

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

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16 **Abstract.** Carbon monoxide (CO) concentrations in the troposphere are decreasing globally, with Africa as an
17 exception. Yet, the region is understudied, with a deficit of ground-based observations and highly uncertain CO
18 emission inventories. This paper reports multi-year observational CO data from the Mt. Kenya Global Atmosphere
19 Watch (GAW) station, as well as summertime CO isotope observations from both Mt. Kenya and Nairobi, Kenya.
20 The CO variability at Mt. Kenya is characterized by slightly increased concentrations during dry periods and a
21 strong influence of short-term pollution events. While some data gaps and differences in instrumentation
22 complicate decadal-scale trend analysis, a small long-term increase is resolved. While multi-year data gaps
23 complicate decadal-scale trend analysis, no overall long-term shift can be resolved. High pollution events are
24 consistent with isotopic signal from downwind savanna fires. The isotope fingerprint of CO in Nairobi indicate an
25 overwhelming dominance (near 100%) of primary emissions from fossil fuel combustion - with implications for
26 air pollution policy. In contrast, the isotope signature of CO intercepted at the large footprint Mt. Kenya region
27 suggests at least 70% primary sourced, with a predominance likely from, savanna fires in Africa. Taken together,
28 this study provides quantitative constraints of primary vs secondary CO in the eastern Africa region and in urban
29 Nairobi, with implications for satellite-based emission inventories as well as for chemical-transport and climate-
30 modelling.

31 1. Introduction

32 Carbon monoxide (CO) is the dominant sink for the hydroxyl radical (OH), accounting for over 50% consumption
33 of OH in the atmosphere (Lelieveld et al., 2016). It therefore influences the atmosphere's oxidation and cleansing
34 capacity and, by extension, chemically regulates the atmospheric lifetime and abundance of other reactive gases
35 such as methane and halocarbons (Lelieveld et al., 2016; Zheng et al., 2019). As such, CO is an indirect greenhouse
36 gas with a net positive warming effect on climate (Szopa et al., 2021). In addition to climate effects, CO is a
37 precursor to the formation of ground-level ozone, with implications for human health (Chen et al., 2021; WHO,
38 2016; Zhang et al., 2019). Anthropogenic activities such as biomass burning and fossil fuel combustion are
39 important contributors to the global CO budget, in addition to atmospheric reactions, e.g., oxidation of
40 hydrocarbons (Duncan et al., 2007; Zheng et al., 2019). However, the CO source contributions, mole fractions,
41 and atmospheric residence time are spatially variable, complicating the source-sink assessment.

42 Global CO levels have been declining over the past two decades, but Africa is an exception. The key source of
43 information on CO trends in the African region is satellite-based observations that show an increase in CO mole
44 fractions (Buchholz et al., 2021; Hedelius et al., 2021; Zheng et al., 2019). However, the ground-truthing of the
45 satellite observations is challenged by a deficit of atmospheric observatories and scant continuous long-term
46 observations in the region (DeWitt et al., 2019; Henne et al., 2008b; Kulmala, 2018). Exacerbating this
47 observational deficit, regional CO emission inventories are not well-defined as the continent possesses a unique
48 CO emission profile, different from other regions such as Europe and South Asia (Crippa et al., 2018; Dasari et
49 al., 2022; Hedelius et al., 2021). To advance our understanding of trends in CO over Africa and its source
50 contributions, long-term CO measurements and isotope-based source apportionment studies are required but data
51 availability is scarce.

52 The isotopic composition of CO provides insights into the relative strengths of regional CO sources and
53 atmospheric processing (Brenninkmeijer, 1993; Dasari et al., 2022; Henne et al., 2008b; Röckmann et al., 2002).
54 A particular source of CO possesses a characteristic isotopic signature, with the isotopic composition of the
55 ambient CO reflecting that of the combined sources, sinks, and atmospheric ageing (Brenninkmeijer and
56 Röckmann, 1997; Dasari et al., 2022; Popa et al., 2014; Röckmann et al., 1998, 2002). For example, CO from
57 primary sources (fossil combustion and biomass burning) has a more enriched $\delta^{18}\text{O}$ signature (above +12‰)
58 compared to that of secondary-formed CO, e.g., from oxidation of CH_4 (at ~0‰) and non-methane hydrocarbons
59 (NMHC), at $\sim 2.4 \pm 2.4\text{‰}$ (Brenninkmeijer and Röckmann, 1997).

60 Additional source information can be obtained from the $\delta^{13}\text{C}$ signatures. CO formed from methane oxidation is
61 strongly depleted in ^{13}C ($\delta^{13}\text{C} = -51.9 \pm 1.6\text{‰}$) in contrast to, for example, CO emitted from burning of C_4 plants
62 ($\delta^{13}\text{C} = -14.0 \pm 3.8\text{‰}$), C_3 plants ($\delta^{13}\text{C} = -26.9 \pm 4.9\text{‰}$) or fossil combustion at $-27.8 \pm 1.5\text{‰}$ (Brenninkmeijer et
63 al., 1999). However, the kinetic isotope effect (KIE) during the CO-OH reaction (the main atmospheric CO
64 removal mechanism) results in the enrichment of $\delta^{13}\text{C}$ by 4-5‰ and more depleted $\delta^{18}\text{O}$ signatures (by ~10‰) in
65 the lower troposphere (Brenninkmeijer et al., 1999; Röckmann et al., 1998). Overall, isotope forensics can provide
66 valuable data on CO emissions in remote and urban locations in Africa, especially considering that the region is
67 largely understudied, with very few ground-based CO observations and highly uncertain emission inventories.

68 This study investigates the long-term trends in CO mole fractions at Mt. Kenya GAW station, a high-altitude
69 monitoring site in equatorial East Africa well suited to intercept the regional emission footprint. Online CO mole
70 fractions measurements have been going on at the observatory since 2002, albeit with large data gaps due to
71 technical challenges. Flask-based measurements carried out at Mt. Kenya at different periods by NOAA (2003-
72 2011) were used for gap filling. The online and NOAA flask-measured CO data were obtained from the WMO's
73 World Data Centre for Greenhouse Gases (WDCGG) database. After the CO measurements from Cape Point,
74 South Africa, this is likely the longest-running data available in sub-Saharan Africa and provides observational
75 constraints of the region's long-term trend in CO. The present study additionally provides stable isotope
76 composition data of CO to resolve source attribution of the observed higher summertime CO amount fraction.
77 Furthermore, the data are compared to previously obtained and unpublished isotope data from Mt. Kenya (1996-
78 1997; earlier unpublished work by Röckmann and Brenninkmeijer) and that of an urban site in Nairobi in summer
79 2021 to provide further insights into regional CO sources. This dataset is unprecedented in the region and facilitates
80 improved understanding of the regional CO emission trends and source attribution.

