Response to the reviewer

Reviewer 1

This manuscript presents the spatial distribution, seasonal variability, historical trends, and future evolution of surface BC over Africa using AerChemMIP simulations adjusted with MERRA-2. The models generally underestimate BC concentrations, particularly over Central Africa, and project continued increases across parts of North, West, and East Africa under SSP370SST. The regional focus is important because long-term BC measurements remain limited across Africa. However, the analysis contains substantial methodological and interpretive limitations, including the lack of independent evaluation using ground-based measurements, the assumption of stationary model bias, excessive reliance on correlation and statistical significance, and conclusions based on indirect associations rather than quantitative attribution. The manuscript also contains numerous grammatical, mathematical, and internal-consistency errors that substantially affect readability. Therefore, I do not recommend publication of the manuscript.

We sincerely thank the reviewer for the careful evaluation of our manuscript and for providing constructive comments. We appreciate the reviewer's recognition of the importance of investigating BC concentration across Africa, particularly given the limited availability of long-term BC measurements across the continent. We have carefully revised the manuscript to clarify the limitations associated with the lack of independent ground-based observations for model evaluation and the assumptions underlying the bias-adjustment procedure. We have also revised the interpretation results such as correlation to emphasize associations rather than causal relationships. Furthermore, we have thoroughly reviewed the manuscript to improve grammatical accuracy, mathematical consistency, scientific interpretation and the consistency of the reported results. The reviewer's comments have improved the clarity and overall quality of the manuscript, and we are grateful for the reviewer's comments. A point-by-point response is provided below, and all modifications in the revised manuscript (MS) are highlighted in red.

Major Comments

1. The study presents the final dataset as a bias-adjusted AerChemMIP estimate, but MERRA-2 is itself a model-based aerosol reanalysis constrained primarily by assimilated AOD rather than direct observations of surface BC. Several parts of the manuscript incorrectly refer to MERRA-2 as observed data. Although long-term network measurements remain limited across Africa, numerous individual field campaigns have provided surface BC measurements. These observations are not quantitatively included in the evaluation. Stating that estimated concentrations fall within the broad range of previous measurements is insufficient to establish model performance. The uncertainty in MERRA-2 is also not addressed despite its central role in model selection and bias adjustment.

We thank the reviewer for raising this important issue. We agree that MERRA-2 is a model-based aerosol reanalysis and should not be referred to as direct observations of surface BC. We have therefore revised the manuscript to consistently refer to MERRA-2 as a reanalysis or model-based reference dataset rather than as observational data. We have also included the limitation of applying MERRA-2 for evaluating and adjusting AerChem model bias. In the revised MS, lines 142-149 as; Although MERRA-2 provides a consistent long-term

aerosol record and used as reference data in the present study, for model's 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. Since the adjusted simulations were not independently evaluated against surface BC observations, the model evaluation quantifies discrepancies between the AerChemMIP simulations and the MERRA-2 reanalysis rather than errors relative to independent observations."

We would also like to acknowledge the challenge that even though a number of BC measurements have been reported in Africa, many of these measurements cover only short campaign periods such as days to moths and most are not publicly accessible. Furthermore, most observations are recent, between 2015 and 2025. Consequently, these observations cannot provide a homogeneous quantitative evaluation of the AerChemMIP models during the evaluation period (2004–2014).

2. The delta-change correction appears to be derived for 2004-2014 and then applied to 1981-2100. This assumes that the absolute model bias remains stationary for approximately 120 years, despite substantial changes in BC emissions, aerosol composition, meteorology, transport, and deposition. The correction procedure is also insufficiently described. It is unclear how the offset is calculated. An additive correction may also generate negative concentrations in regions with declining future BC. The role of the bias adjustment in the historical and future trend results is therefore unclear.

We thank the reviewer for highlighting methodological issue. We agree that applying an additive bias correction derived from a limited calibration period to the full 1981–2100 period requires careful justification, particularly because BC emissions, aerosol composition, meteorological conditions, transport and deposition processes may evolve over time. We have therefore clarified the bias-correction procedure and its limitations in the revised manuscript. Importantly, because the correction is a time-invariant additive offset at each grid cell, it only changes the mean concentration level but does not mathematically alter the temporal trend. We also acknowledge that the stationarity of the absolute model bias over the full 1981–2100 period cannot be assumed physically and have therefore revised the manuscript to avoid implying that the bias correction represents a time-varying or dynamically evolving model bias. The bias adjustment is instead used to reduce the mean systematic bias relative to MERRA-2, while the long-term trend analysis is interpreted primarily from the model simulations and its associated uncertainty.

