Manuscript No.: egusphere-2025-5734

Responses to Reviewer #1

This study focuses on the critical environmental and health issue of near-surface ozone pollution in Africa. To address the scarcity of ground-based observational data across the continent, it integrates GEOS-Chem simulations, CMIP6 multi-model data, and a Random Forest (RF) model to construct a research framework with both interpretability and predictability. Innovatively, the study quantifies the independent contributions of climate-driven changes in meteorological conditions and biogenic isoprene emissions to ozone concentrations, thereby illuminating the compound health risks confronting Africa in the context of global warming. The manuscript is detailed, well-structured, and features clear conclusions, making it suitable for publication in the journal Atmospheric Chemistry and Physics. Here are some suggestions for the authors to improve the manuscript.

We thank the reviewer for all the insightful comments. Below, please see our point-by-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in an effort to take into account all the comments raised here.

1. Why did this study choose a hybrid approach combining a random forest model with a chemical transport model (CTM), rather than directly using a multi-model ensemble mean of machine learning models or a single CTM for long-term projections? What specific advantages does this hybrid modeling framework offer for this research?

Response:

In this study, GEOS-Chem model simulations were performed in a nested version of with a horizontal resolution of 0.5° latitude × 0.625° longitude and 47 vertical layers from the surface up to 0.01 hPa, with boundary conditions from 2° latitude × 2.5° longitude simulations. By utilizing the computational resources with 20 nodes (32 cores per node), the historical simulations for 2000–2019 (one node for each year) require more than 3 months. Therefore, the future simulation for the 18 selected CMIP6 models for 2020–2054 requires 3 months * 1.75 * 18 models * 4 scenarios = 31.5 years, which is too impracticable. However, by using the machine learning (ML) model, it only takes minutes for predicting. It will be more computationally expensive and time consuming by using the single chemical transport model (CTMs) compared with the ML method in this study. Therefore, we applied GEOS-Chem simulations as input data integrated with ML method, which could be a practical supplement to traditional CTMs. Conversely, directly using a multi-model ensemble mean of ML models lacks physical and chemical processes, which are important to the non-linear O_3 chemistry. In addition, Africa does not have sufficient O_3 observation for the ML model training.

2. The article mentions that most regions in Africa are in a "NO_x-limited" regime. In such an environment, why does an increase in biogenic isoprene emissions lead to a slight decrease in ozone concentrations?

Response:

Thank you for the comment. The isoprene is a major biogenic volatile organic compound (VOC) that predominates in forested regions such as Africa. In strongly NO_x-limited environments, where VOC are abundant, the increased isoprene emissions tend to reduce O_3 concentrations through oxidation processes and the formation of isoprene nitrates (Pacifico et al., 2012; Squire et al., 2014).

We have clarified in revised manuscript as follows: “In NO_x-limited regions, increased biogenic isoprene emissions tend to reduce O_3 levels through oxidation processes (Pacifico et al., 2012; Squire et al., 2014), resulting in a slight decrease in O_3 levels by less than 0.5 ppb over Central and Western Africa in 2050 relative to 2020 (Figure 7b, O_3_NAT).”

3. The study designed two experiments, O_3_MET and O_3_ALL, to distinguish the contributions of meteorology and biogenic emissions. In the random forest model, how did the authors ensure that fixing one set of variables does not introduce prediction bias due to autocorrelation among features?

Response:

This is a good point. Future climate simulations in CMIP6 follow the Shared Socioeconomic Pathways (SSPs) with various emission scenarios and land use changes. To be sure, the anthropogenic emissions of greenhouse gases (GHGs) are the dominant factor causing the future climate change (Figure 6.24 in IPCC AR6, 2021). The changes in precursor emissions can also influence future climate, but their impacts are much weaker than GHGs. Therefore, many studies identified the impacts of future meteorological parameter changes as climate-driven air pollution variations (e.g., Penrod et al., 2014; Cai et al., 2017; Hong et al., 2019). Therefore, we used the similar method in the ML model to distinguish the impacts of future biogenic emission and meteorological parameters under climate change on O_3 variations.

4. Under high-emission scenarios (e.g., SSP5-8.5), which specific changes in meteorological factors (such as radiation, humidity, and cloud cover) enhance the "ozone climate penalty" effect?

Response:

Among all the input predictors, relative humidity, air temperature, precipitation, and total cloud cover are the top most influential meteorological variables of the model construction for near-surface O_3 concentrations over Africa. The air temperature exhibits a positive influence on O_3 concentrations, while relative humidity and cloud cover are negatively associated with O_3 (Figure 3).

The future increases in air temperature, along with reductions in relative humidity and cloud cover are expected to enhance photochemical O_3 production, which has been illustrated in the manuscript. It will lead to substantial increases in O_3 levels under high-emission scenarios, indicting an O_3 climate penalty over Africa.

5. The results indicate that the impact of rising temperatures on mortality in Africa is far greater than that of ozone concentrations. Is this difference primarily due to variations in the sensitivity of exposure-response functions, or is it determined by the increased frequency of future extreme heat events?

