<u>**Response to Comment RC1**</u>

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

Author(s): Yao et al.

MS No.: acp-2024-3179

General Comment : *This work evaluates the impact of soil nitric oxide emissions (BioNO) on regional air quality over Africa using the RegCM5 regional climate model. The authors applied a revised chemistry scheme within the model, and also used updated BioNO emissions developed from an artificial neural network (ANN). The authors find that by incorporating BioNO, evaluation of model chemical output against observations is improved across various time scales and for multiple chemical species. This manuscript contains numerous grammatical errors and typos, particularly related to poor sentence structure, incorrect verb tense, and missing articles (e.g. "a" or "the"), and there are an abundance of acronyms which are not defined throughout the manuscript. Additionally, there are many instances of phrasing and terminology that deviate from the conventional language typically used in atmospheric science / ACP publications. While not necessarily incorrect, using unconventional language can make the text feel less polished and can be distracting. These grammatical errors and text issues unfortunately detract from the manuscript quality, making it difficult to assess the overall quality of this work objectively. I have suggested some edits in the line by line comments, however I encourage the authors to more thoroughly go through the entire manuscript to correct remaining grammatical errors and to carefully revise the manuscript to improve clarity and readability for an ACP audience before resubmitting. I do find the model evaluation of surface NO_2, O_3 and HNO_3 concentrations against observations to be worthwhile.*

The authors would like to sincerely thank the reviewer for his/her constructive feedback and valuable suggestions to improve the clarity and quality of the manuscript. We have carefully revised the entire text to address grammatical errors, correct typos, and improve sentence structures. We also ensured the consistent use of articles and proper verb tenses throughout the manuscript. All acronyms have now been clearly defined upon their first mention. In addition, we revised phrasing and terminology to align with the conventional language typically used in atmospheric science and ACP publications, following the reviewer's guidance. These modifications have considerably improved the clarity and readability of the manuscript. Furthermore, the entire manuscript has been thoroughly reviewed by a native English speaker to ensure linguistic accuracy and fluency. We greatly appreciate the reviewer's positive remark regarding the model evaluation of surface NO_2, O_3, and HNO_3 concentrations against observations.

Specific Comment 1 : *The word "significant" is used numerous times in this manuscript, when the sentiment is being conveyed that something is substantial, or noteworthy. I encourage the authors to reduce the use of "significant" and "significantly" in such cases, as these words should be reserved for the context of statistical significance, as opposed to something that is notable. Consider using words such as: notable, noteworthy, considerable, or substantial.*

We thank the reviewer for this valuable suggestion. We have carefully reviewed the manuscript and replaced all instances where "significant" and "significantly" were used in a non-statistical context with more appropriate terms such as "notable," "considerable," or "substantial."

Specific Comment 2 : *There are many website links and DOIs that are included within the body of the text, which need to be included in the reference section with only an appropriate citation within the body of the text. Refer to other ACP papers for an example of appropriate citations.*

In accordance with ACP formatting guidelines, we have removed all direct links and DOIs from the main text and properly cited them in the reference section. In the body of the text, we now refer to these sources using standard in-text citations.

Specific Comment 3: *Section 2.2 describing the BioNO parameterization requires attention. In equation 2, how are each of the weights determined? What do the variables S1, S2, and S3 represent? Why does each of S1, S2 and S3 contain a summation cross differing values, e.g. 1 to 7, 8 to 23, and then 17 to 23? What do the subscripts on the weights represent? This is not explained, causing the various equations feel arbitrary. Additionally, why are hyperbolic tangents used to represent this process in equation 1? This is not a common approach when modeling soil NO emissions. Why not use a more common, and newer, approach, e.g. Hudman et al., (2012), which is cited within this manuscript? There needs to be a clearer description of this parameterization and justification for decisions that were made, as the soil emissions and subsequent impacts appear to be the focal point of this manuscript.*

We thank the reviewer for these insightful questions regarding the BioNO parameterization. We have updated Section 2.2 to incorporate these following explanations and ensure clarity regarding the parameterization process (based on the description given in Delon et al., 2007):

