Response to the reviewer

Reviewer 2

Recommendation: Major Revision

The manuscript addresses an important and relevant topic and provides a useful continental-scale assessment of black carbon over Africa using AerChemMIP models, MERRA-2, fire data, meteorological variables, and trajectory analysis. The study has potential value; however, several methodological and interpretational issues should be addressed before publication.

We sincerely thank the reviewer for the positive assessment and for recognizing the importance of this stud. We have carefully considered the reviewer's concerns and revised the manuscript accordingly to improve methodological clarity and ensure that the interpretation of the findings is appropriately supported by the results. Our point-by-point responses to the specific comments are provided below and the corresponding revisions have been incorporated into the revised manuscript.

Major Comments

1. MERRA-2 is used both as the reference dataset for model evaluation and as the basis for bias adjustment. This creates a degree of dependence between the correction and subsequent validation. The authors should clearly acknowledge this limitation and avoid presenting the agreement with MERRA-2 as independent validation.

We thank the reviewer for highlighting this important methodological limitation. We acknowledge that using MERRA-2 both as the reference dataset for model evaluation and as the basis for bias adjustment introduces a degree of dependence between these procedures. To address this concern, we removed the ANOVA-based assessment of bias-adjustment effectiveness from the revised manuscript because evaluating the adjusted simulations against the same reference dataset would not provide an independent assessment. We have also explicitly acknowledged the uncertainties associated with using MERRA-2 for both model evaluation and bias adjustment, given that it is a reanalysis product rather than a direct observational dataset. The revised manuscript clarifies this limitation and emphasizes the need for caution when interpreting the bias-adjusted results and future projections. For instance, lines 142-148 of the revised MS as" Although MERRA-2 provides a consistent long-term aerosol record and used as reference data, for models evaluation and bias adjustment, uncertainties remain in its representation of surface BC concentration. These uncertainties arise partly from the representation of BC emissions, atmospheric transport, vertical mixing and removal processes in the underlying model (Randles et al., 2017; Buchard et al., 2017). Consequently, discrepancies between MERRA-2 and AerChemMIP simulations may reflect uncertainties in both datasets rather than model errors alone. In this study, MERRA-2 was used as the reference for model evaluation and bias adjustment; however, the adjusted simulations were not independently evaluated against surface BC observations because of the limited availability of long-term measurements across Africa during the evaluation period. Therefore, the model evaluation quantifies discrepancies between the AerChemMIP simulations and the MERRA-2 reference dataset rather than errors relative to independent observations."

2. The historical analysis is stated as 1981–2014, while Figure 4 reports a long-term mean for 1980–2023. Similarly, the burned-area period differs between the main manuscript and supplementary material. These inconsistencies should be carefully checked and corrected.

The authors have revised and corrected the mistakes, the historical analysis starts from 1981 and ends on 2014. Figure 4 is revised as "Figure 4. Spatial distribution of (a) long-term mean (1981-2014) and (b) regional average of BC concentration. Full names of the regions are North Africa (NA), West Africa (WA), Central Africa (CA), East Africa (EA), Southern Africa (SA) and Madagascar Island (MD)."

3. Some equations, particularly the Pearson correlation and Index of Agreement, should be carefully checked against their standard definitions and against the actual calculations used. The statistical treatment should also consider temporal autocorrelation and, where appropriate, the issue of multiple spatial significance testing.

We have revised all the equations and we acknowledge the mistakes observed by the reviewer. Pearson correlation (r) and Index of Agreement (IA) have been revised as:

$$r = \frac{\left[1/(n-1)\right]\sum_{i=1}^{n}[(y_{re}-\bar{y}_{re})(y_{mod}-\bar{y}_{mod})]}{\left\{\left[1/(n-1)\right]\sum_{i=1}^{n}(y_{re}-\bar{y}_{re})^2\right\}^{1/2}\left\{\left[1/(n-1)\right]\sum_{i=1}^{n}(y_{mod}-\bar{y}_{mod})^2\right\}^{1/2}} \quad (1)$$

$$IA = 1 - \frac{\sum_{i=1}^{n}(y_{re}-y_{mod})^2}{\sum_{i=1}^{n}(|y_{mod}-\bar{y}_{re}|+|y_{re}-\bar{y}_{re}|)^2} \quad (6)$$

4. Four models are selected based on an Index of Agreement threshold of 0.7, while GISS-E2-1G is excluded. The authors should provide a stronger justification for this threshold and discuss how excluding one model affects the resulting multi-model mean and uncertainty.