81 **2. Methodology**

82 **2.1 Measurement sites**

83 Ambient air sampling was conducted at a remote mountain site, the Mt. Kenya Global Atmospheric Watch (GAW)
84 station, and in Nairobi city. The Mt. Kenya GAW station is located on the north-western slope of Mt. Kenya (0.062
85 °S, 37.297 °E, at 3678 m MSL) in eastern equatorial Africa. The station description, site selection and
86 representativeness, and meteorological characterization are detailed by Henne et al. (2008a, 2008b). In brief, the
87 station lies within a nature conservancy, the Mount Kenya National Park, and contributes to the World
88 Meteorological Organization GAW programme. The closest human settlements and roadways are over 17 km
89 away, and the nearest town (Nanyuki) is at 1900 m MSL. A small touristic infrastructure, the Old Moses Camp, is
90 situated 300 m below and ca. 1.9 km to the NNW of the station. The second site, in the megacity of Nairobi,
91 Kenya, was a rooftop measurement site (~17 m above ground level; 1690 m asl.; 1.279° S, 36.817° E). As described
92 previously, the Nairobi site is representative of the city's ambient conditions (Kirago et al., 2022b).

93 **2.2 Ambient air sampling**

94 Glass flask sampling was conducted in August 2021 at the Mt. Kenya GAW station and in Nairobi with an in-
95 house assembled portable sampler consisting of a diaphragm pump (KNF Neuberger N86E) connected with 1/4"
96 Dekabon tubing. The sampler design, glass flask pre-conditioning protocol and sampling procedure are previously
97 described (Dasari et al., 2022). Briefly, the sampler was designed to fill two pre-conditioned glass flasks (Normag,
98 1L) simultaneously, that is, each sample was collected in duplicates. Ambient air was drawn at a flow rate of 2 L
99 min⁻¹ and dried through a magnesium perchlorate trap. First, the glass flasks were flushed for 20 minutes before
100 compressing the dried air to an absolute pressure of ~1.7 bar. At Mt. Kenya GAW station, 21 nighttime (02:00h
101 local time) and six daytime (14:00h local time) air sample sets were collected. Nine sample pairs were collected
102 in Nairobi (every second day; daytime only; 14:00h local time). The filled glass flasks were sent to the Institute
103 for Marine and Atmospheric Research Utrecht (IMAU), Utrecht University, for processing and stable isotope
104 analysis of CO.

105 **2.3 Measurements of CO mole fractions and stable isotopes ($\delta^{18}\text{O}$ and $\delta^{13}\text{C}$) composition of CO**

106 The CO mole fraction and stable isotopic composition measurements of the collected glass flask samples were
107 performed at IMAU, Utrecht University. A continuous-flow isotope ratio mass spectrometry (CF-IRMS, Thermo
108 Scientific Delta V Advantage) system was used, applying a previously described measurement protocol (Pathirana
109 et al., 2015). In brief, the sample gas was introduced into the analytical system using an automated multi-port unit,
110 via a mass flow controller and under ultra-high-purity helium flow. Here, the air sample was directed through a
111 trap with Ascarite (8 - 20 mesh, Thermo Scientific™) followed by magnesium perchlorate (Sigma-Aldrich), to
112 remove CO_2 and water. A subsequent cryogenic trap (liquid N_2 , -196°C) was used to remove the remaining traces
113 of CO_2 , N_2O and hydrocarbons. The CO in the clean air matrix was then oxidized to CO_2 using Schütze reagent
114 ($\text{I}_2\text{O}_5/\text{H}_2\text{SO}_4$ mixture on granular silica gel) synthesized in-house. Subsequently, the CO-derived CO_2 was cryo-
115 trapped in liquid- N_2 , while other residual gases (e.g., O_2 and N_2) were pumped out. The sample was further purified
116 on a GC column, dried via a Nafion dryer, and subsequently transferred to the CF-IRMS via an open split inlet for
117 stable isotopes ($\delta^{18}\text{O}$ and $\delta^{13}\text{C}$) analysis. (Pathirana et al., 2015) The original CO amount fraction was deduced
118 from the quantity of the derived CO_2 .

119 The isotopic composition is expressed as per mil (‰) enrichment or depletion of the isotope ra-tio in the sample
120 relative to that of international standard materials, which in these cases are the Vienna PeeDeeBelemnite (V-PDB)
121 standard for $\delta^{13}\text{C}$ -CO, and the Vienna Standard Mean Ocean Water (V-SMOW) for $\delta^{18}\text{O}$ -CO measurements ~~The~~
122 ~~isotopic composition is expressed as per mil (‰) enrichment or depletion of the isotope ratio in the sample relative~~
123 ~~to that of international standard materials, which in these cases are the Vienna PeeDeeBelemnite (V-PDB) standard~~
124 ~~for $\delta^{13}\text{C}$ -CO, and the Vienna Standard Mean Ocean Water (V-SMOW) for $\delta^{18}\text{O}$ -CO measurements~~
125 (Brenninkmeijer et al., 1999; Pathirana et al., 2015). A reference cylinder with atmospheric air with known isotopic
126 composition and mole fraction ($\delta^{13}\text{C} = -30.25\text{‰}$; $\delta^{18}\text{O} = +7.10\text{‰}$; $\text{CO} = 180$ ppb) was used for calibration.
127 Periodical measurements of "target" gases were used to monitor the precision and accuracy of the measurements,
128 as well as the long-term stability of the analytical system (Pathirana et al., 2015). In addition, blank runs (without
129 injecting the sample or reference gas) were performed to assess the background CO_2 , mainly from the Schütze
130 reagent. The typical 1-sigma measurement reproducibility during the time of these analyses is estimated at 0.12‰
131 for $\delta^{13}\text{C}$ and 0.16‰ for $\delta^{18}\text{O}$.

132 **2.4 Sampling and isotopic characterization of CO for the 1996/97 campaign**

133 High-volume air samples were collected between July and September 1996 ~~The 1996/97 high volume air samples~~
134 ~~were collected~~ on an exploratory mission around Mount Kenya following the ring road A2/B6, and branching off
135 towards the mountain to locations where sufficient power was available for sample collection. During this
136 campai gn and later incidental samplings in 1997 ~~During this campaign and later incidental samplings,~~ air samples
137 of approximately 500 L volume were compressed into 5 L aluminium cylinders using a modified RIX compressor
138 (Mak and Brenninkmeijer, 1994). Unfortunately, records of precise locations have been lost. The filled cylinders
139 were sent to the Max Planck Institute for Chemistry in Mainz, Germany, for CO isotope analysis on a high-volume
140 extraction unit (Brenninkmeijer et al., 1999; Röckmann et al., 2002). A high CO concentration calibration gas (269

141 ppm) that was used during the 1996/7 measurements has been preserved and is regularly measured in the lab of
142 Utrecht University to assure scale compatibility.