Lines 190-198 as; "To adjust the systematic bias in the MMM, additive delta change method (Sperna et al., 2010; Wu and Huang, 2016) was applied. This approach has been shown to reduce median absolute errors between model simulations and observational or reanalysis datasets (Beyer et al., 2019). Although this approach can reduce systematic mean bias, it may not fully account for changes in model bias associated with evolving emissions, meteorological conditions, atmospheric transport, aerosol composition and deposition processes under a non-stationary climate (Iyakaremye et al., 2021). For each grid cell, the additive bias adjustment was calculated as the difference between the 2004-2014 mean BC concentration from MERRA-2 and the corresponding 2004-2014 mean MMM concentration. This grid-cell-specific adjustment, derived from the 2004–2014 reference period, was

then held constant and applied to the annual MMM BC concentration throughout the entire study period (1981–2100). It was expressed as;

$$y_{adj} = y_{mod} + (\bar{y}_{re} - \bar{y}_{mod}) \quad (7)$$

The manuscript places excessive emphasis on whether $p < 0.05$. The conclusion that bias adjustment is effective because the adjusted multimodel mean is not significantly different from MERRA-2 is not valid. A nonsignificant difference does not demonstrate equivalence. The comparison is also circular because the adjusted data are evaluated against the same dataset used to construct the correction. Spatially and temporally correlated gridded data further violate standard ANOVA independence assumptions. The biomass-burning attribution has similar limitations. Regional correlations vary substantially, but the resulting relationships are interpreted similarly. (Line 260). Correlation alone does not establish causation. Both BC and burned area have strong seasonal cycles, so correlations using raw monthly values may largely represent shared seasonality. Burned area is also not equivalent to BC emissions because fuel type, emission factors, and combustion efficiency vary. Similarly, the correlation between North African BC and European burned area does not establish transport from Europe. Correlation and descriptive comparison indicate association, not quantitative attribution.

Following further consideration and in light of the concerns raised by the other reviewers, we have removed the ANOVA-based assessment of the bias adjustment from the revised manuscript. Because the analysis did not rely on independent observational data, ANOVA was not considered an appropriate basis for evaluating the effectiveness of the bias adjustment. We have therefore removed the corresponding analysis and results to avoid drawing conclusions that are not adequately supported by the statistical approach.

We have also carefully revised the interpretation of the correlation results to clarify that they indicate associations rather than provide evidence of causal relationships in the entire manuscript. For instance, lines 315-317 of the revised MS as; "Furthermore, correlation analysis between BC and total burned areas (Fig. S4) revealed statistically significant positive correlation ($p < 0.05$) in all the regions, suggesting strong association between biomass activities and BC concentration, except over North Africa where non-significant positive correlation ($r \sim 0$, $p > 0.05$) was detected." Lines 333-334 as; "Results in Fig. S5a revealed a statistically significant positive correlation ($p < 0.05$) between BC extracted over North Africa and total burned areas of Europe. Additionally, their monthly pattern (Fig. S5b) exhibited a similar variation, with peaks occurring between July and October, suggesting their strong temporal positive association."

3. The Pearson correlation equation (Eq. 1) appears incorrect because the modeled values in the covariance term are centered using the reanalysis mean rather than the model mean. The Willmott index of agreement (Eq. 4) also appears to omit the leading "1−". If implemented as printed, larger errors would yield larger values, contradicting the stated interpretation that 1 is perfect agreement.

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:

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

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

Where y_re and y_mod are the MERRA-2 reanalysis and AerChem model simulations respectively while $\bar{y}$_re and $\bar{y}$_mod are their corresponding means during the period of evaluation.

Minor Comments

1. Line 81. The justification for using histSST rather than fully coupled historical simulations is not convincing. Suppressing ocean-atmosphere feedbacks does not isolate regional emissions, particularly when large-scale circulation and Mediterranean transport are discussed in the manuscript.

