

Authors' comments to reviews of the ACP MS No.: egusphere-2023-728

Sources and Long-term Variability of Carbon Monoxide at Mount Kenya and in Nairobi

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Overview

We thank the referee 2 and the Editor for their in-depth assessment of our manuscript and overall supportive comments and constructive suggestions, which have helped to further improve the manuscript.

All issues raised by the referee are addressed below, organized such that first the referee/editor comments are given in *italic*, directly followed by our response in **blue color**. The resulting edit in the revised manuscript are given in **bold green**. References in our responses to page and line numbers in the manuscript refer to the revised version.

Responses to Anonymous Referee #2 comments (RC2)

This is a clearly written and easy to understand paper showing multi year measurements of CO and its isotopes from Mount Kenya with some contrasting measurements from Nairobi also shown. The introduction is comprehensive and clear, and the methods (as far as measuring CO and its isotopes) are also clear with details not covered referred to previous papers. The Results and Discussion section presents a nice discussion of the isotopic analyses but the ambient CO observation discussion is less clear with details put into the Supplementary section that to me seem like they belong in the main text.

Overall this study represents an important contribution in an undersampled region of the world and I recommend that the paper is published after some revisions which I consider minor as the required extra analysis and text is not onerous or particularly time consuming.

We thank Referee #2 for the overall positive assessment of our work, and suggestions for improvements. All comments and suggestions have been considered, and especially the discussions on the ambient CO observations expanded.

Comments on this section:

- *Page 6 – “the seasonal variations are not pronounced” – This is actually hard to judge from what you present/ Why not show a combined seasonal average? Eyeballing the S1 figure upper panel I get a strong impression that there is a fairly consistent low in CO mole fraction around May with peaks around March and August. Sure there’s some variability in timing but this looks like a seasonal variation not just random to me. In fact, this bimodal peak feature is explained by the back-trajectories in Figure 2 and explained by in the last paragraph of section 3.1 – with biomass burning from different areas bought by the changing meteorology in different seasons.*

We agree also that the CO measurements at the Mt. Kenya GAW station reveal an annual cycle, based on monthly averages, characterized by peak concentrations during the dry periods as shown in the **revised Figure 1**, and highlighted in the revised manuscript.

Chapter 3.1, Lines 187-188: ‘Overall, peak CO mole fractions were observed during the dry periods.’

- *However these backtrajectories are presented without any explanation of the timeframe that they represent in either the text or the figure caption.*

Well noted. We have included the timeframe, December 2020 – November 2021, in the revised manuscript (**Figure 2**).

- “no clear multi-year trend..”. No details are given are what trend analysis was undertaken and on what data. Did you use daily mean values? All data or median values. What sort of trend analysis was done? A value of approximately zero is stated but with no uncertainty estimate given.

Thank you for highlighting this omission on our part. Initially, our assessment of the long-term trend was based on simple linear regression. Further assessment following an approach by (Thoning and Tans, 1989) reveal a small, but statistically significant, overall decadal positive trend of 6.7 ± 0.4 ppb/10yrs (SI Figure S). Here, a fit function that includes a linear term, a quadratic term as well first and second harmonics was applied on the the daily aggregated CO data for 2002 – 2021. However, we acknowledge that the data gaps and different measurement techniques for the different periods of data availability. We have provided more details in the revised manuscript.

Abstract: ‘While some data gaps and differences in instrumentation complicate decadal-scale trend analysis, a small long-term increase is resolved.’

Chapter 3.1, Lines 189 – 202: ‘Assessment of the long-term CO trend, following the approach by (Thoning and Tans, 1989), reveal a small but statistically significant positive decadal trend of 6.7 ± 0.4 ppb/10yrs. This statistical model is based on a fit function that includes a linear term, a quadratic term, as well as first and second harmonics. For comparison, simple linear regression gives a similar decadal rate of 6.2 ± 0.6 ppb/10yrs (for uncertainty estimation, see (Kirago et al., 2022). Like many types of environmental data, the present CO data display a lognormal-like concentration distribution, suggesting influence by exponential processes such as sink kinetics (Andersson, 2021). This may influence trend analysis. Similarly to linear regression, regression of log-transformed data also gives a significant positive rate, which suggests that the skewed concentration profile has little influence on trend estimation. However, given the large data gaps and different measurement techniques, such interpretations should not be over-emphasized. Nevertheless, the increasing trend here constrained for ground observations of CO is qualitatively consistent with satellite retrievals and model estimates for sub-Saharan Africa (Hedelius et al., 2021; Zheng et al., 2019; Buchholz et al., 2021).’

Conclusions, Lines 304-307: “Although the data gaps in CO mixing ratios and mixed instrumentation complicates detailed analysis, a small decadal increase of 6.7 ± 0.4 ppb/10yrs was resolved for the Mt. Kenya GAW station, in agreement with satellite observations and emission

inventories for the Sub-Saharan region (Buchholz et al., 2021; Hedelius et al., 2021; Zheng et al., 2019).”