143

144 2.5 Long-term CO mole fractions at the Mt. Kenya GAW station

145 High-resolution CO data from Mt. Kenya GAW station are available from the WMO World Data Centre for
146 Greenhouse Gases (WDCGG; <https://gaw.kishou.go.jp/>). A continuous time series is available for the 2002-2006
147 and 2020-2021 periods, with some large gaps attributable mainly to power outages and data quality issues.
148 Between 2010 and 2015, the station was disconnected from the power grid following a bush fire, while
149 performance audits revealed the CO analyzer to be in poor working condition between 2015-2019
150 (decommissioned in 2020), compromising the data quality (Zellweger et al., 2020). Over time, the CO
151 measurements were made using different CO analyzers (Thermo Electron Corporation TEI 48C-TL in 2002-2006,
152 Horiba APMA360 in 2010-2019, and Picarro G2401 in 2020-2021).

153 The instrument calibration, quality control protocols, and data treatment procedures are discussed elsewhere
154 (Henne et al., 2008b; Zellweger et al., 2009, 2020). In brief, ambient air was drawn into the CO instrument using
155 1/4" Teflon (till 2019) and 1/4" Synflex 1300 (after 2019) tubings at a flow rate of 4 l/min via a Nafion drier to
156 remove moisture and a particulate filter. The air inlet was about 7 m above ground and protected against rain, snow
157 and direct wind. These instruments were installed and calibrated by the Swiss Federal Institute for Materials
158 Science and Technology (Empa) in collaboration with the Federal Office of Meteorology and Climatology
159 MeteoSwiss, and operated by the Kenya Meteorological Department (KMD). The instrument calibration and
160 performance audits are conducted regularly by the GAW World Calibration Center hosted at Empa (Zellweger et
161 al., 2020). In addition, flask-based CO data from Mt. Kenya GAW station by NOAA Global Monitoring labora-
162 tory and published in the WDCGG database was used in this study (Petron, 2023).

163 2.6 Trajectory and statistical modelling

164 The air mass back trajectories (10 days; arrival height of 100 m above ground level) were calculated to identify
165 the air mass source region. The NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT,
166 version 4) and GDAS ($1^\circ \times 1^\circ$) archived meteorological datasets were used (Stein et al., 2015). The Bayesian
167 Markov chain Monte Carlo (MCMC) model was used to quantitatively constrain CO fractional contributions and
168 account for source end member variability and measurement uncertainties (Dasari et al., 2022). The MCMC
169 simulations were carried out with MATLAB R2020 with 1,000,000 runs and 10,000 runs for sample burn-in and
170 a data thinning of 100.

171 3. Results and Discussion

172 3.1 CO mole fractions at Mt. Kenya GAW Station and Nairobi

173 The results of the ~~continuous~~ CO observations at Mt. Kenya GAW station, both online and flask measurements,
174 are presented in Figure 1. Part of the data (2002 - 2006) has been comprehensively discussed previously (Henne

175 ~~et al., 2008b). Part of the data (2002–2006) has been comprehensively discussed previously (Henne et al., 2008b),~~
176 ~~and here it will be only compared to the 2021 period with respect to a general long-term trend.~~ Overall, peak CO
177 mole fractions were observed during the dry periods (SI Figure S1). Assessment of the long-term CO trend,
178 following the approach by Thoning and Tans (1989), reveal a small but statistically significant positive decadal
179 trend of 6.7 ± 0.4 ppb/10yrs. This statistical model is based on a fit function that includes a linear term, a quadratic
180 term, as well as first and second harmonics. For comparison, simple linear regression gives a similar decadal rate
181 of 6.2 ± 0.6 ppb/10yrs (for uncertainty estimation, see Kirago et al., 2022a). Like many types of environmental
182 data, the present CO data display a lognormal-like concentration distribution, suggesting influence by exponential
183 processes such as sink kinetics (Andersson, 2021). This may influence trend analysis. Similarly to linear
184 regression, regression of log-transformed data also gives a significant positive rate, which suggests that the skewed
185 concentration profile has little influence on trend estimation. However, given the large data gaps and different
186 measurement techniques, such interpretations should not be over-emphasized. Nevertheless, the increasing trend
187 here constrained for ground observations of CO is qualitatively consistent with satellite retrievals and model
188 estimates for sub-Saharan Africa (Buchholz et al., 2021; Hedelius et al., 2021; Zheng et al., 2019). However, the
189 seasonal variations are not pronounced; the intra-seasonal peak to peak amplitude is larger than the variations
190 between different seasons, implying a strong influence of short-term pollution events. Meanwhile, no clear multi-
191 year trend in CO concentrations were detected (the rate of the long-term trend is ~ 0 ppb/year), albeit the large
192 data gaps and different measurement techniques precludes a detailed analysis.

193 The observed CO levels, ranging between 55 – 250 ppb, are comparable to those previously recorded at Mt. Kenya
194 station (Henne et al., 2008b), but lower than CO concentrations reported at the Rwanda Climate Observatory -
195 another remote site also in Eastern Africa, possibly with a stronger and more direct influence of savanna burning
196 episodes (DeWitt et al., 2019). The daytime and nighttime ambient flask CO concentrations were comparable,
197 similar to observation from Picarro-measured CO measurements though with a slight daytime elevation (SI Figure
198 S1). During the nighttime, the station stands above the atmospheric boundary layer, hence reduced influence from
199 local sources. Overall, changes in source strength, air mass transport pathways and meteorological parameters such
200 as planetary boundary layer thickness are likely to be key drivers of the observed temporal variations. In Nairobi,
201 the CO concentrations during August 2021 range between 200 – 700 ppb ($\sim 0.2 - 0.8$ mg m⁻³, assuming average
202 weather conditions), well within the WHO recommended short-term (24-h average) air quality guideline level of
203 4 mg m⁻³ (WHO, 2021). While CO is not a major direct health concern in Nairobi nor in other urban settings
204 (Chen et al., 2021), it affects the presence of health-detrimental components such as ground-level ozone and
205 secondary aerosols.

206 Back-trajectories calculated with the HYSPLIT model were combined with the CO data to learn more about source
207 regions incident with elevated CO mole fractions. Air masses originating from different geographical areas, such
208 as from eastern Africa, Arabian Peninsula, northern Africa, South Asia, and south-eastern Africa, as well as cleaner
209 air masses from the Indian Ocean, are intercepted at Mt. Kenya GAW station (Figure 2). This underlines the
210 suitability of Mt. Kenya GAW station to capture both the regional and intercontinental footprints. The elevated
211 summertime (June - August) CO mole fractions are linked to the arrival of south-easterly air masses, coinciding
212 with large-scale savanna fires in southern Africa and Madagascar. The air masses shift north-easterly during winter

213 (December - March), and coincide with savanna fires in northern Africa (Andersson et al., 2020; Kirago et al.,
214 2022a). Although the intercepted air masses do not directly flow over West-Central Sub-Saharan Africa, where
215 most fires occur, the atmospheric residence time of CO is sufficient for regional and intercontinental mixing. Air
216 masses with elevated CO loadings from South Asia and the Arabian Peninsula are also intercepted during winter.
217 High wintertime CO amount fractions have been reported from a South Asian receptor site in the northern Indian
218 Ocean (Dasari et al., 2022). Taken together, the seasonal variability in CO mole fraction can partly be explained
219 by regional emission events, combined with a contribution from other geographical source regions such as South
220 Asia.