We thank the reviewer for raising this important point. We acknowledge that the prescribed-SST experimental design does not isolate regional emission effects and does not eliminate the influence of atmospheric circulation or long-range transport. We have revised the manuscript to clarify the justification and scope of the histSST and ssp370SST experiments in AerChem and also ensure that the interpretation of BC concentration patterns does not imply independent attribution to regional emissions or circulation. Lines 81-85 of the revised MS as; "In this study, we used the histSST and SSP370SST experiments, which prescribe sea-surface temperatures and thereby constrain interactive ocean-atmosphere feedbacks. This experimental design helps to assess changes in BC concentration under specified emissions and atmospheric forcing conditions, with reduced influence from coupled ocean-atmosphere variability. However, it does not isolate regional emission effects from changes in atmospheric circulation and transport, which can also influence BC distribution (Griffiths et al., 2025)."

2. Line 86. The description of how BC is simulated in each model is insufficient, particularly because the models may use different aerosol schemes, size representations, aging treatments, deposition parameterizations, and chemistry configurations.

We have added the description of how BC is simulated, in the revised MS lines 87-98 as; "Although AerChemMIP models follow common CMIP6 experimental protocols and emission inventories (Hoesly et al., 2018), they differ in their representation of aerosol processes and atmospheric chemistry. As summarized in Table 1, the models employ different chemistry configurations and aerosol schemes, including SPRINTARS, MASINGAR, MAM, and Oslo Aero (Yukimoto et al., 2019; Danabasoglu et al., 2020; Kelley et al., 2020; Seland et al., 2020; Shiogama et al., 2023). These schemes differ in their treatment of aerosol size distribution and microphysical processes, which can influence how BC is represented and evolves in the atmosphere. Differences in BC aging, including the conversion of hydrophobic to hydrophilic particles where explicitly represented, can affect aerosol-cloud interaction and the efficiency of wet removal. In addition, differences in wet and dry deposition parameterization, atmospheric transport and model physics can influence BC lifetime, spatial distribution and temporal variability. Consequently, inter-model differences in simulated BC concentration may arise not only from differences in emissions but also from the representation of atmospheric processes governing BC transport and removal."

3. Line 104. The exact MERRA-2 collection, temporal resolution, version, and screening procedure are not specified.

We have revised and include MERRA-2 collection, temporal resolution, version, and screening procedure. Lines 218-220 of the revised MS as; "In this study, we used monthly surface BC mass concentration from the MERRA-2 M2TMNXAER aerosol diagnostics collection (version 5.12.4) to evaluate AerChem models in simulating BC over Africa. During pre-processing, negative BC concentrations were excluded and only positive values were retained for subsequent analysis."

4. Line 143. Only absolute MB and RMSE are reported, while normalized metrics are not considered despite regional differences in mean BC concentrations.

We have included normalized MB and RMSE in the revised MS. In the revised MS lines 238-251 as; "The normalized bias indicates that the AerChem models generally underestimate BC concentration relative to MERRA-2 across much of Central, Southern and West Africa, as well as along parts of the Mediterranean coast. In contrast, positive NMB values indicate overestimation in parts of North and East Africa. The spatial pattern of underestimation overlaps with regions strongly influenced by seasonal biomass burning, including savanna fires (Keita et al., 2021), suggesting that uncertainties in biomass-burning emissions may contribute to the simulated BC deficits. Furthermore, several regions with negative NMB experience substantial high rainfall (Novella and Thiaw, 2013), suggesting that other factors such as differences in wet-deposition processes associated with high rainfall between the AerChem models and MERRA-2 may also contribute to the discrepancies. The RMSE and NRMSE results (Fig. 2d-e) further reveal that all selected models exhibit relatively high RMSE values across Central, parts of West, East and Southern Africa, indicating substantial discrepancies between the AerChem model simulations and MERRA-2 in these regions. In contrast, relatively low RMSE and NRMSE values, approaching zero across much of North Africa, indicate relatively smaller errors and closer agreement between the AerChem model simulations and MERRA-2 in these regions."