We thank the reviewer for this important comment. We would like to clarify that an IA threshold of 0.7 was not used as a predefined criterion for model selection. > 0.7 for Index of Agreement (IA) is mentioned as how high other models perform Rather, model selection was based on a comprehensive evaluation of performance using correlation, mean bias, normalized mean bias, root mean square error, normalized root mean square error and the IA. Across these metrics, GISS-E2-1-G exhibited comparatively poorer performance than the other models, including substantially lower IA (approximately 0.25) and larger negative biases across extensive parts of Central and West Africa. These results collectively informed its exclusion from the multi-model mean, rather than any single metric or predefined threshold. We have revised the manuscript to clarify this multi-metric selection rationale. We have also clarified that including GISS-E2-1-G would have affected the MMM and inter-model spread due to its poor performance. in lines 255-261 of the revised MS as "The IA analysis (Fig. S2) further highlights differences in model performance, where MIROC6, MRI-ESM2-0, CESM2-WACCM and NorESM2-LM exhibited high IA values (> 0.7), whereas GISS-E2-1-G had a substantially lower value of approximately 0.25. MIROC6, MRI-ESM2-0, CESM2-WACCM and NorESM2-LM were therefore selected for the MMM based on their overall performance across correlation, MB, NMB, RMSE, NRMSE and IA. GISS-E2-1-G was excluded because of its comparatively poorer overall performance, particularly its substantial underestimation of BC concentration across extensive parts of Central and West Africa as it could influence the MMM and inter-model spread. It can however be noted that selected models also exhibited biases relative to MERRA-2 (Figs. 2 and S2); therefore, the MMM was subjected to bias adjustment using the method described in Section 2.2.1."

5. The lack of a statistically significant difference between the bias-adjusted MMM and MERRA-2 should not by itself be interpreted as proof that the bias correction is "effective" or that the two datasets are equivalent. This interpretation should be revised and supported with appropriate performance metrics.

We thank the reviewer for this important comment. We agree that the lack of a statistically significant difference between the bias-adjusted MMM and MERRA-2 does not, by itself, demonstrate the effectiveness of the bias adjustment or establish statistical equivalence between the two datasets. Accordingly, we have removed the analysis testing the effectiveness of the bias adjustment because independent data were unavailable to validate the adjusted MMM. We have revised the manuscript accordingly to avoid drawing conclusions about the effectiveness of the bias adjustment that cannot be independently verified

6. The relationships between BC and burned area, as well as the PSCF and trajectory results, provide useful evidence about possible source regions. However, these methods do not provide quantitative source apportionment. Statements concerning specific emission sectors should therefore be presented more cautiously unless supported by additional emissions data.

We thank the reviewer for this helpful comment. We have revised the interpretation of the results to avoid causal language and to present the relationships between BC and burned area, as well as the PSCF and trajectory results, as evidence of potential source regions and transport pathways rather than quantitative source apportionment. We have also revised statements concerning specific emission sectors to avoid attributing BC concentrations to particular sources without sufficient supporting evidence. For instance, lines 342-350 of the revised MS as "PSCF results (Fig. 6c) further identify northern regions, extending into parts of Europe, as potential source regions associated with elevated BC concentration over North Africa. High PSCF values were mainly observed over eastern Europe during DJF and MAM (Fig. 6c). During JJA, when BC concentration peaks, high PSCF values extended north-eastward from the tracking location toward the Mediterranean Sea. This spatial pattern suggests that the transport of air masses from northern and Northeastern regions may have contributed to the elevated BC concentration over North Africa. Furthermore, given the intensive maritime activity in the Mediterranean region, shipping emissions may also represent a potential contributing source. These results hence suggest a potential association between elevated BC concentration over North Africa and air masses originating from northern regions, including polluted areas of Europe, as well as the possible influence of maritime transport over the Mediterranean Sea."