221 3.2 Isotopic constraints of sources to CO from Mount Kenya and Nairobi

222 ~~The stable isotope composition of CO ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) for ambient samples from Mt. Kenya GAW station during~~
223 ~~August 2021 varied temporally with the CO mole fractions (SI Figure S2). The stable isotope composition of CO~~
224 ~~($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) for ambient samples from Mt. Kenya GAW station during August 2021 varied temporally and~~
225 ~~inversely with the CO mole fractions.~~ The $\delta^{13}\text{C}$ ranged between -31.5% to -28.0% , while $\delta^{18}\text{O}$ ranged between
226 2.5 to 10.0% (SI Figure S2). However, there were no distinct temporal or diurnal trends in the recorded isotopic
227 values (both daytime and night-time samples were measured). The air masses were consistently southeasterly
228 during the three weeks study period (SI Figure S3). Comparable $\delta^{18}\text{O}$ composition was observed in 1996/97
229 samples (ranged between 3.7 to 10.4%), but was more enriched in $\delta^{13}\text{C}$ (-28.4% to -26.6%). The isotopic
230 composition in the Mt. Kenya background region was distinct from that of the urban Nairobi location that recorded
231 highly enriched $\delta^{18}\text{O}$ values ($17.5 \pm 2.2\%$; SI Figure S2).

232 The Keeling plot approach provides insights into the regional CO sources. Here, a linear relationship is observed
233 between the isotope signatures and the inverse of the CO amount fractions ($\delta^{13}\text{C}$ vs $1/[\text{CO}]$ and $\delta^{18}\text{O}$ vs $1/[\text{CO}]$)
234 both at Mt. Kenya and in Nairobi (Figure 3). ~~This implies that the CO dynamics in this system can be described~~
235 ~~by a two-component mixture: a relatively stable background fraction and a regional varying source (Dasari et al.,~~
236 ~~2021; Keeling, 1958). This implies that the CO dynamics in this system can be described by a two component~~
237 ~~mixture; a relatively stable background fraction and a regional source.~~ The y-axis intercept in this relation
238 represents the source signature. At Mt. Kenya, analysis of the recently-obtained dataset (2021) reveals the stable
239 isotopes signature of the source of $\delta^{18}\text{O} = 14.0 \pm 1.2\%$ and $\delta^{13}\text{C} = -27.7 \pm 0.6\%$. For the samples collected during
240 the 1996/97 campaign, the $\delta^{18}\text{O}$ signature is very similar and indistinguishable ($\delta^{18}\text{O} = 14.2 \pm 2.1\%$), while the
241 source is more enriched in ^{13}C ($\delta^{13}\text{C} = -24.7 \pm 0.7\%$; Figure 4). The latter suggests differences in the relative
242 strengths of the contributing sources, possibly a relatively higher contribution from C_4 plants burning or a relatively
243 smaller influence of secondary CO from atmospheric reactions during the 1996/97 campaign. It should be kept in
244 mind that the 1990s samples were obtained at a lower altitude location on the slopes of Mt. Kenya. C_4 plants like
245 maize and sugarcane are commonly grown in Kenya, while also biomass usage (including crop residuals for
246 household energy) and agricultural burning are prevalent in the region (World Bank, 2011). In Nairobi, a clearly
247 distinct source signature is noted, especially for $\delta^{18}\text{O}$ ($\delta^{13}\text{C} = -26.0 \pm 0.4\%$ and $\delta^{18}\text{O} = 22.9 \pm 0.8\%$; Figures 4).
248 The highly enriched $\delta^{18}\text{O}$ source signature in Nairobi indicates almost exclusively high temperature combustion
249 sources, while a mixed source regime (both combustion sources and CO emanating from atmospheric reactions)
250 is observed at Mt. Kenya; these can be quantitatively resolved using an isotopic mass balance approach

251 The source signatures can be used to quantitatively constrain the fractional contributions of CO in the regional
252 background and urban atmosphere (Dasari et al., 2022). However, information was available for only two isotopes,
253 $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$, against five potential sources that can contribute to the overall CO isotopic signature (C_3 plants,
254 fossil, C_4 plants, CH_4 oxidation and NMHC oxidation), yielding a mathematically under-determined scenario.
255 Furthermore, the weak linear correlation for $\delta^{13}\text{C}$ in the Keeling plot ($\delta^{13}\text{C}$ vs $1/[\text{CO}]$; $R^2 = 0.34$ for Mt. Kenya)
256 limits its application in the statistical model. Therefore, only $\delta^{18}\text{O}$ signatures were here modelled ($R^2 = 0.64$ for
257 both Mt. Kenya; $R^2 = 0.89$ for Nairobi; $P < 0.05$). Hence, the CO sources were partitioned into two major classes:
258 primary/combustion (fossil, C_3 and C_4 biomass) and secondary (i.e., oxidation of methane and NMHC).

259 A Bayesian statistical model, drawing upon the model described in Dasari et al. (2022), was used to estimate the
260 contribution of secondary ($f_{\text{secondary}}$) vs primary (f_{primary}) CO sources. In this model the relative contributions of
261 primary vs. secondary CO for the temporally varying source is computed, corresponding to the $\delta^{18}\text{O}$ values at the
262 limit where $1/\text{CO}$ approaches zero (the $\delta^{18}\text{O}$ intercept in the Keeling plot). First, the source end members for the
263 two fractions were established. Unlike the oxidation of NMHC, the CH_4 -oxidized CO fluxes have little variability
264 (CH_4 has a long atmospheric lifetime) and largely contribute to the background signal (Dasari et al., 2022; Worden
265 et al., 2019; Zheng et al., 2019). Therefore, the temporally-varying secondary CO end member was assigned that
266 of the NMHC oxidation source ($\delta^{18}\text{O}_{\text{secondary}} = 2.4 \pm 2.4\text{‰}$).

