Manuscript egusphere-2025-5272, "Long-term trace gas and black carbon measurements at the high-altitude station Mount Kenya: tropical atmospheric variability and the influence of African emissions", by Leonie Bernet et al.

Replies to review RC1 from Anonymous Referee #1, on 12 Jan 2026

We thank Reviewer 1 for the careful assessment and constructive suggestions. Below, we provide the answers to his/her comments (in blue). Some of the new text added to the revised manuscript is given in italics.

Reviewer 1, general comments:

This paper follows a pair of papers that reported the installation (Henne et al., 2008a, JAMC) and first 5 years of CO/CO2/CH4/O3 data at Mt Kenya (2002-2006, Henne et al., 2008b, ACP) by adding roughly 5 more years of data (2020-2024). Seasonality of the data and precipitation & temperature cycles are displayed. Flexpart analysis of air parcel origins at the free tropospheric (FT) Mt Kenya site are described using the same regional partitioning as in Henne2008b. Comparisons of data with CAMS are included for the more recent data attempting to learn more about source apportionment; interpretation is somewhat limited because CAMS and data agreement are not very good. Black carbon (BC) data for the more recent period are displayed.

As a Data Report, the new data are useful and should be published.

Thank you for your general comments.

As a scientific analysis, the paper is uneven in quality and not as well organized as the 2008b paper. Mis-statements about uniqueness of the network are made (easy to fix).

Adequate references were added to acknowledge other global networks as suggested below.

However, more importantly, the paper is not as well organized as an ACP Research Article should be. The paper would have greater value to the scientific community if the authors focused less on the CAMS comparisons and more on trajectories for source apportionment, updating some of the most useful figures in Henne2008b. Even more, the obvious analysis to add is a comparison of the newer and first 5-year period concentrations. In view of two new papers that computed FT ozone trends (~2000-2023) from the Nairobi sonde FT record (Thompson et al., 2025; Van Malderen et al., 2025), comparing the earlier and more recent Mt Kenya records is an important result. Although Henne2008b and your Fig 3c show some offsets between the sondes at Mt Kenya level and your GAW data, the correlations are very good. It is recommended that the paper analyses be augmented with a trends calculation and ordered as follows: Climatology, seasonality, diurnal patterns, trajectory/sector analysis (like Fig 8 in Henne2008b), trends for the 4 species in Fig C1, finally source apportionment. Additional recommendations and references follow.

We thank the reviewer for this constructive suggestion. In response, we substantially extended the manuscript with a dedicated trend analysis for all four trace gases, including comparisons with published regional and global trends. While we did not fully reorganize the manuscript as proposed, the current structure already closely follows the recommended progression. Specifically, the revised “Time series and trend analysis” section now addresses the long-term behaviour and climatological context, followed by sections on seasonality, diurnal patterns, and finally trajectory- and sector-based source attribution. This structure allows trends to be interpreted consistently in the context of observed variability and source influences, without fragmenting the analysis. We therefore retained the overall structure while ensuring that all elements suggested by the reviewer are now explicitly covered. The discussion of trends in the revised “Time series and trend analysis” section now reads:

"Trends for different periods as distinguished by different colours in Figure C1 and are listed in Table C1. Monthly aggregates of both the continuous measurements and the flask analyses were used for the determination of trends by linear regression. The CO_2 trend intensified over the 23-year record. While flask measurements for 2002–2011 indicated an increase of less than 2 ppm yr^{-1}, recent observations show a rise of nearly 2.4 ppm yr^{-1}. This rate is consistent with global increases reported by the NOAA network (NOAA, Global Monitoring Laboratory, 2025) but is slightly below the most recent decadal global growth rate documented by WMO (World Meteorological Organization (WMO), 2025). CH_4 trends during 2002–2011 were characterized by the temporary global near-steady state in atmospheric CH_4, before growth resumed in 2007 (Rigby et al., 2008). As a result, the early-period trend is comparatively weak, while recent observations show a significant increase of more than 12 ppb yr^{-1}, even exceeding the global mean growth rate of 10.6 ppb yr^{-1} over the past decade (World Meteorological Organization (WMO), 2025). This acceleration aligns with recent assessments that the tropics dominate the latitudinal contribution of global emissions (Saunois et al., 2025) and highlight Central Africa and tropical South America as the dominant contributors to the current net methane flux to the atmosphere (Niwa et al., 2025).