7. The manuscript sometimes moves directly from spatially elevated BC concentration to a specific emission source (e.g., industry, traffic, mining, or biomass burning). However, atmospheric concentration reflects the combined effects of emissions, transport, dispersion, chemical & physical removal, and meteorological conditions. The authors should distinguish more clearly between where BC is concentrated and where it originates.

We have revised the presentation of the BC distribution to better distinguish between regions of elevated BC concentration and potential emission sources. We have also incorporated the role of meteorological conditions in influencing the regional distribution of BC and included relevant references to previous studies documenting the long-range transport of biomass-burning aerosols across Africa. For instance, lines 282-285 of the revised MS as; "Apart from emission sources, meteorological conditions also influence the regional distribution of BC across Africa by modulating atmospheric transport and dispersion. Previous studies have documented the long-range

transport of biomass-burning aerosols from parts of Southern toward Central and West Africa (Sinha et al., 2004; Haslett et al., 2019, Nyasulu et al.,2021), highlighting the role of regional atmospheric circulation in shaping aerosol distribution such as BC over the study domain." Furthermore, we already detailed meteorological influence in the seasonal analysis, where variations in atmospheric circulation and other meteorological conditions play important roles in BC accumulation and transport, please see section 3.4. Throughout the revised manuscript, we have adopted more cautious language when interpreting potential emission sources and transport pathways, avoiding definitive source attribution where it is not directly supported by our analyses.

8. Bias adjustment may affect the interpretation of hotspots. The authors should clarify how much of the identified hotspot structure is derived from the original models versus the bias-adjustment procedure.

We thank the reviewer for this important comment. The spatial comparison between the unadjusted and bias-adjusted MMMs shows that the principal BC hotspots are already present in the original model simulations and are retained after bias adjustment. In particular, elevated BC concentrations over Central Africa and West Africa, especially Nigeria, are evident in both MMMs. However, bias adjustment modifies the spatial distribution and magnitude of BC concentrations, resulting in more pronounced elevated concentrations over northern Egypt, central Ethiopia, and northern South Africa. Thus, the main hotspot structure is derived from the original model simulations, whereas bias adjustment modifies the regional expression of this structure. We have revised the manuscript to clarify this distinction. We have also included the figure (Figure 3) with both unadjusted and the bias-adjusted, the revised figure as;