267 The primary CO end member is a composite of the three combustion sources; C_4 biomass ($+20.2 \pm 4.9\text{‰}$), C_3
268 biomass ($+16.3 \pm 5.1\text{‰}$) and fossil fuel combustion at $+19.2 \pm 4.9\text{‰}$ (Dasari et al., 2022). Although the relative
269 contributions are uncertain, the $\delta^{18}\text{O}$ end members largely overlap. Model estimates show biomass burning in
270 Africa accounts for 80 - 90% of the surface CO emissions (Zheng et al., 2018). Similar contributions to black
271 carbon (different but co-emitted incomplete combustion product) were observed using isotopic constraints with
272 near-equal contributions from C_3 and C_4 biomass in the eastern Africa background atmosphere (Kirago et al.,
273 2022c). Therefore, the relative source contributions were estimated at 50% from C_3 biomass and 50% from C_4 and
274 fossil sources at Mt. Kenya. Hence, a primary CO end member was established at $\delta^{18}\text{O}_{\text{primary}} = 18.4 \pm 3.5\text{‰}$. In
275 Nairobi, fossil fuel combustion was estimated to contribute to 85% of the CO emission in the city ($\delta^{18}\text{O}_{\text{primary}} =$
276 $+19.2 \pm 4.9\text{‰}$). ~~Since primary source endmembers largely overlap, the model simulations were generally insen-~~
277 ~~sitive to chosen priors, as investigated by sensitivity analysis. Since individual source end members in the primary~~
278 ~~fraction largely overlap, the model simulations were generally insensitive to chosen priors, as investigated by~~
279 ~~sensitivity analysis.~~

280 A similar $\delta^{18}\text{O}$ source signature ($\sim 14.0 \pm 2.1\text{‰}$) was observed at the two Mt. Kenya campaigns (1996/97 and
281 2021). Applying the established endmembers, we estimate the contribution of CO from primary/ combustion
282 sources at the regional background site to be at least 70%. In contrast, we found an almost exclusively primary CO
283 component for the urban Nairobi case. Nairobi is a strong air pollution source region, and the CO loadings largely
284 reflect the city's CO emissions. CO is, e.g., a precursor to low-level ozone, and thus emissions deteriorate air
285 quality. Present findings show that air quality policy should target primary emissions, especially from traffic
286 (Kirago et al., 2022b). In contrast, Mt. Kenya GAW station captures a more regional footprint with a dominant
287 contribution from savanna fires.

288 4. Conclusion

289 This study provides ground-observational constraints that broadly supports earlier suggestions that savanna fires
290 are the main emitters and modulators of CO loadings over Sub-Saharan Africa. Although the data gaps in CO
291 mixing ratios and mixed instrumentation complicates detailed analysis, a small decadal increase of 6.7 ± 0.4
292 ppb/10yrs was resolved for the Mt. Kenya GAW station, in agreement with satellite observations and emission
293 inventories for the Sub-Saharan region (Buchholz et al., 2021; Hedelius et al., 2021; Zheng et al., 2019). ~~Albeit~~
294 ~~data gaps in CO mixing ratios prevent detailed analysis, no clear long-term trend was resolved for the Mt. Kenya~~
295 ~~GAW station.~~ Isotope-based source apportionment shows that at least two thirds of the CO emitted from East
296 African savanna fires are of primary origins, while for Nairobi primary sources approach 100%. The latter has
297 implications for air quality policy, suggesting primary emissions such as traffic should be targeted, in line with
298 previous findings for BC (Kirago et al., 2022b). These findings put constraints on satellite-based emission
299 inventories and chemical-transport and climate modelling. Overall, this study corroborates earlier findings that in
300 order to reduce the secondary climate warming effect from CO over Sub-Saharan Africa, man-made savanna fires
301 should be reduced (Andersson et al., 2020).

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319 Author Contribution

320 Conceptualization of the study by AA. Design and execution of field campaigns by LK, AA, SG and MJG.
321 Management of Mt. Kenya GAW station and instrument calibration by DN, JK, CZ, CF & MS. Isotope analysis
322 by MEP. 1996/97 sampling campaign and sample analysis by TR. Data analysis by LK with support from AA,
323 ÖG, SLH and SG; Manuscript writing by LK with support from co-authors.

324 **Competing interests**

325 The authors declare no competing interests.

326 **Additional information**

327 Supplementary Information (SI) contain the flask-based CO measurement data for the 1996/97 and 2021 sampling
328 campaigns and the respective stable isotopes of CO (Table S1- S3).

