Response to Reviewer and Editor

Title: Influence of biogenic NO emissions from soil on Atmospheric chemistry over Africa: a regional modelling study

Author(s): Yao et al.
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The authors would like to sincerely thank the editor and Referee #3 for their constructive and insightful comments, which have helped us further improve the manuscript. In this revised version, we clarified the description and limitations of the ANN-based BioNO parameterization, including its applicability to different ecosystems, and expanded the discussion of its large impact on NO_2 concentrations compared to anthropogenic and biomass burning emissions. We now explicitly acknowledge uncertainties when extrapolating the ANN approach to continental scale, as well as the potential influence of missing sources such as lightning NOx and the underestimation of BB emissions. We also discussed the consistency of our findings with the recent TROPOMI-based inversions of Opacka et al. (2025), which suggest that natural NOx sources remain underestimated in current inventories.

In addition, we improved the description of model physics and deposition schemes (e.g., u* thresholds, Rg adjustments, ISORROPIA-II), defined acronyms at first use, enhanced figure readability, and clarified how the BioNO emissions dataset can be shared.

We are confident that these revisions address all concerns raised by Referee #3 and have strengthened the manuscript. We thank the reviewer and editor once again for their valuable feedback.

With our best regards,
On behalf of all the co-authors

Sincerely yours
Eric Martial Yao and Fabien Solmon

REFEREES' REPORTS:

REFEREE RC3

Comment 1: *1. Section 2.2, ANN description:*

Ref 1 asked for further information on this ANN, and the added explanation helps the text to some extent. I am a little nervous about a scheme that needs 27 weights, but agree with the authors that the Hudman scheme would not be a good choice either, and I think that the community should explore different approaches.

However, Ref 2 asked about the appropriateness of the Delon scheme for the land-use types used in this study. The authors respond that "we can assume that the model correctly reproduces the expected spatial distribution", based upon a statement that "these processes are well described in the literature". Are they? No literature is cited, and I see no link between the uncited literature and the 27 coefficients of the ANN. In fact, my default assumption would be that an ANN trained on one set of ecosystem would fail on a different set, and since it is effectively impossible to understand the complex set of equations I would like to see some evidence that the ANN is indeed robust over a wider range of conditions.

(Connected to this, are the values of the weights identical to those given in Delon et al. 2007? If not, explain. If so, why has there been no progress since then?)

Author's response: We thank Reviewer for this constructive comment, which helps clarify the assumptions and limitations associated with the use of the Delon et al. (2007) ANN scheme for biogenic NO emissions.

First, we would like to clarify that the implementation used in our study is based on the original ANN formulation described in Delon et al. (2007), but with revised weights introduced in Delon et al. (2008). The architecture and input variables of the ANN remained unchanged between the two versions; however, the weights were recalibrated in Delon et al. (2008) using updated observations and improved calibration techniques. This update aimed to better capture emissions dynamics, particularly in semi-arid tropical ecosystems, in order to better represent emission pulses after rewetting. We apologize for not making this distinction clearer in the manuscript. We have now corrected this in Section 2.2 and explicitly referred to both sources (Delon et al., 2007 and 2008).

Regarding the reviewer's question on whether there has been any evolution in the ANN approach since then, we note that the weights from Delon et al. (2008) are still commonly used in more recent applications (e.g. Delon et al., 2010, 2015), especially for simulations over Sahel and Delon et al. (2012) for simulations over West Africa in wet and dry savanna ecosystems. There has been no major recalibration of the ANN weights since 2008, because results in wet and dry savannas were considered as sufficiently robust. . However, this could indeed be a direction for future work.