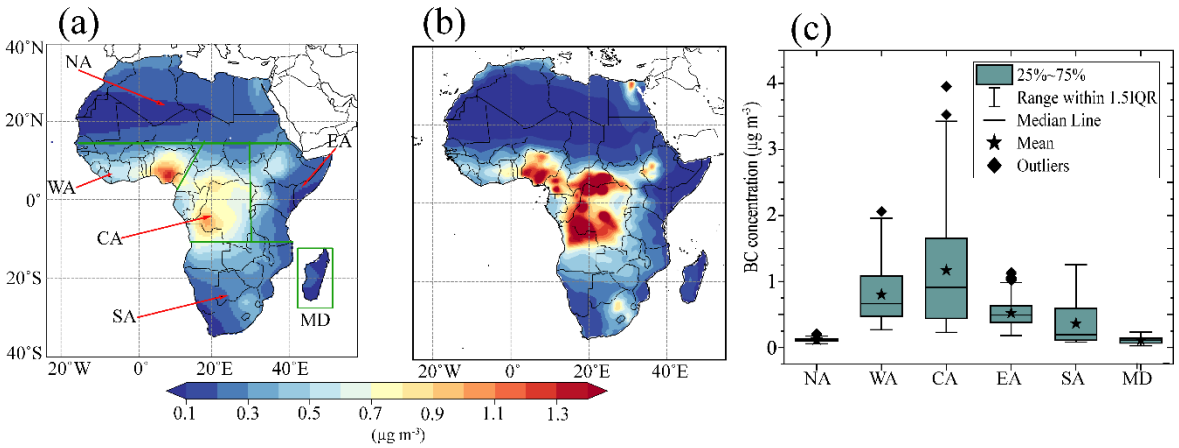


Figure 3. Spatial distribution of BC concentration for (a) bias unadjusted multi-model mean (MMM), (b) bias adjusted MMM and (c) regional average. Full names of the regions are North Africa (NA), West Africa (WA), Central Africa (CA), East Africa (EA), Southern Africa (SA) and Madagascar Island (MD).

The revised texts lines 268-270 as; "The annual MMM (Fig.3a-b) revealed high concentration of BC in Central, followed by West Africa, specifically over Nigeria for both bias-unadjusted MMM and bias-adjusted MMM. However, the bias-adjusted MMM additionally reveals elevated BC concentrations over northern Egypt, central Ethiopia, and northern South Africa, indicating that bias-adjustment modifies the spatial distribution and extent of BC .hotspots."

9. The manuscript associates dry-season BC maxima primarily with biomass burning and wet-season reductions with reduced burning and wet deposition. This is plausible, but the observed seasonal covariance does not establish that biomass burning is the dominant cause everywhere. Meteorological transport, boundary-layer height, precipitation, and regional circulation may also strongly control the seasonal cycle.

We agree that meteorological conditions play an important role in modulating the seasonal distribution of BC in Africa. We have also detailed already on how changes in in meteorological conditions are associated with the seasonal distribution of BC in Section 3.4 of the revised manuscript, by considering changes in wind speed and direction, mean sea-level pressure, boundary-layer height and precipitation. Please see section 3.4 of the MS.

10. Some increasing BC trends are attributed to industrialization, fossil-fuel use and traffic, but sector-specific emission data are not directly analysed. These explanations should either be supported by additional evidence or expressed as possible contributing factors.

We have carefully revised the presentation and interpretation of the results, expressing these emission sources as possible contributing factors rather than established causes of the observed trends. In the revised MS lines 386-387 as; These increasing trends may be associated with expanding industrial activities, increased fossil-fuel use and growing traffic emissions in these regions amid rapid economic development in recent years (Anand et al., 2024; Chiloane et al., 2017).” Line 397 as; “These abrupt trend shifts indicate turning points in BC concentration levels across Africa and may be associated with regional changes in industrial activities, traffic emissions and biomass-burning practices (Keita et al., 2021).”

11. The manuscript uses terms such as “driving factors” and makes causal interpretations based mainly on correlations between BC, burned area and meteorological variables. Correlation alone does not establish causation. I suggest replacing strong causal wording with “associated factors” or “possible contributing factors” where direct attribution has not been performed.

Response: We have revised the entire manuscript to replace strong causal expressions with more cautious terms, including “associated factors” and “possible contributing factors,” wherever direct attribution was not performed to ensure that our interpretations remain consistent with the evidence provided by the correlation analyses.

12. The future results should include clearer information on inter-model spread or agreement. Since the analysis is based on a selected subset of models, the projections should be described as scenario-based projections rather than predictions.

Response: We have revised the projections following the reviewer’s suggestion and describe the projections as scenario based rather than predictions. For instance, Lines 405-415 of the revised MS as; “The projected trends under the SSP370SST scenario (Fig.9) indicate predominantly positive BC trends across North and West Africa during the near-term (2015-2040), with an increase exceeding ~0.5-1.0% yr^{-1}, suggesting a potential intensification of BC concentration under this scenario. In contrast, Central and Southern Africa exhibit weak to moderate negative trends, suggesting potential localized emission reductions. By mid-century (2041-2070), positive trends persist and strengthen over West Africa and parts of North Africa, indicating these regions are future hotspots of BC, hence require enhanced intervention under the SSP370SST scenario. On the other hand, Southern Africa shows negative trends, which might reflect mitigation progress. Central, East Africa and

Madagascar Island project mixed patterns during mid-century, indicating emerging regions of concern where emissions may increase without targeted policies. In the late-century period (2071-2100), a marked shift occurs, with North Africa transitioning to negative trends and widespread declines across Central and Southern Africa., In contrast, localized positive trends persist in parts of East Africa and Madagascar, indicating areas where BC concentration could continue to increase under SSP370SST and where targeted mitigation efforts may be particularly important."