The revised figure as;

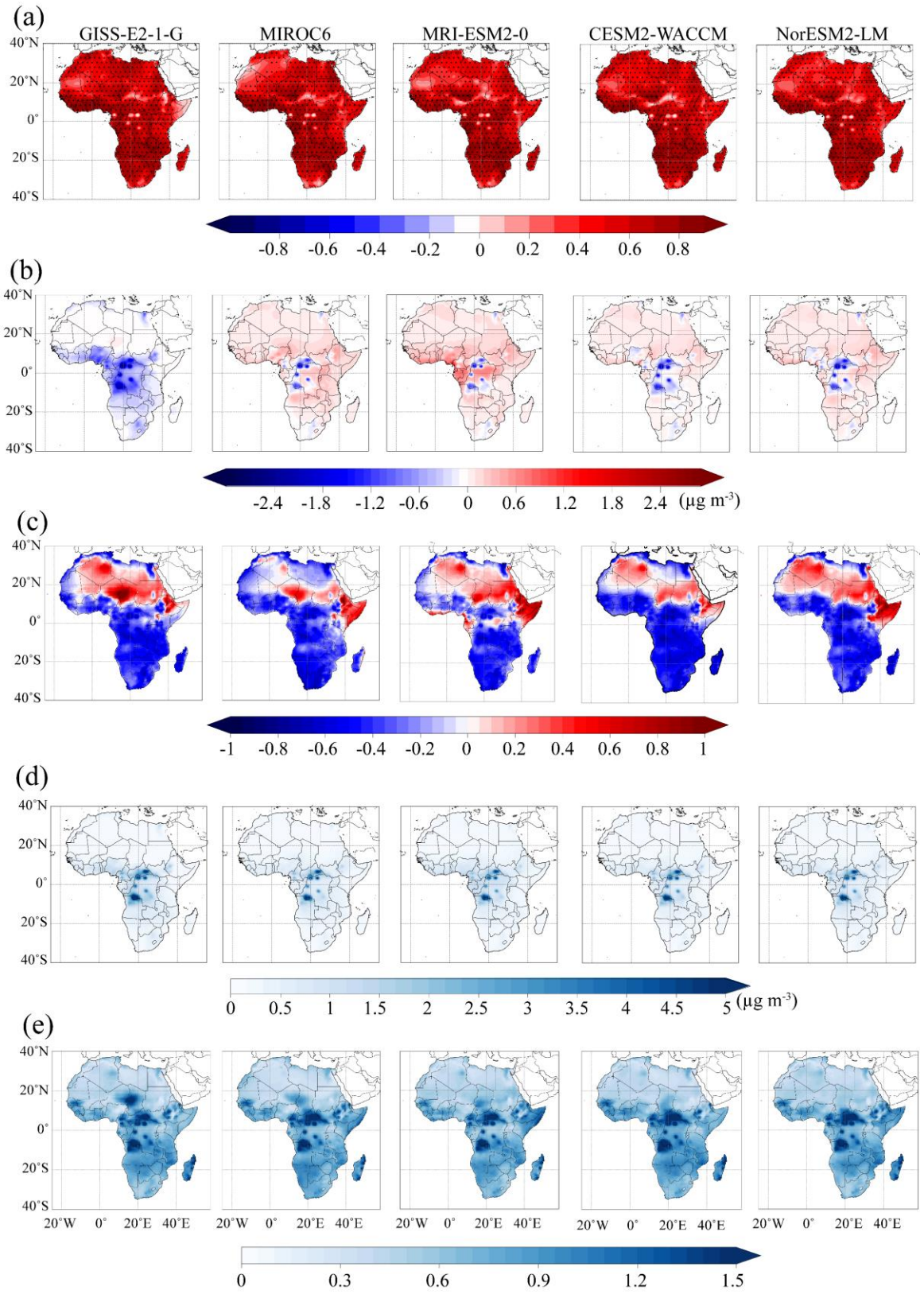


Figure 2. Spatial distribution of (a) correlation, (b) MB, (c) NMB, (d) RMSE and (e) NRMSE between AerChemMIP models (GISS-E2-1, MIROC6, MRI-ESM2-0, CESM2-WACCM, and NorESM2-LM) and MERRA2 during the evaluation period (2004-2014).