The combined O_3 time series for 2002–2024 is the most internally consistent among the four trace gases discussed here, as it is based on continuous in-situ measurements using the same technique throughout. A statistically significant positive trend is found for the full period (2002–2024) of 0.20±0.07 ppb yr^{-1}. Comparison of sub-periods indicates a slowdown in the ozone growth rate, with a stronger increase during the early years (2002–2006: 0.42±0.88 ppb yr^{-1}) and a smaller trend in the more recent period (2015–2024: 0.14±0.21 ppb yr^{-1}). This behavior is consistent with SHADOZ observations showing that free-tropospheric and total tropospheric ozone above Nairobi exhibit weak, statistically insignificant long-term trends over 2000-2022 of 0.05 to 0.08 ppb yr^{-1} depending on the trend calculation approach (Van Malderen et al., 2025), with earlier positive tendencies leveling off in recent years (Thompson et al., 2025). Our results also complement the conclusions of Cooper et al. (2020), who analyzed long-term ozone trends from 27 remote ground-based sites worldwide but could not include East African stations owing to the lack of sustained observations at the time. While highlighting Africa as a major observational gap, they concluded that, globally, free-tropospheric ozone has increased since the mid-1990s, in contrast to Europe and North America, where surface ozone trends have largely flattened or declined since around 2000.

The long-term CO trend is the most difficult to interpret because it combines several measurement techniques, including in-situ NDIR and CRDS observations, different calibration strategies, and a mixture of in-situ, flask, and later again in-situ data. In addition, CO observations in the tropics are strongly influenced by biomass-burning emissions and by the varying strength of El Niño and La Niña events. These factors produce temporary

CO enhancements, resulting in large interannual variability that often dominates any long-term trend signal. For the period 2002–2011, the CO trend derived from flask measurements

is negative (-0.48±1.28 ppb yr−1), consistent with overall patterns observed at remote background sites (Patel et al., 2024). However, Patel et al. (2024) also highlight that trends in the tropics are less consistent than at higher latitudes because of the strong influence of biomass burning and climate variability. In contrast, the most recent period (2020–2024) shows a slightly positive trend, similar to that obtained for the full dataset. Depending on whether the early continuous measurements or the

flask data are used, the overall CO trend ranges between 0.11±0.33 and 0.63±0.29 ppb yr−1. These differences are likely related to the varying sensitivity of continuous versus intermittent flask sampling to episodic fire emissions. The long data gap from 2012 to 2019 also compromises robust trend determination."

Reviewer 1, specific comments:

Abstract Comment. Also applies to the Introduction. (1) It is important to acknowledge the other two African WMO/GAW stations on the continent and then point out as you do that the Mt Kenya is the only equatorial station in Africa…sample wording follows.

Abstract. Long-term observations of atmospheric composition are essential for understanding regional and global climate impacts. Although the Global Atmosphere Watch (GAW) programme provides a network of worldwide measurements, continuous atmospheric measurements across Africa are limited to three stations. This study presents in-situ measurements of trace gases and black carbon from the Mount Kenya GAW station (MKN) from 2020 to 2024, offering a unique dataset from equatorial Africa. Its location exposes MKN to air masses from both hemispheres, enabling detection of emissions and providing insights into tropical variability such as seasonal and diurnal cycles. We present carbon dioxide (CO2), methane (CH4), carbon monoxide (CO), ozone (O3), and black carbon (BC) measurements and describe seasonal and diurnal characteristics. Trajectory calculations with emissions estimates for CO and methane give African vs non-African contributions to greenhouse gases and air pollution. CO and BC were mainly linked to household fuel use and industrial energy, with biomass burning contributing during dry seasons. Methane variability was driven by agriculture and seasonal wetlands, but large uncertainties remain in all emission estimates. We also compare the 2020-2024 measurements to Mt Kenya trace-gas data from 2002-2012. (give % change). Comparison of the observations with Copernicus Atmospheric Monitoring Service (CAMS) model products shows CAMS fails to capture O3 and BC dynamics during rainy seasons. Our findings confirm the value of MKN observations for evaluating atmospheric models and emission inventories, highlighting the need to expand African measurement infrastructure. 235 words

(2) The wording above reflects a strong Recommendation to compute trends or "% changes" from means over the first 5-10 years and the more recent 5-yr period. Because your attributions (line 13-14) have such large uncertainties, better to put concrete results on the changes in the abstract and to call attention to the changes in the Introduction and Conclusions sections! That will be of more value to readers and likely enhance citations to the paper.