Regarding the generalization of this parameterization across ecosystems: we fully agree with the reviewer's concern that an ANN trained and calibrated primarily on semi-arid and temperate environments may not accurately represent emissions from other land-cover types, notably humid tropical forests. The domain used in our study encompasses a wide range of ecosystems, from arid and semi-arid zones in the Sahel to dense evergreen forests. We acknowledge that the ANN performs better in regions characterized by strong dry-wet seasonality, where NO pulses are mainly driven by microbial activity following wetting events. Its applicability in forests, where high and stable soil moisture levels and thick canopy cover modify the controlling processes, is more limited. These limitations are already discussed in detail in Section 5 (BioNO fluxes). Following the reviewer's suggestion, we have now added a sentence in Section 2.2 to signal this early on, and we refer the reader to the further discussion later in the manuscript.

The biophysical mechanisms embedded in the ANN, such as the role of soil moisture, texture, pH, and nitrogen availability, are well established in the literature. For instance, moisture-pulse-driven NO emissions are a robust feature of dry and semi-arid regions (e.g., Delon et al., 2007, 2008, 2010, 2012, 2015) while persistent soil moisture and canopy interception in forests tend to reduce emissions through both biological inhibition and physical shielding (e.g., Davidson et al., 2000; Pilegaard, 2013). These key references are already cited in Section 5 to reinforce our statement that "these processes are well described in the literature".

Author's changes in manuscript: *Modified Line 213 to2137*

Author's changes in manuscript: *Modified Line 226 to 229*

Comment 2: *2. Other emissions (especially lightning)? The text discusses how BioNO contributes to large changes in O3 and NO2 bias, and Fig. 10 suggests that BioNO sometimes has a huge impact on the NO2. Is this realistic? Is there any evidence that these other emissions are underestimated? Are these suggestions consistent with other studies, e.g. the TROPOMI-based inversions of Opacka et al., 2025? In fact, I miss a proper discussion of the importance of uncertainties in all the emissions, including those of BB and lightning (which isn't mentioned, but might often correlate with BioNO pulsing).*

Author's response: We thank the reviewer for raising this important point. We acknowledge that Fig. 10 shows a relatively large impact of BioNO on modeled surface NO_2 concentrations, sometimes exceeding the contribution from the base-case anthropogenic and biomass burning (BB) emissions in certain regions and seasons. This outcome results from the combination of spatial variability in emission sources and the temporal dynamics of BioNO (interactive emissions), which can lead to strong pulses of NO emissions in semi-arid regions following soil wetting events.

This does not imply that total BioNO emissions systematically exceed anthropogenic or BB emissions at the continental scale. Rather, the large relative impact observed locally reflects the high sensitivity of NO_2 concentrations to episodic BioNO pulses.

We also recognize that the magnitude of the modeled NO_2 increase at the regional scale appears larger than typically reported in studies using standard inventories such as CAMS-GLOB-ANT or GEOS-Chem, which often include only minimal or empirical representations of BioNO. However, direct comparisons are difficult due to differences in model configurations and the more dynamic, process-based approach used here.

Furthermore, while our ANN-based BioNO parameterization shows good agreement with observed NO_2 levels at specific INDAAF stations, its extrapolation over the entire domain, especially in ecosystems poorly represented in the ANN training dataset (e.g., dense tropical forests), introduces significant uncertainty. We acknowledge this limitation and have revised the discussion to reflect that regional-scale impacts should be interpreted with caution, especially in areas far from the training conditions.

A further important point is that our simulations include BioNO but do not account for lightning NOx emissions, which are known to be a major contributor to the tropical NOx budget, particularly in the upper troposphere (Jaeglé et al., 2005). Their exclusion likely leads to an underestimation of total natural NOx, and may therefore artificially amplify the relative role of BioNO in our results.

Similarly, biomass burning (BB) emissions in Africa are subject to considerable uncertainty. Although we used the widely adopted GFED4 inventory, multiple studies (e.g., van Wees and van der Werf, 2019; Giglio et al., 2013) suggest that fire emissions are likely underestimated in this region. Since BB and BioNO emissions can be seasonally correlated, it is conceivable that part of the model-observation improvement obtained by adding BioNO could reflect a compensating effect for missing or underestimated BB emissions.