5. Line 157. The temporal basis of the grid-level correction is unclear.

We have clarified the temporal basis of the grid-level adjustment, in the revised MS lines 195-197 as; “For each grid cell, the additive bias adjustment was calculated as the difference between the 2004-2014 mean BC concentration from MERRA-2 and the corresponding 2004-2014 mean MMM concentration. This grid-cell-specific adjustment, derived from the 2004-2014 reference period, was then held constant and applied to the annual MMM BC concentration throughout the entire study period (1981-2100).”

6. Figs. 2 and 3. The figures present the same diagnostics for different models and could be combined.

We have combined the figures, please see figure 2 in the revised MS.

7. Line 203. The conclusion that the BC underestimation reflects underestimated emissions is not demonstrated. Other factors could also contribute.

We have revised and modified the conclusion, lines 240-245 of the revised MS as; “We noted that regions with underestimated BC concentration often coincide with areas affected by seasonal biomass burning, including savanna fires (Keita et al., 2021), suggesting that differences in the representation of biomass-burning emissions between the AerChemMIP models and MERRA-2 may contribute to the simulated BC deficits. Furthermore, several regions with negative NMB experience substantial high rainfall (Novella and Thiaw, 2013), suggesting that other factors such as differences in wet-deposition processes associated with high rainfall between the AerChem models and MERRA-2 may also contribute to their discrepancies.”

8. Line 211. The statement that GISS-E2-1-G “highly underestimated the spatial distribution” is unclear.

Response: The sentence has been revised to; “Generally, all models underestimate BC concentration in isolated parts of Central Africa, with highest magnitude of underestimation detected in GISS-E2-1G.” Please see line 233 of the revised MS.

9. Line 243. The text refers to Fig. 7, although the relevant results appear in Fig. 6.

We have made a correction and include relevant figure, in the revised MS as “Fig. 5”. Please see line 298.

10. Line 229. Residential cooking and heating emissions are not adequately considered.

We have revised the sentence where residential cooking and heating emissions are included, lines 278-276 as; “In addition, residential combustion of solid fuels, particularly wood and charcoal for cooking and heating, represents another important source of pollutants in many African communities (Adeeyo et al., 2022; Endalew et al., 2022; Eriksson et al., 2022), hence influencing BC concentration in the atmosphere.”

11. Line 290. The attribution to maritime shipping is speculative.

We thank the reviewer for this observation. We agree that attributing elevated BC concentration to maritime shipping without source-specific evidence would be speculative. Moreover, the method used (PSCF) is limited to identifying potential sources rather than quantifying the sources. We have revised the relevant discussion to avoid implying a direct source attribution and use of causal language. In the revised MS lines 248-250 as; "Furthermore, given the intensive maritime activity in the Mediterranean region, shipping emissions may also represent a potential contributing source. These results indicate 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."

12. Line 340. The historical trend discussion remains largely descriptive. General references to industrial activity, traffic emissions, and biomass burning cannot explain the distinct trend patterns among regions.

We agree that the discussion is descriptive because we did not analyse specific source contribution to the trends, however, we have revised the it 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). Furthermore, changes in regional meteorological conditions that influence wet deposition and atmospheric transport may also contribute to the observed BC trends; however, their relative contributions require further investigation.". Please see lines 397-100 in the revised MS.

13. The manuscript reports several conflicting analysis periods. In particular, Fig. 4 is described as representing 1980–2023, even though the historical AerChemMIP simulation ends in 2014. The stated GFED period in the Methods also conflicts with the Supplement caption.

We have made corrections to the mistakes, Fig.4. as; Figure 4. Seasonal spatial distribution of BC concentration over Africa for the period 1981-2014. Panels represent the four major global climatological seasons: December-February (DJF), March-May (MAM), June-August (JJA), and September-November (SON).", the time period in methods 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 the influence association between changes in burning activities on distribution of BC over Africa." and in the supplementary figure, Fig. S3 as; "Figure S3. Seasonal spatial distribution of burned area across Africa for DJF, MAM, JJA and SON between 2000 and 2014 based on Global Fire Emission Database (GFED5)."

Technical Corrections

Numerous grammatical errors are present throughout the manuscript. Examples include:

We have carefully revised the entire manuscript including grammar errors.

"Seasonal dynamics is dominated" in Line 17;

Revised as "Seasonal dynamics are dominated",

"large uncertainties on… remain high" in Line 41;

Revised as “Despite Africa’s importance in the global BC budget, substantial uncertainties remain regarding BC concentration levels”,

“knowledge gap undermines Africa’s effective development, region-specific mitigation strategies” in Line 57;

The entire sentence has been removed.