We largely followed the reviewer's suggestions by adding a qualitative discussion of trends for the different periods and by summarizing the corresponding quantitative values in tables provided in the Appendix. We deliberately did not list specific numerical trend values in the Abstract, as including multiple period-dependent numbers would risk overloading and potentially confusing the reader. Instead, all trends for the different time periods are now discussed in detail in Sect. 3.2 and given in tables in the appendix.

To reflect the reviewer's request at a higher level, the Abstract has been revised to include a qualitative summary of the trend behaviour. The relevant sentence now reads:

"We further compare the measurements from 2020-2024 with trace-gas data from 2002-2012. More positive trends were observed for CO2 and CH4 in agreement with global patterns, whereas O3 exhibited non-significant positive trends in the recent period, consistent with findings from previous studies. CO trends were less conclusive due to the influence of sporadic biomass burning events, which complicate long-term trend detection."

Introduction Comment: Not enough credit and context to other studies. The section about Lamto and Rwanda measurements sounds rather dismissive.

More appropriate reference was made to the Rwanda, Lamto and Maido observations.

"In addition to the continental East African site MKN, the Maïdo station (2155 m a.s.l) on Réunion Island (Callewaert et al., 2022) and the Lamto station (155 m a.s.l.) in Côte d'Ivoire (Tiemoko et al., 2021, 2023) provide tropical data on GHGs and air pollutants. Observations of CO2, CH4, and CO at Réunion Island showed that surface mole fractions are dominated by local influences (urban emissions at Saint-Denis, and biospheric fluxes at the high-altitude Maïdo station), whereas column measurements mainly reflect long-range transport from Africa, Madagascar, and distant biomass-burning regions. Because Lamto is a low-elevation site, multi-year observations showed that local wetland emissions, agricultural activities, and seasonal biomass burning, together with regional seasonal transport patterns, strongly shaped the variability of CO2, CH4, and CO in West Africa."

Suggested Introduction Edits-

*The World Meteorological Organization's Global Atmosphere Watch (GAW) Programme is one of only a few internationally coordinated initiatives (NDACC, de Maziere et al. [2018] and its affiliated networks, e.g., AGAGE, GML's HATS, SHADOZ), dedicated to long-term, systematic observations of atmospheric composition on a global scale (World Meteorological Organization (WMO), 2014). *Through a network of hundreds of stations, GAW delivers high-quality data from spatially representative sites that monitor atmospheric conditions, with an emphasis on sites with minimal local influence. This global framework is essential for understanding large-scale patterns and long-term trends in atmospheric composition. However, despite its wide reach, significant observational gaps remain—particularly across tropical regions and the Global South (*consider omitting this expression; it can be controversial).* Africa, though one of the most climate-vulnerable continents, is typically under-represented in atmospheric monitoring networks, including for greenhouse gases (GHGs), primarily due to different national priorities, limited resources and infrastructure in emerging economies.

Continuous GHG observations are essential for verifying and reducing uncertainties in bottom-up emission estimates, as demonstrated in Europe (Henne et al., 2016; Saboya et al., 2024) or other regions (Bukosa et al., 2025). Despite a few monitoring stations (e.g. Morgan et al. (2015); Labuschagne et al. (2018); Tiemoko et al. (2023)), much of Africa still lacks the comprehensive GHG monitoring needed for robust emission assessments. This study explores the lessons learnt from the longterm data measured at the remote, high-altitude GAW station on Mount Kenya (*give latitude/longitude, alt)*. Besides an extensive data analysis to investigate atmospheric variability, we simulate air masses with an atmospheric transport model and combine them with emissions from bottom-up emission inventories. This integrated approach allows us to explore not only temporal variability but also the spatial and sectoral origins of the observed species. This assessment is made with full recognition that a denser GHG observation network across the continent is ultimately needed for robust verification and constraint of emission estimates.