As mentioned, the inclusion of BioNO improves model agreement with surface NO_2 observations at specific locations and time periods, especially when soil and climate conditions fall within the ANN training domain. However, the extension to continental scale, especially in areas with different land surface or meteorological characteristics, remains uncertain. We fully agree with the reviewer that the regional extrapolation of the effect deserves further investigation in future work.

Finally, we note that recent inversion results from Opacka et al. (2025) lend support to the idea that natural NOx sources remain underestimated in bottom-up inventories over Africa. Using TROPOMI-based inversions of NO_2 and HCHO, they suggest that soil NO emissions are underestimated by ~26%, and that lightning NOx emissions may be underestimated by a factor of 4. These findings are consistent with our conclusion that current inventories likely miss substantial contributions from natural sources. Although our study does not include all such components (e.g., lightning NOx), it shows that the inclusion of process-based BioNO parameterizations alone can lead to improved agreement with observations in many regions.

We have revised the discussion accordingly to better address the reviewer's comment, and we thank them again for highlighting this important issue.

Author's changes in manuscript: *Modified Line 527 to 549*

Author's changes in manuscript: *Modified Line 698 to 701*

Author's changes in manuscript: *Modified Line 708 to 711*

Comment 3: *3. How are the results of this study relevant to the wider community? The ANN method used is so complex that few others can replicate the results, or would want to. The BioNO emissions themselves might be of interest to others though, and I think these should be made available through zenodo. (The data availability section says that results can be made available via email to the corresponding author, but that is a poor substitute for a long-term repository.)*

Author's response: Thanks for this important point. First, regarding the relevance of the study for the wider community: although the ANN-based method used to estimate soil NO emissions is indeed complex, the main objective of the study is not to promote the method itself, but rather to evaluate how an improved representation of soil NO emissions affects atmospheric composition over Africa. We believe these results are of broad interest for the modeling and air quality community, particularly in regions where bottom-up inventories are sparse or highly uncertain. Our study highlights key differences in spatial patterns, seasonality, and impact on tropospheric chemistry that can inform future model developments and field measurement campaigns.

Concerning data availability, we agree that the BioNO emissions may be of interest to others. To be considered as a community dataset, the fluxes and inventory would need to be more robust, notably through additional validation using multiple models, reanalysis data, and longer time series. The parametrization used to estimate soil NO emissions is publicly available. Model outputs can of course be provided upon request.

Other comment 1: *L54 - mangled sentence after some words removed.*

Author's response: The sentence has been revised to restore grammatical clarity and improve readability in the revised version of the manuscript.

Author's changes in manuscript: *Modified Line 53 to 54*

Other comment 2: *L95 - I would skip the word "will" here*

Author's response: We agree with the reviewer and have removed "will" for a more concise formulation.

Author's changes in manuscript: *Modified Line 78 to 82*

Other comment 3: *L105 - change "in regards to" to "with regard to"*

Author's response: The expression “in regards to” has been replaced with the correct form “with regard to.

Author's changes in manuscript: *Modified Line 91*

Other comment 4: *L101 on: As requested by Ref 2, the authors have added a section on the CBM-Z, which builds upon CBM-IV. This mechanism is rather old now, and as noted by Ref 2 the scheme may oversimplify some interactions. Further comments comparing to more recent versions would be useful, e.g. to CBM-6 (Cai et al., 2021, already cited) or CBM-7 (Yarwood et al, 2024). Would any of the updates affect conclusions about the impact of BioNO emissions in the troposphere?*

Author's response: Recent developments such as CBM-6 (Cao et al., 2021) and CBM-7 (Yarwood and Tuite, 2024) have introduced improved VOC speciation, more explicit treatment of isoprene oxidation pathways, and updated peroxy radical chemistry. These enhancements can influence the formation of ozone and secondary nitrogen species under certain regimes. However, given the focus of our study on the large-scale impacts of BioNO emissions on tropospheric NOx and O_3 levels, the added complexity of these newer mechanisms would likely yield only marginal changes in the overall trends and spatial patterns reported here. Moreover, the uncertainties in precursor emissions (especially soil NO and VOCs over Africa) remain a dominant source of variability, limiting the relative benefit of switching to a more detailed mechanism. Nonetheless, future work could explore the sensitivity of BioNO impacts to alternative chemical schemes in high-resolution setups.