329 Data from this study will be available in the Bolin Centre Database (bolin.su.se/data/).

330

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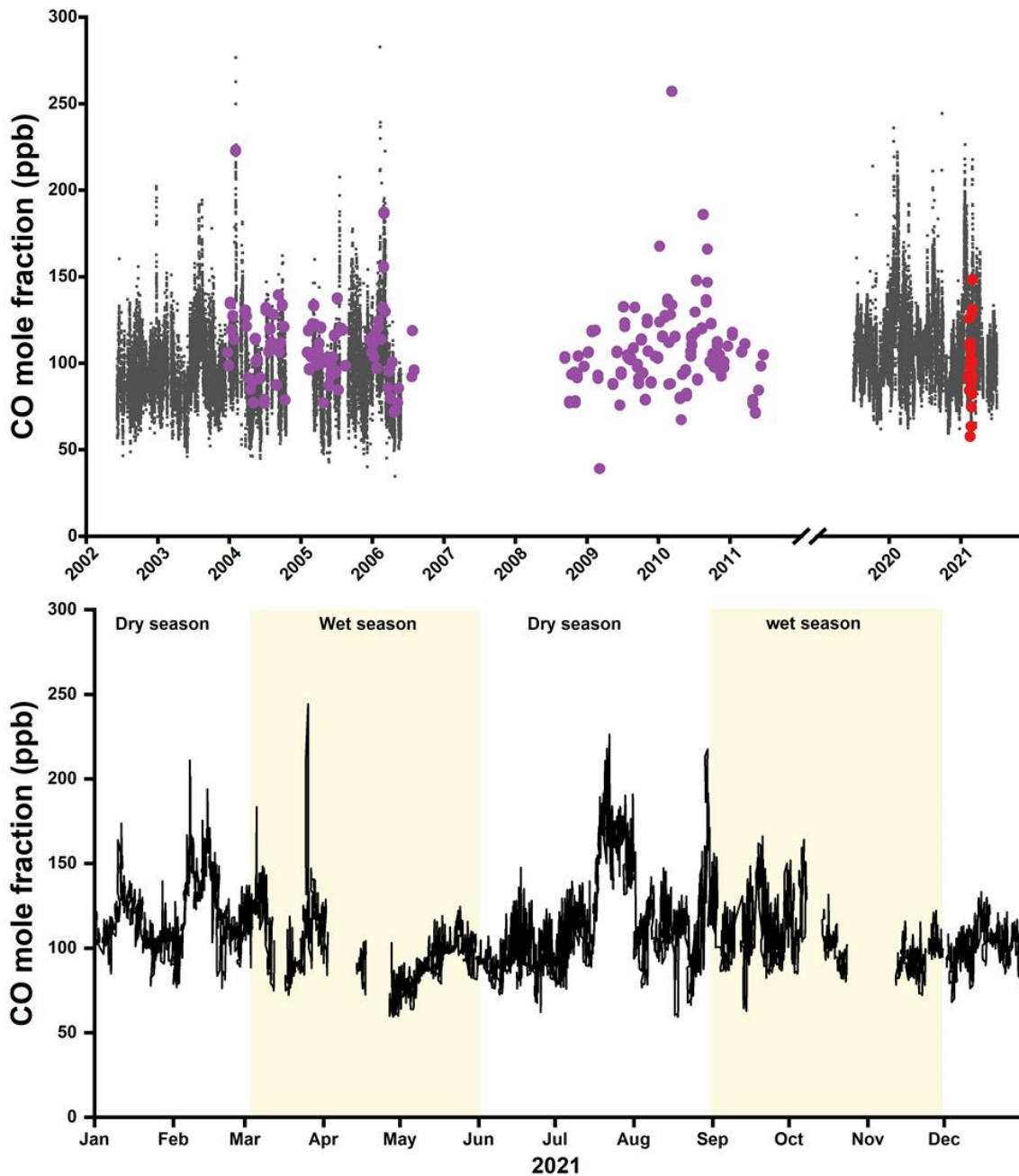
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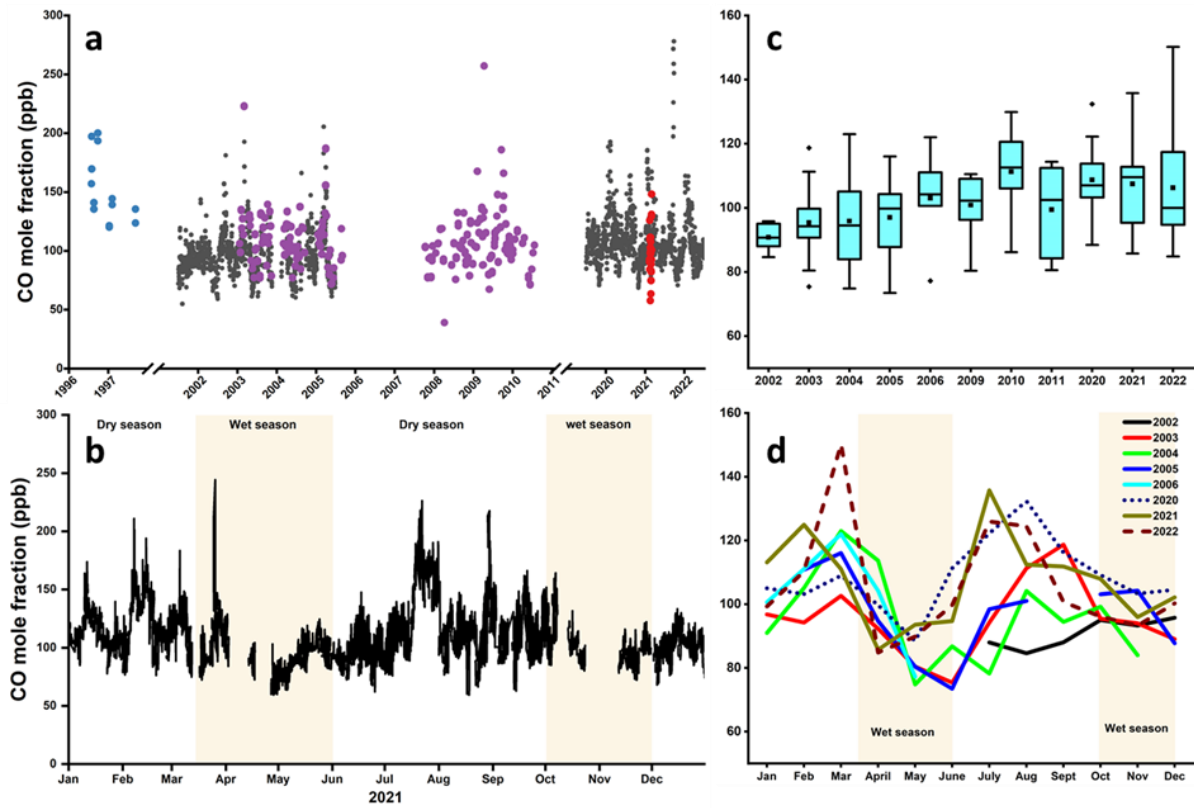
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441

442 Figure 1: Time-series of CO mole fractions at Mt. Kenya-GAW station. a) Daily-resolution
 443 continuously measured CO mole fraction (Picarro) at Mt. Kenya-GAW is represented by black dots.
 444 The CO data was retrieved from the WMO's WDCGG database covering 2002 to 2021. Different
 445 instrumentations were used over time, but similar instrumental calibration, quality control, and
 446 assurance protocols were applied. Flask-based measurements by NOAA at the station are presented
 447 in purple symbols, while flask samples during our 2021 campaign are shown in red symbols. b)
 448 Variations in CO mole fractions for the year 2021. The prevailing typical weather conditions are

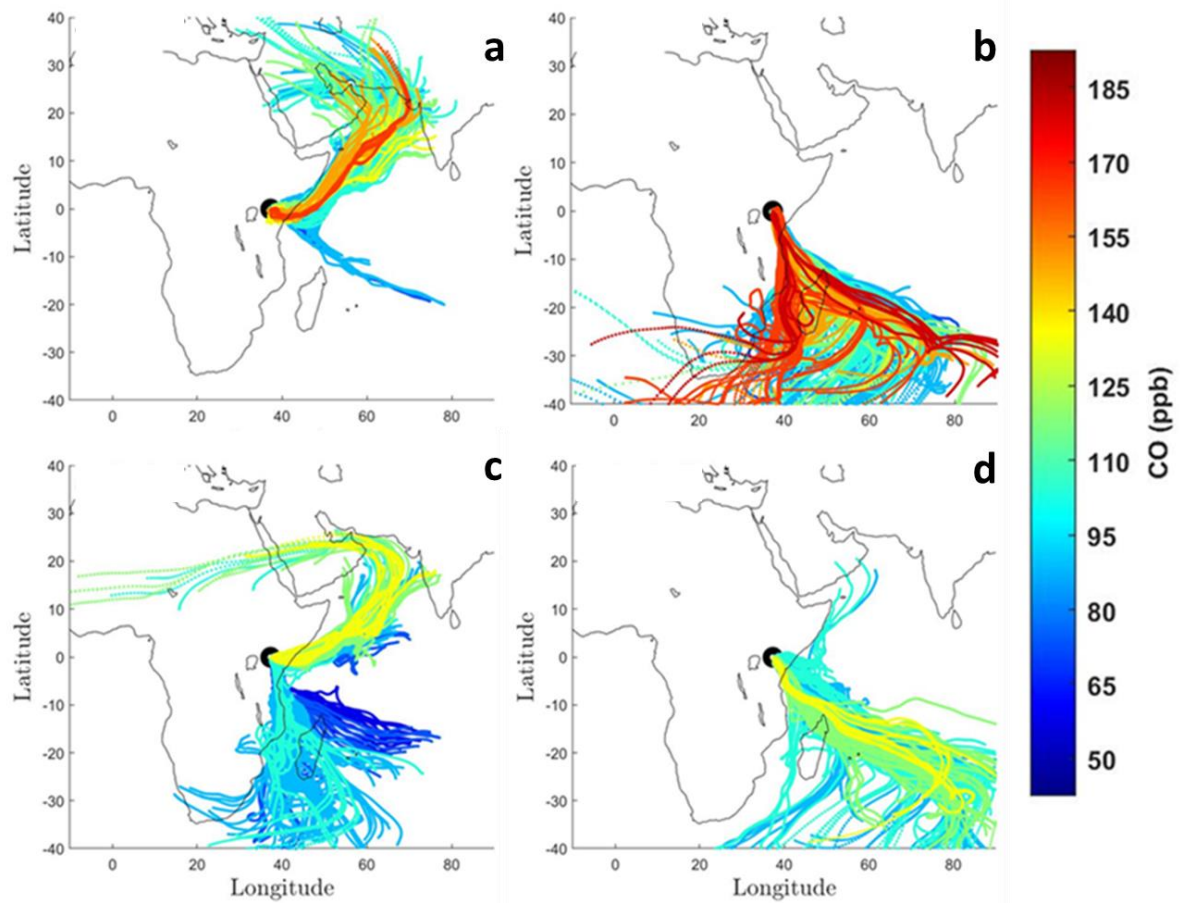
449 indicated.