The Mount Kenya station (MKN), operational since 1999, represents a unique monitoring site in tropical Africa. The recurrent meridional migration of the Intertropical Convergence Zone (ITCZ) that oscillates between approximately 20 °N and 5–8 °S depending on boreal and austral seasons (Henne et al., 2008a; Lashkari and Jafari, 2021; Hu et al., 2007), exposes the station to fundamentally different advection regimes throughout the year. These include continental air from the northeast during boreal winter and marine tropical air from the southeast in boreal summer. Moreover, more local anthropogenic processes

and biomass burning emissions also influence the station, particularly during daytime………*Summarize the Henne et al, 2008b findings here. More recently a global tropospheric ozone study included evaluation of east African ozone trends for more than 20 years of Nairobi ozone soundings. Total tropospheric ozone changes (surface to tropopause) were estimated at ~(1.5-3.5)%/dec for the period ~2000-2023 (Thompson et al., 2025; Van Malderen et al., 2025) with the lower value representative of a trend at the altitude corresponding to Mt Kenya. Increases near the surface were closer to + 5%/decade because Nairobi is a polluted city of ~4 million.*

The continuous and comprehensive greenhouse gas and air pollution datasets at MKN are unprecedented in the tropical African region and underline the importance of the MKN measurement site. While early carbon monoxide (CO) and surface ozone (O3) data were reported previously (Henne et al., 2008b), the renewal of the power line has enabled largely gap-free, continuous aerosol measurements since 2015, and measurements of carbon dioxide (CO2), methane (CH4), CO, and surface O3 since December 2019. Multiple trace gases were measured with flask samples at MKN by the Global Monitoring Lab (GML) of the National Oceanic and Atmospheric Administration (NOAA) from 2003 to 2011 (e.g. Lan et al. (2025)), but were not continued afterwards. Kirago et al. (2023) investigated MKN CO in-situ and flask measurements up until 2022, but more recent years and other species have not yet been explored. Indeed, this study presents the first comprehensive analysis of the recent continuous datasets, focussing on the period 2020 to 2024.

Few studies have investigated similar compounds in the region. DeWitt et al. (2019) studied GHGs and air pollutants at the Rwanda Climate Observatory (Mt. Mugogo, 2590m a.s.l), but data were limited to 2015-2017. Earlier black carbon (BC) and aerosol measurements in East Africa have been analysed at urban and rural sites (Kirago et al., 2022; Gatari and Boman, 2003; Makokha et al., 2017; Khamala et al., 2018), but the recent multi-year BC data from MKN were not included. In addition to the continental East African site MKN, the Maido station (2155m a.s.l) on Reunion Island (Callewaert et al., 2022) and the Lamto station (155m a.s.l.) in Cote d'Ivoire (Tiemoko et al., 2021, 2023) provide tropical data on GHGs and air pollutants. However, the former cover only 20 months of Maido data, and Lamto is strongly influenced by local sources due to its low altitude. The continuous, remote MKN measurements therefore fill a critical gap in the tropical observation network.

In this study, we (i) analyse in-situ trace gas and aerosol measurements at MKN, (ii) evaluate the performance of Copernicus Atmospheric Monitoring 60 Service (CAMS) model products for several species *(suggest putting this last)*, (iii) compare surface ozone with vertical profiles from ozonesondes launched in Nairobi, (iv) simulate atmospheric transport using the particle dispersion model FLEXPART, and (v) combine transport simulations with bottom-up emission inventories for fires, wetlands, and anthropogenic sources ……

The introduction was thoroughly revised and extended following the reviewer's suggestions.

Most relevant additions were:

"The WMO Global Atmosphere Watch (GAW) Programme of the World Meteorological Organization (WMO) is a truly international framework dedicated to long-term, systematic and global observations of atmospheric composition (World Meteorological Organization (WMO), 2014). At the same time, GAW is complemented by several more specialized international networks that focus on particular species, techniques, or regions. Among these, the Network for the Detection of Atmospheric Composition Change (NDACC) (De Mazière et al., 2018), a contributing network to GAW, coordinates high-quality remote-sensing observations, and NDACC in turn includes affiliated cooperating networks such as AGAGE (Advanced Global Atmospheric Gases Experiment) (Western et al., 2025), SHADOZ (Southern Hemisphere Additional OZonesondes) (Thompson et al., 2025), and the NOAA Halocarbons and other Atmospheric Trace Species (HATS) program (Montzka et al., 1999). Together, these networks of hundreds of stations in total form an integrated global observing system that strengthens and broadens the capabilities of GAW."