“Under SSP370,… slow technological development and high challenge to mitigation” in Line 78;

Revised as; “Under SSP370SST, economic development is slow, reliance on fossil fuels remains high, technological development is limited, and climate change mitigation faces substantial challenges.”,

“Lower values…indicates” in Line 141;

Revised as; “Lower values of RMSE and NRMSE indicate smaller errors relative to the reference concentration level, whereas higher values indicate larger relative discrepancies.”,

“to outperforms” in Line 154;

It has been revised as; “This approach has been shown to reduce median absolute errors between model simulations and observational or reanalysis datasets (Beyer et al., 2019).”

“The spatial distribution…show” in Line 184;

Revised as; “The spatial distribution of BC across all the models shows”

“exhibits the highest underestimation of compared to” in Line 188;

The sentence has been removed

“RMSE further support” in Line 205; “BC decrease” in Line 238;

The sentence has been removed

“These findings…is” in Line 242;

Revised as; “These findings demonstrate that BC has high seasonal variation across Africa and are consistent with previous studies”.

“a peak in BC concentration… is low” in Line 245;

Revised as; “In North Africa, BC concentration is observed high during JJA and low during MAM, while West Africa exhibits high BC during DJF and low during JJA.”

"West Africa… minima" in Line 245; "portions of BC was sourced" in Line 287;

Revised as; "In North Africa, BC concentration is observed high during JJA and low during MAM, while West Africa exhibits high BC during DJF and low during JJA." Please see line

"increase in BC…pose" in Line 342.

Revised as; "The significant increase (Z-score > 1.96) in BC observed in most regions poses substantial risks to both climate and public health, hence regionally tailored policies to reduce BC levels in the atmosphere are essential across Africa." Please see line 400.

The manuscript requires general language editing rather than correction of only the examples listed above.

We sincere appreciate for the constructive recommendation, we have carefully revised the entire manuscript and correct the errors.

Several typographical and terminology errors are also present. Examples include "SO4-2" in Line 96;

SO_4^{-2} modified to SO_4^{2-}

"conversional observation datasets" in Line 100;

Revised as; "conventional observational datasets"

"Global Fire Emissions (GFE) Database" in Line 118;

Modified to "5th the version of Global Fire Emissions Database (GFED5)",.

"mass mixing ration" in Line 123;

Modified to "mass mixing ratio",

"Montgomery.2017" in Line 158;

The sentence has been removed.

"Figs.2" in Line 211;

Revised as Fig. 2b

"mean sea level pressure (MSL)" in Line 302;

Revised as; "Figure 7. The composite distribution of BC, wind and MSLP between 1981 and 2014 during different seasons over Africa. The units for MSLP are in millibars (mb)."

“Mitterrandian Sea” in Line 293.

Modified to “Mediterranean Sea”,

SSP370SST is not defined in the Abstract,

We have revised as; “Future projections under Shared Socioeconomic Pathway 3–7.0, with prescribed sea surface temperatures and a radiative forcing level of approximately 7.0 W m^{-2} by 2100 (SSP370SST)”. Please see line 22.

y_{cor} is not defined after Eq. 5,

we have revised the equation, in the revised MS as equation 7;

$$y_{adj} = y_{mod} + (\bar{y}_{re} - \bar{y}_{mod}) \quad (7)$$

Where y_{adj} is the bias adjusted MMM.

PSCF is not defined in the main text.

We have revised and defined PSCF as; “potential source contribution function (PSCF)”, please see line 326.

“AerChemMIP” “AeroChemMIP” are both used, as are “CMIP6,” “CMIP-6,” and “CMIP 6.”

We have corrected the inconsistence; the right abbreviations are “AerChemMIP” and “CMP6”

Grid resolutions also use inconsistent symbols and spacing, including 0.25°x0.25°, 0.5° x 0.625°, and 0.5°×0.5°.

We corrected the error, in the revised MS as; “0.25°×0.25°, 0.5°×0.625°, 0.5°×0.5°”

Figure numbering, abbreviations, equations, captions, and reference formatting require comprehensive checking.

We have carefully revised them all, all modifications are highlighted in red.