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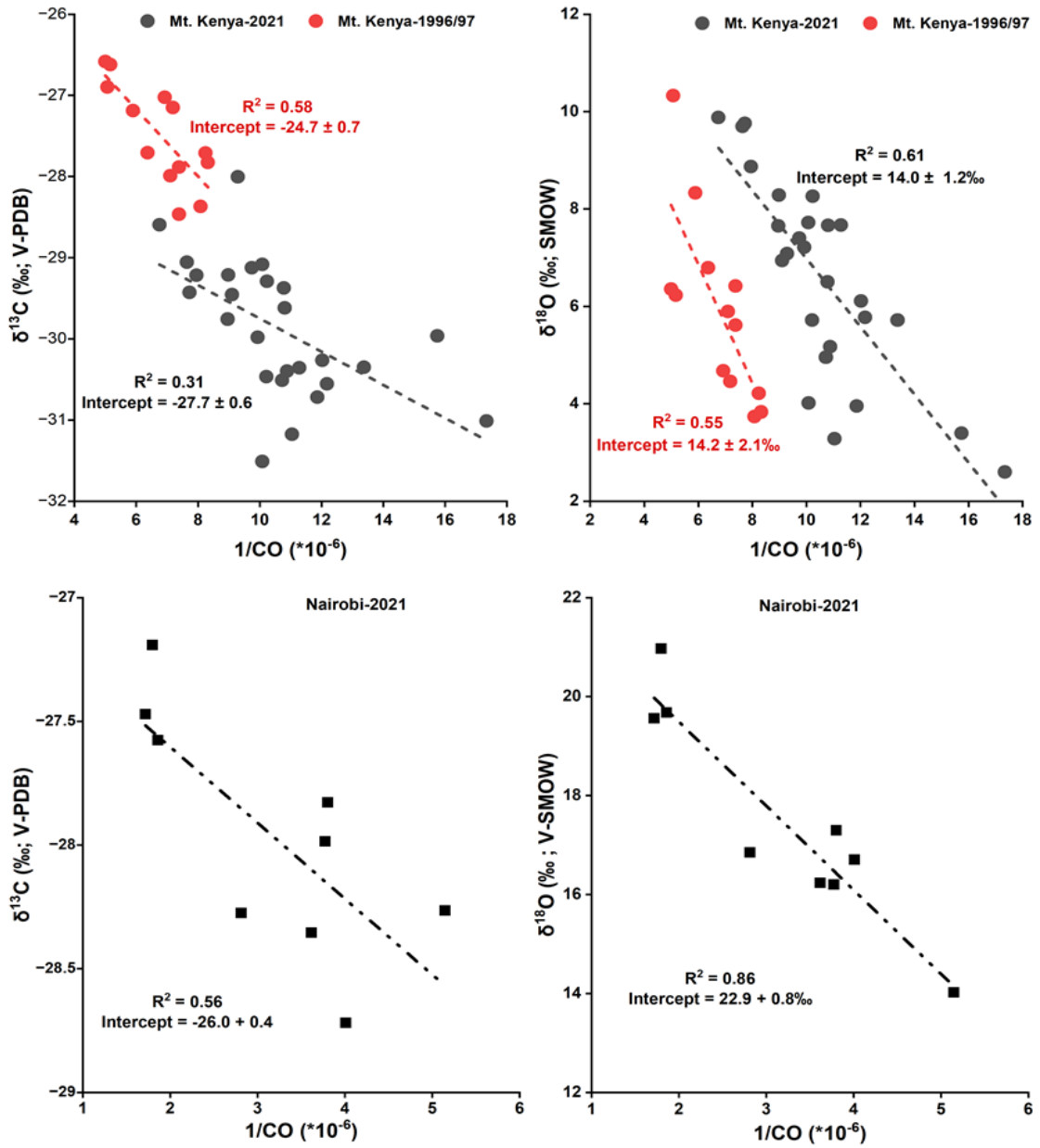
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452 **mole fraction (Picarro) at Mt. Kenya GAW is represented by black dots. The CO data was retrieved from the WMO's-**
453 **WDCGG database covering 2002- 2021. Different instrumentations were used over time, but similar instrumental**
454 **calibration, quality control, and assurance protocols were applied. Flask-based measurements by NOAA at the station**
455 **are presented in purple symbols, while flask samples during 1996/97 campaigns are shown as blue dots and flask**
456 **samples from the 2021 campaign are shown in red symbols, in good agreement with the online measurements, as shown**
457 **in SI Figure S4. b) Variations of CO mole fractions for the year 2021. The prevailing typical weather conditions are**
458 **indicated. c) Annual averaged CO mixing ratios. The boxes represent the 25th and 75th quantiles, and the black line**
459 **represents the median value. The bottom/top whiskers are the minimum and maximum values, respectively, while**
460 **diamonds represent the outliers. d) Inter-annual cycles of monthly averaged CO mole fractions (coloured lines represent**
461 **individual years).**

462



463

464 **Figure 2: Seasonal changes in CO concentration-coded back trajectories intercepted at Mt. Kenya; (a) December –**
 465 **February 2021, (b) June – August 2021, (c) March – May 2021, and (d) Sep-tember – November 2021.** (a) December –
 466 **February, (b) June – August, (c) March – May, and (d) September – November. Ten days air masses back trajectories**
 467 **are calculated at an arrival height of 100 m above ground level.**