13. The conclusions recommend region-specific emission-control strategies based on the identified BC hotspots and trends. While this is a reasonable implication, the study does not quantify individual emission sectors or/& their contributions. The policy recommendations should therefore be presented as indicative rather than directly derived from quantitative source attribution.

We appreciate the reviewer for the constructive suggestion, we have revised and present recommendations as indicative rather than directive. Lines 456-461 of the revised MS as; "These findings highlight the importance of considering region-specific BC mitigation strategies, including potential control on industrial activities, traffic emissions and household energy use, particularly in regions exhibiting persistent or increasing BC concentration. However, these implications are scenario-based on the projected spatial and temporal patterns of BC concentration under SSP370SST, rather than conclusions derived from quantitative attribution to individual emission sectors. The findings hence provide a potential regional evidence base to inform future air pollution mitigation planning and support further research into the contribution of different emission sources to BC trends across Africa."

Minor Comments

1. Use MERRA-2 reanalysis dataset rather than "observed BC" where appropriate.

We have revised the entire MS with "observed BC" presented as "MERRA-2 reanalysis"

2. Clearly define "near-surface" BC and the atmospheric density used to convert BC mass mixing ratio to concentration.

We have defined "near-surface" in the revised MS, line 98 as; "In this study we used near surface BC concentration, defined as the BC concentration represented by the lowest vertical level of the atmospheric model output (Turnock et al.,2020)." We have also included the details of converting BC mass mixing ratio into concentration in the supplementary materials. S1 of the supplementary materials as;

"The AerChemMIP models simulate mass mixing ration of BC (BC_{mmr}) particles in the air (kg kg^{-1}), therefore, the simulated BC_{mmr} was converted into concentration (kg m^{-3}) by multiplying BC_{mmr} with the density of the air, expressed as:

$$BC_{con} = BC_{mmr} \times \rho_0 \quad (S1)$$

Where ρ_0 is the density of the air. At standard pressure (1000hPa) and temperature (273.15 K), the density of air (dry) is 1.295kg m^{-3}. However, the density of the air is dependent of temperature and pressure in the atmosphere which affects concentration of BC (Xu et al., 2022). Density from Eq. S1 was therefore expressed as:

$$\rho_0 = \frac{p}{RT} \quad (S2)$$

Where p is the ambient pressure, R and T are the specific gas constant and temperature respectively. For dry air, R = 287.058 J $kj^{-1}K^{-1}$. For consistence, both temperature and pressure were extracted from the models during

conversion. The final BC mass concentration was expressed in micrograms per cubic metre (μg m^{-3}) by multiplying the concentration in kg m^{-3} by a conversion factor of 10^9." We have also clarified in the main text of the revised MS details are provided in the supplementary materials, please see line 138 of the revised MS.

3. State whether regional averages are area-weighted.

We used only area average

4. Define precisely what is meant by "near-surface BC."

Same as in previous comment above, we have defined "near-surface" in the revised MS, line 98 as; "In this study we used near surface BC concentration, defined as the BC concentration represented by the lowest vertical level of the atmospheric model output (Turnock et al.,2020)."

5. Provide the exact fire-product name and version consistently throughout the manuscript.

We have made a correction and provided name and version consistently throughout the manuscript, eg lines 132 of the revised MS as; Burning area data between 2002 and 2014 was sourced from 5th the version of Global Fire Emissions Database (GFED5) (van der Werf et al., 2025) to investigate influence of changes in burning activities on the distribution of BC over Africa. In the supplementary Figure as S1 as; "Figure S1. The map of Africa with the sub-divided regions represented by the dotted polygons (a) and the spatial distribution of annual banned area due to forest fires and total annual burned areas based on 5th version of Global Fire Emission Database (GFED5) between 2002 and 2014 (b)."