Response:

The difference is primarily due to variations in the sensitivity of exposure-response functions. The mortality is more sensitive to the temperature change than the change in O_3 concentrations. Wang et al. (2022) also indicated that higher mortality risk was associated with elevated air temperatures rather than O_3 concentrations over North China Plain.

6. The study used multiple CMIP6 model datasets. Why were these particular models selected?

Response:

We used all available model datasets, rather than selected particular models. In this study, a variety of monthly meteorological variables, including air temperatures at 2 m, 850 hPa, and 500 hPa, wind fields at 850 and 500 hPa, precipitation rate, total cloud cover, relative humidity, sea level pressure, and incoming shortwave radiation at the surface, are selected to predict O_3 concentrations, since that these meteorological factors have shown substantial influences on O_3 concentrations. In total, 18 CMIP6 models (ACCESS-CM2, ACCESS-ESM1-5, CanESM5, CESM2-WACCM, CMCC-CM2-SR5, EC-Earth3-Veg, EC-Earth3, FGOALS-f3-L, FGOALS-g3, GFDL-ESM4, INM-CM5-0, IPSL-CM6A-LR, MIROC6, MPI-ESM1-2-HR, MPI-ESM1-2-LR, MRI-ESM2-0, Nor-ESM2-LM, and NorESM2-MM) have all of these monthly meteorological fields under four future scenarios (SSP1-2.6, SSP2-4.5, SSP3-7.0, SSP5-8.5). We have clarified the reason of selected CMIP6 models in Section 2.2.

7. What key input data did the GEOS-Chem model primarily provide during the machine learning training phase?

Response:

To train the ML model for predicting future biogenic isoprene emissions and O_3 concentrations, the input data provided by GEOS-Chem model include: (1) historical near-surface O_3 concentrations across Africa for 2000–2019 simulated by GEOS-Chem model; (2) MERRA-2 reanalysis data used to drive GEOS-Chem simulations, including temperatures at 2 m, 850 hPa, and 500 hPa, wind fields at 850 and 500 hPa, precipitation rate, total cloud cover, relative humidity, sea level pressure, and incoming shortwave radiation at the surface; and (3) the O_3 precursor emission inventory applied in GEOS-Chem model, including NO_x, CO, CH_4, and NMVOCs.

8. The authors adopt a "single-year verification" strategy, designating the period 2000–2009 and 2011–2019 as the training set, with 2010 serving as the sole test year. This approach may fail to fully assess the model's generalizability across different time periods and climate backgrounds. Ozone concentrations are significantly influenced by interannual meteorological fluctuations, and a single test year cannot cover diverse interannual variability scenarios, potentially underestimating the uncertainty of the model's projections for future interannual fluctuations. Please supplement the core rationale for selecting 2010 as the exclusive test year. Have more stringent temporal cross-validation methods been implemented (e.g., setting multiple independent test years, time segment splitting such as 2010–2014, or rolling window validation) to evaluate the model's robustness?

Response:

Thank you for pointing this important setting. The most significant feature under future climate change is the rising temperature. However, the present-day GEOS-Chem model simulations and ML model training may not capture the O_3 feature in a much warming climate in the future. To address this concern, we train the ML model using data in 19 years (2000–2009 and 2011–2019) when the air temperatures in Africa were relatively low and then validate performance of the ML model using testing data in 2010, the warmest year in Africa among the twenty years (see Fig. A below). With this data splitting method, the key information of warmer climate is included in the model training and testing. As mentioned above, the trained ML model has a good ability to predict future O_3 changes, even in the condition of extrapolation with higher temperature.

We have clarified the reason in manuscript as follows: "The 2010 records are used to validate the performance of RF model, as that land surface air temperature over Africa reached its peak during 2000–2019, which facilitates future projections in a warming climate. This data splitting strategy ensures the critical information related to rising temperature trends is captured in RF model projections."

