

## 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  $0.5^\circ$  latitude  $\times$   $0.625^\circ$  longitude and 47 vertical layers from the surface up to 0.01 hPa, with boundary conditions from  $2^\circ$  latitude  $\times$   $2.5^\circ$  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  $\times$  1.75  $\times$  18 models  $\times$  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<sub>3</sub> chemistry. In addition, Africa does not have sufficient O<sub>3</sub> observation for the ML model training.

2. The article mentions that most regions in Africa are in a "NO<sub>x</sub>-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<sub>x</sub>-limited environments, where VOC are abundant, the increased isoprene emissions tend to reduce O<sub>3</sub> 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<sub>x</sub>-limited regions, increased biogenic isoprene emissions tend to reduce O<sub>3</sub> levels through oxidation processes (Pacifico et al., 2012; Squire et al., 2014), resulting in a slight decrease in O<sub>3</sub> levels by less than 0.5 ppb over Central and Western Africa in 2050 relative to 2020 (Figure 7b, O<sub>3</sub>\_NAT)."

3. The study designed two experiments, O<sub>3</sub>\_MET and O<sub>3</sub>\_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<sub>3</sub> 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<sub>3</sub> concentrations over Africa. The air temperature exhibits a positive influence on O<sub>3</sub> concentrations, while relative humidity and cloud cover are negatively associated with O<sub>3</sub> (Figure 3).

The future increases in air temperature, along with reductions in relative humidity and cloud cover are expected to enhance photochemical O<sub>3</sub> production, which has been illustrated in the manuscript. It will lead to substantial increases in O<sub>3</sub> levels under high-emission scenarios, indicating an O<sub>3</sub> 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<sub>3</sub> concentrations. Wang et al. (2022) also indicated that higher mortality risk was associated with elevated air temperatures rather than O<sub>3</sub> 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<sub>3</sub> concentrations, since that these meteorological factors have shown substantial influences on O<sub>3</sub> 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<sub>3</sub> concentrations, the input data provided by GEOS-Chem model include: (1) historical near-surface O<sub>3</sub> 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<sub>3</sub> precursor emission inventory applied in GEOS-Chem model, including NO<sub>x</sub>, CO, CH<sub>4</sub>, and NMVOCs.

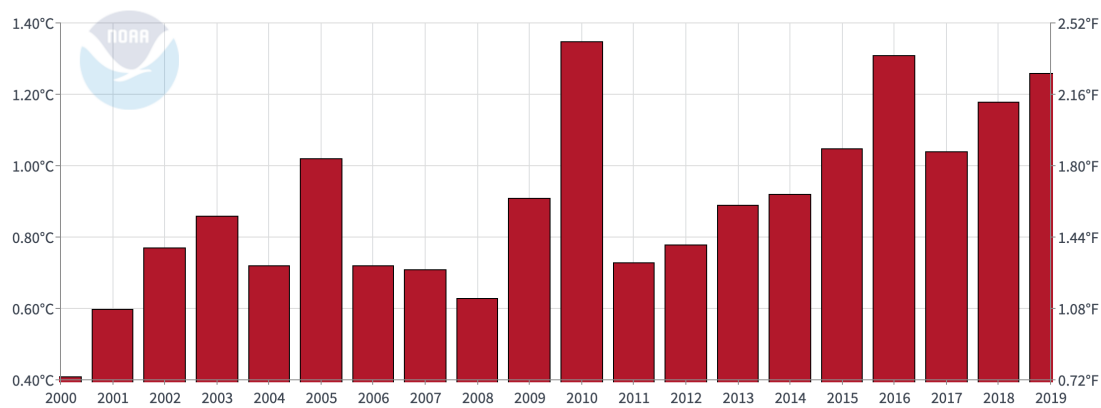
8. The authors adopt a "single-year verification" strategy, designating the period 2000–2009 and 2011–2019 as the training set, with 2010 serving as the sole test year. This approach may fail to fully assess the model's generalizability across different time periods and climate backgrounds. Ozone concentrations 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<sub>3</sub> 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<sub>3</sub> 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."

**Africa Average Temperature Anomaly**



**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., Singaas 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., Singaas 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<sup>2</sup>/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<sup>2</sup>/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<sub>3</sub> production, leading to substantial increases in O<sub>3</sub> levels, with a maximum increase of 2.0 ppb.

There are some uncertainties in the future near-surface O<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub> concentrations and quantify variable contributions at regional scales”

References:

Cai, W., Li, K., Liao, H., Wang, H., and Wu, L.: Weather conditions conducive to Beijing severe haze more frequent under climate change, *Nat. Clim. Change*, 7, 257–262, <https://doi.org/10.1038/nclimate3249>, 2017.

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmos. Chem. Phys.*, 6, 3181–3210, <https://doi.org/10.5194/acp-63181-2006>, 2006.

Hong, C., Zhang, Q., Zhang, Y., Davis, S. J., Tong, D., Zheng, Y., Liu, Z., Guan, D., He, K., and Schellnhuber, H. J.: Impacts of climate change on future air quality and human health in China, *P. Natl. Acad. Sci. USA.*, 116, 17193–17200, <https://doi.org/10.1073/pnas.1812881116>, 2019.