Author's changes in manuscript: *Modified Line 118 to 125*

Other comment 5: *L119. Although the original referees did not comment on this, I wonder about gas-particle partitioning for coarse particles. The manuscript only notes that ISORROPIA-II is "mostly relevant for fine particle":*

Author's response: We thank the reviewer for the observation. We agree that ISORROPIA-II is a general-purpose thermodynamic equilibrium model that applies to both fine and coarse aerosol particles. To avoid any ambiguity, we have removed the reference to fine particles and now refer more generally to the gas-aerosol partitioning and speciation of inorganic compounds calculated with ISORROPIA-II.

Author's changes in manuscript: *Modified Line 126 to 128*

Other comment 6: *L126(a). Although the order of words has been changed in this section, I didn't understand the context - did the original model get very wrong O3 over the ocean? (If so, it must have had very low resistances.). Also, the modification is not really explained ("values are adjusted by lowering them" - this is not a scientific explanation). What was really done?*

Author's response: Indeed, the default resistance value (2000 s m^{-1}) led to deposition velocities that were too low, resulting in an overestimation of surface ozone concentrations over oceanic regions. In our study, the default Rg value for ozone over water, set at 2000 s m^{-1} in Zhang et al. (2003), was reduced to 1000 s m^{-1}. This adjustment was made to align the simulated dry deposition velocities with those reported in the literature (e.g., Charusombat et al., 2010; Wu et al., 2011; Zhang et al., 2002), which indicate higher deposition velocities over oceanic surfaces than those predicted by the default values. This modification improves the accuracy of simulated surface ozone concentrations, For clarity, we have revised the text at Line 108 to include these specific details.

Author's changes in manuscript: *Modified Line 134 to 139*

Other comment 7: *L129(b) The paper cites the Padro et al. criteria that the Richardson number should be maintained below 0.21 for stable conditions. In order to do this the authors set a lower threshold of 0.4 /s for u*. Do they mean that they excluded data which did not satisfy that criteria, or did they set a minimum value of u*. To do the latter would be incorrect I think. Also, a lower threshold of u* = 0.4 m/s is set, but this seems excessive. Figure 1 of*

Flechard et al. (2011) suggests that such a threshold would exclude quite a high percentage of European forests!

Author's response: To ensure the Richardson number (Rib) < 0.21 for realistic dry deposition velocities (Vd) per Padro et al. (1991), we recalculated u* in the RegCM model by imposing minimum thresholds of 0.4 m/s in forests and 0.1 m/s in savannas, rather than excluding data. These thresholds, derived from statistical analysis of simulations, showing Rib > 0.21 when u* falls below these values, aligned with studies such as Adon et al. (2013), who used 0.2 m/s for forests and 0.1 m/s for savannas in their offline simulation. The higher forest threshold partly reflects the lower modeled u* values over forested regions in our simulations, which may arise from limitations in the model's representation of subgrid turbulence. Values of u* were recalculated from the interactive u* according to these thresholds.

We acknowledge that the 0.4 m/s threshold may appear high compared to studies in temperate regions, such as Flechard et al. (2011), which found that such thresholds exclude a large fraction of forest data in Europe. However, tropical African forests experience generally weaker mean winds and higher atmospheric stability, which justifies a higher u* threshold in this context.