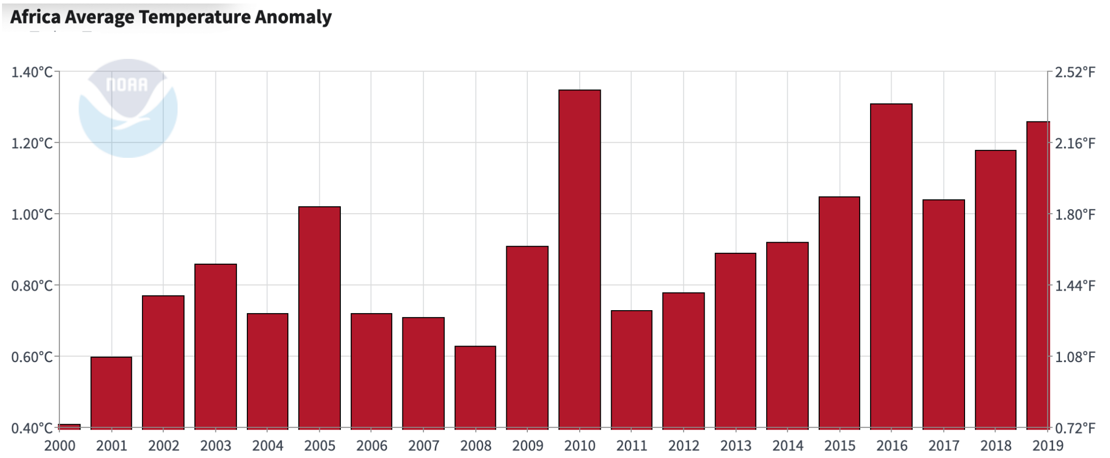

Figure A. Time series of the Africa annual mean surface air temperature over land during 2000–2019 from NOAA National Centers for Environmental Information.

9. SHAP analysis reveals that certain factors contribute most significantly to predicting biogenic isoprene emissions. Why are these factors particularly influential?

Response:

Variations in biogenic isoprene emissions are generally determined by meteorological parameters, with air temperature playing a leading role, as indicated by the feature contribution calculated in the SHAP analysis. A positive relationship exists between air temperature and isoprene emissions, which is consistent with previous studies (e.g., Singsaas and Sharkey, 1998). Moreover, wet weather conditions are negatively associated with isoprene emission rates in Africa, as increases in the frequency and intensity of precipitation influence plant functions (Strada et al., 2023).

Furthermore, isoprene emissions are strongly dependent on ecosystem type. Changes in human population and massive deforestation are expected to alter land cover, thereby affecting the functionality of terrestrial biosphere and exerting negative impacts on biogenic isoprene emissions (Rosenkranz et al., 2015). In contrast, isoprene emissions respond positively to increases in vegetation productivity, with broadleaf trees and shrubs exhibiting particularly high emission potentials (Guenther et al., 2006).

We have clarified in revised manuscript as follows: “The LC and NDVI are identified as the strongest positive drivers of isoprene emissions. This is consistent with the physics that biogenic isoprene is emitted mainly from terrestrial vegetation, and its emissions are directly influenced by LC types and the associated plant species (Guenther et al., 2006; Rosenkranz et al., 2015). Among all meteorological variables, air temperature plays a dominate role in regulating isoprene emissions, exhibiting a positive correlation, in line with prior work (e.g., Singsaas and Sharkey, 1998). The isoprene emission rates show opposite responses to wet weather conditions, as increases in the frequency and intensity of precipitation can influence plant functions (Strada et al., 2023)."

10. Which geographical region in Africa is predicted to experience the most significant increase in biogenic isoprene emissions, and why?

Response:

The projected biogenic isoprene emissions in Central Africa display the most significant increasing trend, with a maximum growth rate exceeding 1.0 $g/m^2/yr$ under four SSPs scenarios. According to the feature contributions derived from SHAP analysis, the land use (including land cover and normalized difference vegetation index) is the most dominant variable positively affecting isoprene emissions. The Central Africa has the vast tropical forests and woodlands, which serves as a major source of biogenic isoprene emissions on the continent. Besides, rising air temperature and dry conditions can further enhance isoprene release. In addition, the reductions in humidity over Central Africa contributes to the increase in isoprene emissions.

We have clarified in revised manuscript as follows:

"Central Africa has the vast tropical forests and woodlands, serving as the major source of isoprene emissions and exhibiting the largest increases in biogenic isoprene emissions across Africa, with a maximum growth rate exceeding 1.0 $g/m^2/yr$ under four scenarios." "Besides, rising air temperature and dry conditions can further enhance isoprene release. The enhancement of projected isoprene emissions induced by drought stress is particularly pronounced over Central and Southern Africa (Figure 6)."

11. Apart from temperature and isoprene, what other factors may influence future ozone concentrations under climate change? What are the main sources of uncertainty mentioned in the article, particularly regarding assumptions about land use and population density?

Response:

Future increases in air temperature, along with reductions in relative humidity and cloud cover will enhance photochemical O_3 production, leading to substantial increases in O_3 levels, with a maximum increase of 2.0 ppb.

There are some uncertainties in the future near-surface O_3 projections over Africa, which may arise from the input data, GEOS-chem model, CMIP6 climate models, and the ML model. The auxiliary data such as land use, topography and population density only for one specific year, which can also vary under future climate change. As a result, the constructed O_3 concentrations could be biased due to the lack of temporal variations of these variables.

12. To improve the accuracy of regional predictions, what enhancements does

the author suggest could be made in machine learning or chemical transport modeling in the future?

Response:

To improve the accuracy of regional projections, we can separately train the ML model for different regions of Africa based on region-specific input data, as O_3 variability differs distinctively depending on local emissions, meteorological conditions, land use, topography, and population density. By applying regionally customized ML model allows us to better capture the spatial and temporal characteristics of local-scale O_3 levels in each region, thereby enhancing the predictive performance. We have stated that “future work should train region-specific RF models to estimate near-surface O_3 concentrations and quantify variable contributions at regional scales”

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