IPCC: Summary for Policymakers. In: *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change* [Masson-Delmotte, V., P. Zhai, A. Pirani, S. L. Connors, C. Péan, S. Berger, N. Caud, Y. Chen, L. Goldfarb, M. I. Gomis, M. Huang, K. Leitzell, E. Lonnoy, J.B.R. Matthews, T. K. Maycock, T. Waterfield, O. Yelekçi, R. Yu and B. Zhou (eds.)]. Cambridge University Press, 2021.

Pacifico, F., Folberth, G. A., Jones, C. D., Harrison, S. P., and Collins, W. J.: Sensitivity of biogenic isoprene emissions to past, present, and future environmental conditions and implications for atmospheric chemistry, *J. Geophys. Res.-Atmos.*, 117, D22302, <https://doi.org/10.1029/2012JD018276>, 2012.

Penrod, A., Zhang, Y., Wang, K., Wu, S., and Leung, L. R.: Impacts of future climate and emission changes on U.S. air quality, *Atmos. Environ.*, 89, 533–547, <https://doi.org/10.1016/j.atmosenv.2014.01.001>, 2014.

Rosenkranz, M., Pugh, T. A. M., Schnitzler, J. P., and Arneeth, A.: Effect of land-use change and management on biogenic volatile organic compound emissions—Selecting climate-smart cultivars, *Plant Cell Environ.*, 38, 1896–1912. <https://doi.org/10.1111/pce.12453>, 2015.

Singsaas, E. L., and Sharkey, T. D.: The regulation of isoprene emission responses to rapid leaf temperature fluctuations, *Plant Cell Environ.*, 21, 1181–1188, <https://doi.org/10.1046/j.1365-3040.1998.00380.x>, 1998.

Squire, O. J., Archibald, A. T., Abraham, N. L., Beerling, D. J., Hewitt, C. N., Lathi  re, J., Pike, R. C., Telford, P. J., and Pyle, J. A.: Influence of future climate and cropland expansion on isoprene emissions and tropospheric ozone, *Atmos. Chem. Phys.*, 14, 1011–1024, <https://doi.org/10.5194/acp-14-1011-2014>, 2014.

Strada, S., Fern  ndez-Mart  nez, M., Pe  uelas, J., Bauwens, M., Stavrakou, T., Verger, A., and Giorgi, F.: Disentangling temperature and water stress contributions to trends in isoprene emissions using satellite observations of formaldehyde, 2005–2016, *Atmos. Environ.*, 295, 119530, <https://doi.org/10.1016/j.atmosenv.2022.119530>, 2023.

Wang, P., Yang, Y., Li, H., Chen, L., Dang, R., Xue, D., Li, B., Tang, J., Leung, L. R., and Liao, H.: North China Plain as a hot spot of ozone pollution exacerbated by extreme high temperatures, *Atmos. Chem. Phys.*, 22, 4705–4719, <https://doi.org/10.5194/acp-22-4705-2022>, 2022.

## Responses to Reviewer #2

The manuscript's novelty is not yet convincingly established. Several of the stated innovations appear to be incremental combinations of existing data sources and standard analytical tools rather than clearly original methodological contributions. There are multiple global ozone projections under the CMIP6 scenarios employing Climate Chemistry models as well as data-driven methods, such as Li et al., 2026; Ni et al., 2026; Ma et al., 2026; Brown et al., 2022. The current manuscript did not provide enough innovations to construct ozone concentration datasets at either global or regional scale. Meanwhile, it looks like a repetitive work of their own work which applied similar ML techniques in studying global ozone changes under the same CMIP6 scenarios (Ni et al., 2026).

### Response:

We thank the reviewer for the insightful comment. We agree that several previous studies have projected or analyzed future O<sub>3</sub> variations using chemistry–climate models and data-driven approaches. Nevertheless, we believe our study provides important additional contributions to the community, particularly by offering a comprehensive projection of O<sub>3</sub> changes **across Africa under multiple scenarios** driven by climate change, along with a detailed decomposition of the associated **driving factors** and the associated **health burden** changes.

With a data-driven approach to estimate O<sub>3</sub> concentrations, Ma et al. (2026) selected air temperature as the sole predictor variable, and constructed a regression relationship between air temperature and O<sub>3</sub> using an L2 regularization function, mapping temperature to O<sub>3</sub> concentrations at a single target grid point. This method was then applied to two climate models to predict O<sub>3</sub>. This study provides a ML-based approach for estimating O<sub>3</sub>, however, uncertainties remain since the dominant meteorological factors driving O<sub>3</sub> variations often vary by region and season. Relying exclusively on temperature while neglecting other key variables such as solar radiation, relative humidity, and wind speed may oversimplify the complex photochemical and dynamical processes governing O<sub>3</sub> formation and transport.