6. Resolve the discrepancy between the stated burned-area period and the period shown in

Supplementary Figure S3.

Same as previous comment above we have resolved the discrepancy, the period used in 2002-2014.

7. Define how the Sen's slope is converted into percentage per year.

We have defined how Sen's slope was converted to percentage per year, Lines 207 of the revised MS as; "In this study, Sen's slope at each grid cell was expressed as a percentage per year (% yr^{-1}) by dividing the estimated annual trend by the mean value of the variable over the study period and multiplying by 100."

8. Clarify the statistical significance criterion shown in Supplementary Figure S8.

We have clarified significance criterion, the revised Figure caption as; "Figure S8. Spatial distribution of BC trends estimated using Sen's slope and the Mann–Kendall (MK) trend test for 1981-2014. Black dots indicate grid cells with statistically significant trends at the 5% significance level ($p < 0.05$)."

9. Correct apparent typographical errors such as "Mitterrandian Sea" to "Mediterranean Sea."

"Mitterrandian Sea" has been modified to "Mediterranean Sea.", please see line 350 of the revised MS.

10. Please specify the trajectory starting heights and duration in the Supplementary Figure S6 caption & Methods.

We have revised the figure caption, specified starting height and duration and also updated the Method section in the supplementary materials. The revised figure caption as; "Figure S6. Five-day backward trajectories arriving at the observation site (red dot), calculated with a starting height of 500 m above ground level (AGL) for the period January-December 2023.". in the method as;

S2. Airmass trajectories and potential source contribution function

The study has employed the backward airmass trajectories and potential source contribution function (PSCF) model (Ashbaugh, 1985) to model the distant source of BC, specifically over North Africa. We calculated the 5-day backward trajectories arriving at the tracking site, 500 m above ground level (AGL) ending at 12:00 h UTC were calculated for every 6 h (00:00, 06:00, 12:00 and 18:00 h UTC) using the Global Data Assimilation System (GDAS) meteorological data between January and December 2023. TrajStat, a GIS-based software tool was used to visualize air-mass trajectories (Wang et al. 2009). To track the potential sources, PSCF was used."

11. The captions of the supplementary figures should state the relevant data period and dataset wherever possible.

Response: We have revised figure captions and include the relevant data period and dataset wherever possible.

12. The climate discussion should distinguish BC concentration from radiative forcing, since radiative forcing is not calculated in this study.

Response: We thank the reviewer for this comment. The revised manuscript does not discuss radiative forcing, as it was not calculated in this study.

13. The reference list should be carefully checked for typographical and bibliographic inconsistencies.

We have carefully revised all the references following reviewer's recommendation

14. Cross check figure numbering, captions, units, and all reported time periods.

We have carefully cross checked all the figures; all changes are tracked in red

15. The conclusions should be slightly moderated where causal attribution is not directly demonstrated.

We thank the reviewer for the comment. We have revised and moderated the language, where causal attribution is not directly demonstrated. For instance, Lines 443-460 of the revised MS as; "The seasonal distribution exhibit peaks during the dry seasons in most regions, strongly associated with seasonal biomass burning and prevailing meteorological conditions while long-range transport, such as from Europe is associated with BC concentration in North Africa." Line 453-460 as; "BC increase across North and West Africa and parts of East Africa and Madagascar through the mid-21st century could exacerbate air quality and contribute to regional atmospheric warming, potentially complicating climate mitigation efforts. In contrast, the projected declining trends across Southern and parts of Central Africa could improve air quality and suggest potential reductions in BC concentration, although the underlying drivers cannot be established from the concentration trends alone. These findings highlight the importance of considering region-specific BC mitigation strategies, including potential control on industrial activities, traffic emissions and household energy use, particularly in regions exhibiting persistent or increasing BC concentration."