Page 7: “The stable isotope composition of COvaried temporally and inversely with CO”. This is stated but not shown (perhaps point to Keeling plots as evidence?). What was the correlation factor – with CO ambient mole fractions?

The stable isotope composition of CO varied inversely to 1/CO (Keeling relation), that is, proportional to CO mole fractions (for example at Mt. Kenya, $R^2 = 0.67$ for $\delta^{18}\text{O}$ and $R^2 = 0.39$ for $\delta^{13}\text{C}$). This typo has been rectified. In addition, the temporal variation of CO concentrations and isotope composition in Nairobi and at Mt. Kenya is presented in **SI Figure S2**.

Chapter 3.2, lines 234-236: ‘The stable isotope composition of CO ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) for ambient samples from Mt. Kenya GAW station during August 2021 varied temporally with the CO mole fractions (SI Figure S2).’

- *Although Keeling plots are now text book stuff – I think a reference would be appropriate? Perhaps in sentence stating that the linear relation implies two-component mixture?*

Thank you for pointing out this. We have included two references describing and applying this approach, including the original reference, (Keeling, 1958)

Chapter 3.2, lines 245-247: ‘This implies that the CO dynamics in this system can be described by a two-component mixture; a relatively stable background fraction and a regional varying source (Keeling, 1958; Dasari et al., 2022).’

References

Andersson, A.: Mechanisms for log normal concentration distributions in the environment, *Sci Rep*, 11, 1–7, <https://doi.org/10.1038/s41598-021-96010-6>, 2021.

Buchholz, R. R., Worden, H. M., Park, M., Francis, G., Deeter, M. N., Edwards, D. P., Emmons, L. K., Gaubert, B., Gille, J., Martínez-Alonso, S., Tang, W., Kumar, R., Drummond, J. R., Clerbaux, C., George, M., Coheur, P. F., Hurtmans, D., Bowman, K. W., Luo, M., Payne, V. H., Worden, J. R., Chin, M., Levy, R. C., Warner, J., Wei, Z., and Kulawik, S. S.: Air pollution trends measured from Terra: CO and AOD over industrial, fire-prone, and background regions, *Remote Sens Environ*, 256, 112275, <https://doi.org/10.1016/j.rse.2020.112275>, 2021.

Dasari, S., Andersson, A., Popa, M. E., Röckmann, T., Holmstrand, H., Budhavant, K., and Gustafsson, Ö.: Observational Evidence of Large Contribution from Primary Sources for Carbon Monoxide in the South Asian Outflow, *Environ Sci Technol*, 56, 165–174, <https://doi.org/10.1021/acs.est.1c05486>, 2022.

Hedelius, J. K., Toon, G. C., Buchholz, R. R., Iraci, L. T., Podolske, J. R., Roehl, C. M., Wennberg, P. O., Worden, H. M., and Wunch, D.: Regional and Urban Column CO Trends and Anomalies as Observed by MOPITT Over 16 Years, *Journal of Geophysical Research: Atmospheres*, 126, 1–18, <https://doi.org/10.1029/2020JD033967>, 2021.

Keeling, C. D.: The concentration and isotopic abundances of atmospheric carbon dioxide in rural areas, *Geochim Cosmochim Acta*, 13, 322–334, [https://doi.org/10.1016/0016-7037\(58\)90033-4](https://doi.org/10.1016/0016-7037(58)90033-4), 1958.

Kirago, L., Gustafsson, Ö., Gaita, S. M., Haslett, S. L., deWitt, H. L., Gasore, J., Potter, K. E., Prinn, R. G., Rupakheti, M., Ndikubwimana, J. de D., Safari, B., and Andersson, A.: Atmospheric Black Carbon Loadings and Sources over Eastern Sub-Saharan Africa Are Governed by the Regional Savanna Fires, *Environ Sci Technol*, <https://doi.org/10.1021/acs.est.2c05837>, 2022.

Thoning, K. W. and Tans, P. P.: Atmospheric carbon dioxide at Mauna Loa Observatory. 2. Analysis of the NOAA GMCC data, 1974–1985, *J Geophys Res*, 94, 8549–8565, <https://doi.org/10.1029/JD094iD06p08549>, 1989.

Zheng, B., Chevallier, F., Yin, Y., Ciais, P., Fortems-Cheiney, A., Deeter, M. N., Parker, R. J., Wang, Y., Worden, H. M., and Zhao, Y.: Global atmospheric carbon monoxide budget 2000–2017 inferred from multi-species atmospheric inversions, *Earth Syst Sci Data*, 11, 1411–1436, <https://doi.org/10.5194/essd-11-1411-2019>, 2019.