By using three chemistry-climate model outputs (UKESM1-0-LL, GISS-E2-1-G, and MRI-ESM2-0) from the Aerosol-Chemistry Model Intercomparison Project (AerChemMIP), Brown et al. (2022) evaluated the climate-driven changes in near-surface O<sub>3</sub> concentrations over South America and Africa under the SSP3-7.0 scenario. This prior modelling study revealed the ozone-climate penalty over Africa, which supports several conclusions in our study with an integrated machine learning (ML) framework, although they found some disagreement of O<sub>3</sub> responses between models. To reduce uncertainties, our

study applied climate projections from 18 global climate models under four future scenarios (SSP1-2.6, SSP2-4.5, SSP3-7.0 and SSP5-8.5). To make one step further, individual contributions of changing meteorological conditions and changing natural isoprene emissions under climate change are separately evaluated using sensitivity ML-based simulations, rather than simplified linear regression. In addition, the corresponding future health risks in Africa related to increasing O<sub>3</sub> and rising temperature are also assessed.

We appreciate that the reviewer also noticed our recent work about O<sub>3</sub> **bias-correction** and future predictions over **China, the United States, and Europe** (Ni et al., *ES&T*, 2026). Although both these two studies used ML models, the methods are completely different. The CMIP6 multi-model and CESM2 model simulations exhibit systematic biases regarding the spatial distributions, long-term variations, and magnitudes of O<sub>3</sub> concentrations. To address these potential biases, Ni et al. (2026) integrated near-surface O<sub>3</sub> observations over China, the United States, and Europe from 2014–2020 based on a ML algorithm LightGBM to project O<sub>3</sub> and correct model outputs. The ML model in Ni et al. (2026) is to correct the model bias rather than to predict O<sub>3</sub> concentration, and the effectiveness of this method is largely constrained to regions with abundant observational data. Therefore, the ML-based bias-correction method can only be applied to China, the United States, and Europe. However, the monitoring stations are sparse and unevenly distributed in Africa, where the bias-correction method is not applicable.

In this study we project near-surface O<sub>3</sub> concentrations over Africa during 2020–2050 under four different scenarios (SSP1-2.6, SSP2-4.5, SSP3-7.0, and SSP5-8.5) based on a ML approach. The input data include GEOS-Chem simulations, climate projections from 18 global climate models, precursor emissions, and many other auxiliary datasets (land use and population). The novel method in our study lies in taking advantage of the chemical transport model in simulating O<sub>3</sub>, and incorporating future meteorological variables from ScenarioMIP multi-model ensemble to reduce the uncertainties inherent in climate model. In addition, the climate-driven changes in biogenic isoprene emissions and meteorological conditions are individually analyzed in this study, thereby providing a comprehensive understanding the impacts of climate change on O<sub>3</sub> variations. Moreover, the associated impacts on mortality from changing O<sub>3</sub> due to increasing isoprene emissions, changing meteorological conditions and rising temperature are separately evaluated, which was rarely involved in previous studies.

We have now clarified the novelty in the manuscript, as “Compared with previous studies that relied on simplified linear regression relationships or limited numbers of chemistry-climate model outputs, our study incorporates projections from 18 global climate models under four future scenarios to better constrain uncertainties in future climate-driven O<sub>3</sub> changes across Africa. In addition, the individual contributions of climate-driven changes in biogenic isoprene emissions and meteorological conditions are separately evaluated

through ML-based sensitivity simulations, providing a more comprehensive understanding of the impacts of climate change on O<sub>3</sub> variations. Furthermore, the associated mortality burdens attributable to changing O<sub>3</sub> concentrations due to increasing isoprene emissions and changing meteorological conditions, as well as mortality burdens linking to rising temperature are separately quantified, offering an integrated assessment of future air quality and health risks across Africa.”

## References:

Brown, F., Folberth, G. A., Sitch, S., Bauer, S., Bauters, M., Boeckx, P., Cheesman, A. W., Deushi, M., Dos Santos Vieira, I., Galy-Lacaux, C., Haywood, J., Keeble, J., Mercado, L. M., O'Connor, F. M., Oshima, N., Tsigaridis, K., and Verbeeck, H.: The ozone–climate penalty over South America and Africa by 2100, *Atmos. Chem. Phys.*, 22, 12331–12352, <https://doi.org/10.5194/acp-22-12331-2022>, 2022.

Ma, Y., Abraham, N. L., Versick, S., Ruhnke, R., Schneidereit, A., Niemeier, U., Back, F., Braesicke, P., and Nowack, P.: mloz: A highly efficient machine learning-based ozone parameterization for climate sensitivity simulations. *J. Adv. Model. Earth Syst.*, 18, e2025MS005459, <https://doi.org/10.1029/2025MS005459>, 2026.

Ni, Y., Yang Y., Wang, H., Wang, P., Li, K., Chen, L., Zhu, J., Li, B., and Liao, H.: Machine learning-based bias-corrected future projections of ozone concentrations from a chemistry-climate model, *Environ. Sci. Technol.*, 60, 3135–3147, <https://doi.org/10.1021/acs.est.5c11992>, 2026.