In addition, we note that at this spatial resolution and with parameterized convection, the modeled wind speed and u* are uncertain, especially under weak synoptic conditions. The model does not resolve intermittent subgrid thermal gusts, which can lead to an underestimation of the effective u*. Therefore, the forest-specific u* threshold value used in our dry deposition scheme may compensate in part for this limitation.

Therefore, the applied u* thresholds are a pragmatic correction applied exclusively within the dry deposition scheme to avoid unrealistically high aerodynamic resistances and resulting biases in deposition velocities. This adjustment does not modify u* for other processes or model components.

We have revised the manuscript text to clarify these points.

Author's changes in manuscript: *Modified Line 140 to 153*

Other comment 8: *L137. Here the authors say that they "substantially improve" the wet deposition scheme, but this "improvement" is undocumented and unproven.*

Author's response: Compared to Shalaby et al. (2012) and Ciarlo et al. (2021), our RegCM interface enhances wet deposition by directly using cloud-to-rainwater production and precipitation rates from Nogherotto et al. (2016) stratiform and Tiedtke (1989) convective schemes, better capturing the interaction of chemistry and convective precipitation processes in African tropical rainy seasons. This yields wet deposition estimates closer to literature values (e.g., Delon et al., 2010; Ossohou et al., 2021). The text is revised for clarity.

Author's changes in manuscript: *Modified Line 156 to 160*

Other comment 9: *L183-184. Be more precise. What is meant by "surface" and "deep" here? Over what depth is sand percentage and pH expected. Which height is wind-speed?*

Author's response: We thank the reviewer for this important point. We have revised the manuscript to clarify the meaning and units of the input variables (x1 to x7) used in the NO flux parameterization. Specifically:

- Surface soil temperature refers to the soil temperature measured at 0–5 cm depth
- Deep soil temperature corresponds to the temperature at 20–30 cm depth
- Sand percentage and pH refer to soil properties integrated over the topsoil layer (typically the upper 30 cm)
- Wind speed is taken at 10 m from the soil.

These clarifications have been added to ensure the reproducibility of the parameterization and to avoid ambiguity.

Author's changes in manuscript: *Modified Line 204 to 207*

Other comment 10: *L212. The acronym INDAAF is used before it is explained in the main text. (The abstract should summarize the main text, and the main text should not rely on definitions being in the abstract.)*

Author's response: We agree that acronyms should be defined at their first occurrence in the main text to ensure clarity and readability. Accordingly, we have removed the full definition of INDAAF from the abstract and now introduce and define the acronym at its first occurrence in the main text.

Other comment 11: *L389. The Simpson and Darras emissions were calculated under CAMS, but EMEP MSC-W isn't part of CAMS. (BTW, the S&D citation has "no" and should be "NO")*

Author's response: The emissions presented in Simpson and Darras (2021) were indeed calculated in the context of the CAMS project, but using the EMEP MSC-W model, which itself is not part of the CAMS system. We have clarified this point and corrected the citation typo ("no" to "NO") in the revised manuscript.

Author's changes in manuscript: *Modified Line 417 to 420*

Other comment 12: *The acronym ERA5 is used twice (p4,6) before it is explained on p10. Explain on 1st mention.*

Author's response: We agree with the reviewer that acronyms should be defined at first mention. We have therefore introduced the definition of ERA5 directly in the first occurrence, and removed the redundant definition later in the text..

Author's changes in manuscript: *Modified Line 100 to 102*

Author's changes in manuscript: *Modified Line 279 to 280*

Other comment 13: *The acronym CAMS is spelled out on p5, but then again on p17. No need for double explanations.*

Author's response: We thank the reviewer for this remark. The acronym CAMS (Copernicus Atmosphere Monitoring Service) is now defined at its first occurrence, and all redundant definitions in subsequent sections have been removed for clarity and consistency.

Other comment 14: *Fig. 8. It is difficult to read the text above the sub-figures. Enlarge this text.*

Author's response: We have increased the font size of the titles above the sub-figures in Figure 8 to improve readability. The updated version is now clearer and easier to read.