and

"*Based on early carbon monoxide (CO) and surface ozone (O3) between 2002 and 2006, Henne et al. (2008b) showed that night-time observations reliably represent free-tropospheric background air. Using trajectory clustering, the authors identified six distinct regional flow regimes that drive the site's semi-annual CO cycle, including northern-hemisphere winter outflow and southern-hemisphere biomass-burning influence. Overall, the weak O3 to CO correlations and rare pollution events demonstrated that Mt. Kenya provides a baseline perspective on tropical free-tropospheric composition, with interannual variability largely governed by southern African biomass-burning emissions. More recently a global tropospheric O3 study included evaluation of east African O3 trends for more than 20 years of Nairobi O3 soundings. Total tropospheric O3 changes (surface to tropopause) were estimated at approximately 1.5 to 3.5% per decade for the period of 2000 to 2023 (Thompson et al., 2025; Van Malderen et al., 2025) with the lower value representative of a trend at the altitude corresponding to Mt Kenya. Increases near the surface were closer to +5% per decade because Nairobi is a polluted city of more than 4 million inhabitants.*"

References to be included: …

The reviewer's suggested references, along with additional literature supporting the trend discussion, have been added to the manuscript.

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Replies to review RC2 from C. Labuschagne, 13 Feb 2026

We thank Reviewer 2 for his careful assessment and constructive suggestions. Below, we provide the answers to his comments (in blue).

Reviewer 2, general comments:

This study of Bernet *et al*, presenting data from a renewed effort for continuous of multi-year in-situ measurements of trace gases and black carbon from the Mount Kenya GAW station (MKN) is well articulated and presented. […] The introduction section explained precisely what the authors had set out to do […] In this regard, the various objectives of the paper were achieved.

Thank you for your positive feedback.

It is noted that in-situ observations at MKN have been conducted since the early 2000s, with a focus in this work on the continuous data from the more recent 2020 to 2024 time period. It would be useful to extend the discussion a bit further to also include the first period of measurements, but due to the large data gaps it might not be possible and will introduce more bias to the overall data set. One simply cannot extend trend calculations over periods where such large data interruptions have occurred. I think the authors should use this argument to explain / elaborate on this, in order to clarify the reason(s) for not extending the discussion to include the entire data set since inception.

The discussion of trends covering the full dataset is now expanded, also in view of the comments of reviewer #1.

With regards to the long-term greenhouse gas measurements at MKN, the authors managed to show increasing trends consistent with global patterns. They should perhaps expand that aspect of the discussion bit more, and mention & discuss the tropical derived values against the global background observations. This will further highlight the need for tropical and equatorial observations.

An extended discussion of long-term trends is now added to section 3.2.

Reviewer 2, specific comments:

Figure 1 – please enlarge (similar sizing as Fig. 2) and add a placemark for the MKN station.

Thanks for this valuable suggestion. The domain of panels shown in Figure was enlarged to match the domain shown in Figure 2.

Line 283: correct typo “ground”

done

Line 344: Please rephrase – I did not see the 2x minima for CO2 clearly, however, the general remark of NE – SE seasonal displacement of the ITCZ still holds.

The seasonal CO2 pattern is discussed in a separate paragraph thirty lines below (lines 372 ff. of the initial manuscript). It is now clarified in the beginning of the seasonality section that the opening statements do not hold true for all species.

Line 397: Please check the statement related to CO2 diurnal amplitudes – I could not easily discern the 417PPM CO2 and stated ~4PPM reduction during nighttime.

Thank you for spotting the inconsistency between the numbers shown in Fig. 4 and those mentioned in the text. The text referred to an earlier version of the analysis that did not yet include the final year (2024), which explains why the values no longer matched the figure. We have revised the text accordingly and removed the reference to the absolute nighttime values. The updated sentences now read: "*Nighttime CO2 values drop rapidly by approximately 3 to 4 ppm after sunrise (Fig. 4a) with larger decreases typically observed during the wet season. These daytime reductions reflect lower CO2 levels in the PBL driven by ecosystem uptake*.".

Line 444: correct typo CH4 (subscript)

done