Fig. 1b shows the simulated contributions from the traffic (orange) and heating (cyan) sectors. The traffic (biofuel plus fossil fuel) sector leads to an average  $\Delta\text{CO}/\Delta\text{ffCO}_2$  ratio of  $7.72 \pm 0.08$  ppb/ppm ( $R^2=0.93$ ), which is less than 10% lower compared to the observed average ratio of 8.44 ppb/ppm. However, the heating (wood plus fossil) sector leads to a much lower average  $\Delta\text{CO}/\Delta\text{ffCO}_2$  ratio of  $3.36 \pm 0.09$  ppb/ppm ( $R^2=0.65$ ). This might indicate that the  $\text{CO}/\text{ffCO}_2$  emission ratios from the TNO heating sector are too low in the main footprint of Heidelberg. This could be explained, for example, by an incorrect distribution of the use of fossil and bio fuels in the heating sector.

We included these plots and a discussion in our manuscript. (Fig. 3b and A3 in the manuscript; p. 12, l. 313-314; p. 18-19, l. 474-499)

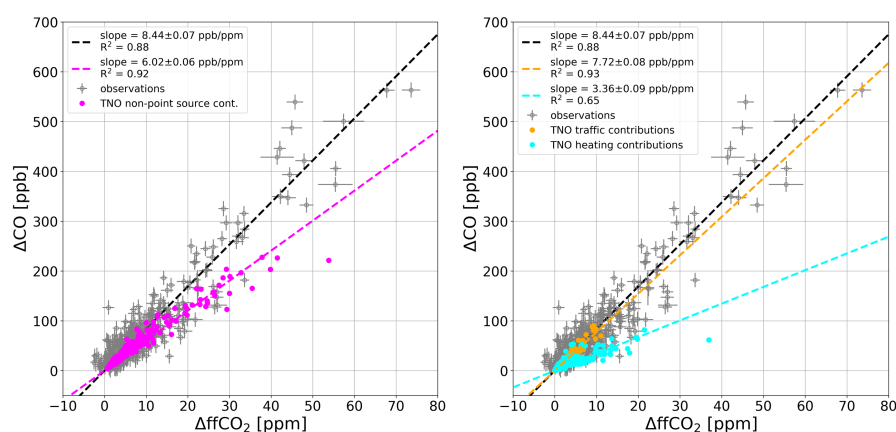


Figure 1:  $\Delta\text{CO}$  and  $^{14}\text{C}$ -based  $\Delta\text{ffCO}_2$  observations from the Heidelberg flasks (in grey) and (a) simulated non-point-source  $\Delta\text{CO}$  and  $\Delta\text{ffCO}_2$  contributions for the flask events (magenta), and (b) simulated  $\Delta\text{CO}$  and  $\Delta\text{ffCO}_2$  contributions from the traffic (orange) and heating (cyan) sectors for the flask events.

## Specific comments

Suggested edits and comments are embedded in the manuscript .pdf. Blue highlights indicate those that are language oriented and yellow for science/conceptual issues.

Thank you for your comments. We directly replied to them in the .pdf document.

## References:

Maier, F. M., Rödenbeck, C., Levin, I., Gerbig, C., Gachkivskyi, M., and Hammer, S.: Potential of  $^{14}\text{C}$ -based versus  $\Delta\text{CO}$ -based  $\Delta\text{ffCO}_2$  observations to estimate urban fossil fuel  $\text{CO}_2$  ( $\text{ffCO}_2$ ) emissions, EGUsphere [preprint], <https://doi.org/10.5194/egusphere-2023-1239>, 2023.