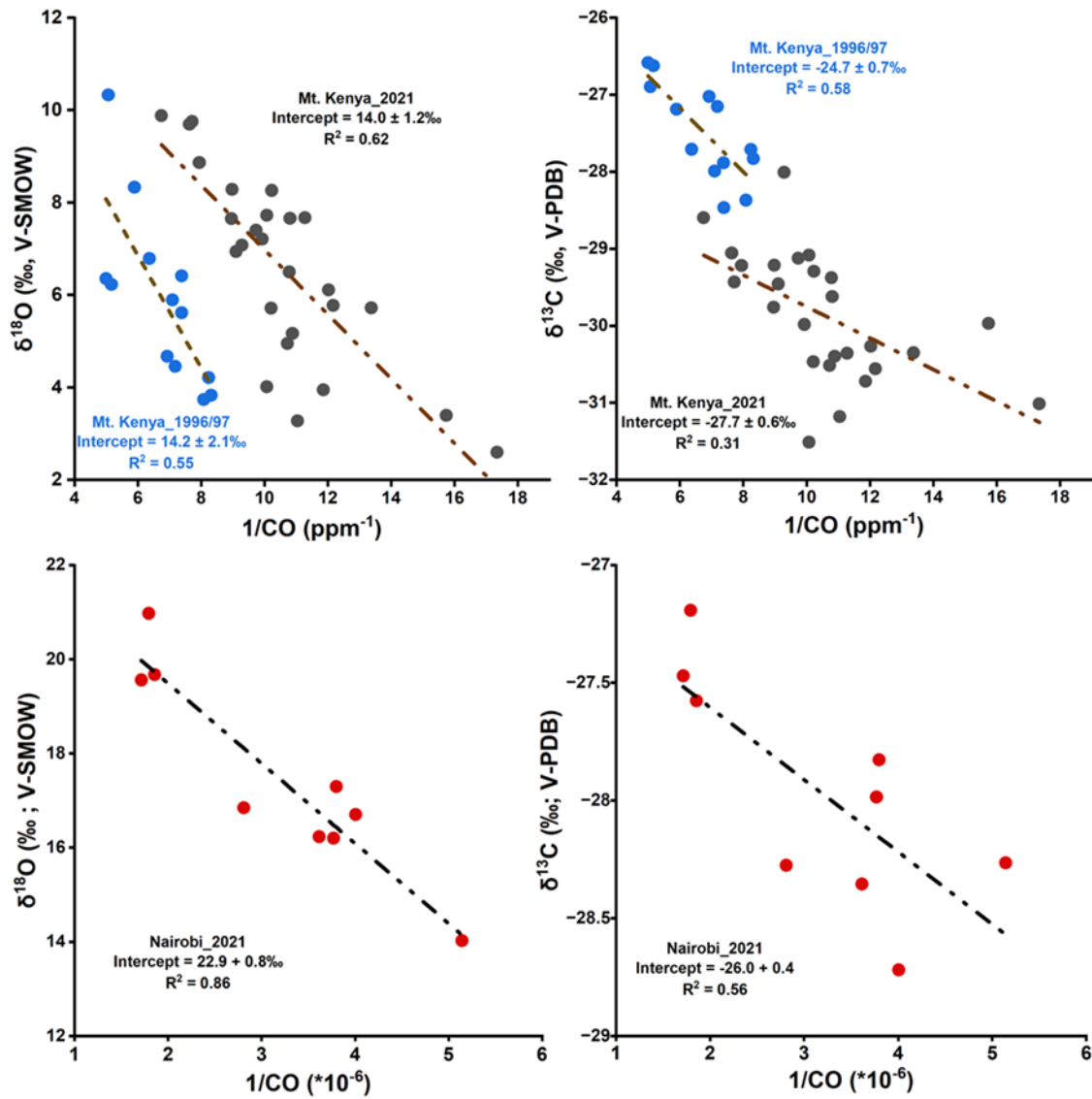
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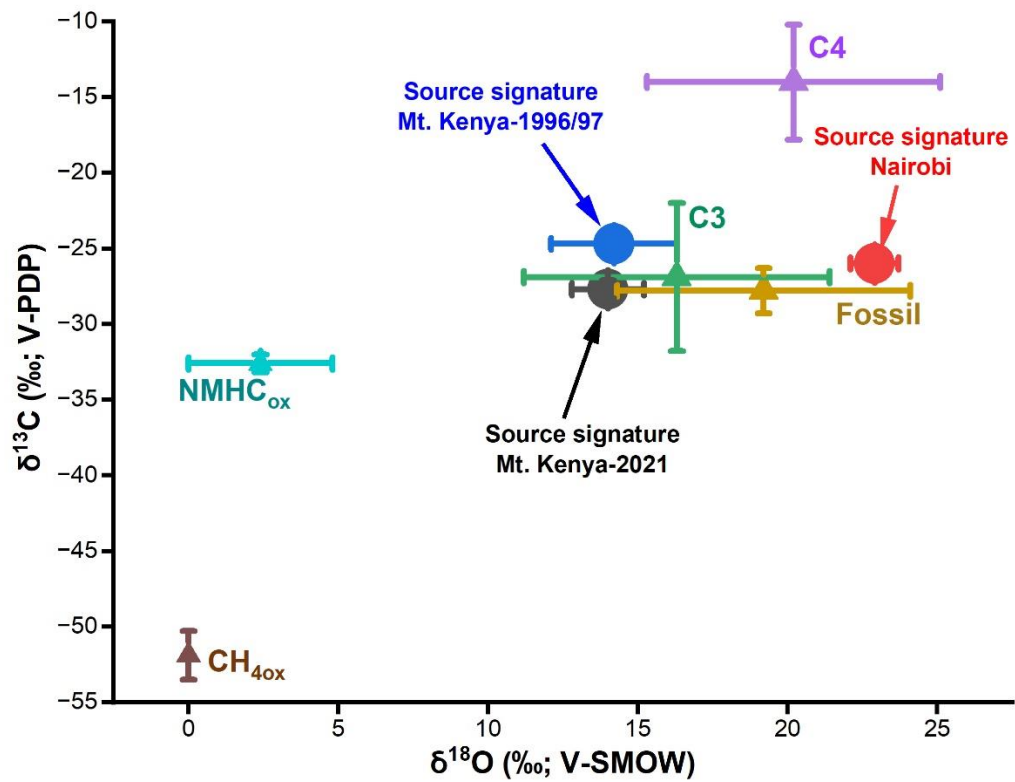
471

Figure 3: The Keeling relation plots (i.e., isotopic signature vs the inverse of the measured CO mole fraction) for Mt. Kenya (top panel) and Nairobi (bottom panel). The y-intercept in the Keeling relationship represents the source signature.



472

473 Figure 3: The Keeling relation plots (i.e., signatures of the two isotopic systems vs the inverse of the measured CO
 474 mole fraction) for Mt. Kenya (top panel) and Nairobi (bottom panel). The y-intercept in the Keeling relationship
 475 represents the source signature.



476

477 Figure 4: Stable isotope ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) source signatures of CO for Nairobi and Mt. Kenya (2021 and 1996/97), and
 478 the source end members. The source end members are adopted from Brenninkmeijer et al. (1999).