- The weights (wi) in the Artificial Neural Network (ANN) equations are determined through a supervised training process. Initially, these weights are assigned random small values to ensure that no input variable dominates the model from the start. The network is trained using a backpropagation algorithm, which iteratively adjusts these weights to minimize the error between the predicted NO fluxes and observed values. In each iteration, the network calculates the prediction error, propagates this error backward through the network, and updates the weights accordingly. This process continues until the error is minimized, ensuring that the final weights reflect the true influence of each input variable on NO emissions.
- The tanh (hyperbolic tangent) function is employed as the activation function in the network. It introduces non-linearity, which is essential for capturing the complex interactions between environmental variables and NO emissions. The tanh function also normalizes intermediate outputs to a range between -1 and 1, stabilizing the learning process and preventing extreme values from skewing the results.
- The sub-equations (S1, S2, S3) structure the network into separate layers, each designed to capture different aspects of the relationships between environmental variables and NO emissions. Although they all use the same input variables (x1 to x7, representing factors like WFPS, soil temperature, pH, etc.), they apply different weights in each sub-equation. This design allows the network to explore multiple combinations of the inputs and better capture non-linear dependencies. Each sub-equation acts as a filter, emphasizing specific patterns or interactions among the variables. The outputs of these sub-equations are then combined using the tanh function to produce the final normalized NO flux.
- The use of this ANN approach, instead of models like Hudman et al. (2012) approach, which was developed at a global scale, was chosen for its ability to model complex and non-linear relationships directly from empirical data. This is particularly valuable in regions like Africa, where in situ measurements of BioNO are limited. The ANN allows the model to adapt to diverse environmental conditions and better generalize across the domain.

Revised text (Lines 186-198): *" The weights wi … the normalized NO flux."*

Specific Comment 4: *While section 2 describes some general details about the RegCM5 model, no details are provided about the specific model domain for this study. Is the model being run for all of Africa, or a subset? Or is the model run globally, and only a subset is being shown to focus on Africa? A clearer description is needed. This could be added to section 2.3.*

We have revised Section 2.3 to include a detailed description of the model domain used in this study. We clarified that the model is not run globally but focuses solely on a specific domain of Africa.

Revised text (Lines 208-213): *"The model domain extends from 19.35°S to 35.48°N ... the INDAAF measurement sites."*

Line-by-Line Technical Correction – Line 4 (Abstract): *I would change "atmospheric NO_2 emissions" to "atmospheric NO_2 concentrations". Also, please define NO_2, e.g. nitrogen dioxide (NO_2). Please define all acronyms at their first instance before using an acronym.*

We have replaced "NO_2 emissions" with "NO_2 concentrations". Additionally, we have defined NO_2 as "nitrogen dioxide (NO_2)" upon its first mention and reviewed the manuscript to ensure all acronyms are properly introduced when first used.

Line-by-Line Technical Correction – Line 6 (Abstract) : *Please define INDAAF acronym.*

As mentioned in our response to the previous comment regarding acronyms, we have now defined INDAAF (International Network to study Deposition and Atmospheric chemistry in Africa) upon its first mention in the abstract.

Line-by-Line Technical Correction – Line 9 (Abstract): *The sentence starting with "Large differences are however…" is a run-on sentence, and the wording is confusing. Please rephrase. This sentence also appears to contradict the claim in the previous sentence that model accuracy has been improved.*

We thank the reviewer for this insightful comment. We acknowledge that the original sentence was overly complex and could lead to misinterpretation. We wanted to say while the integration of BioNO emissions improves the overall model performance particularly for NO_2, HNO_3, and O_3 seasonal cycles, it does not eliminate all discrepancies. To clarify this point and avoid confusion, we have rephrased the sentence to distinguish between the general improvements in model accuracy and the specific biases that still remain, especially regarding surface ozone concentrations. This also resolves the run-on sentence issue.

Revised text (Lines 10–12, Abstract): *"Despite these improvements, ... model used for comparison."*

Line-by-Line Technical Correction – Line 16 & 17: *"... a significant source of gaseous and particulate emissions, affecting the regional…."*
"…, there are also a significant…"

We have revised the sentences to avoid redundancy and improve the flow. The first occurrence of "significant" was replaced with "major", and the second with "substantial" to maintain the intended meaning while enhancing readability.

Revised text (Lines 17–20): *"Tropical Africa is a major ... the tropics (e.g., Aghedo et al., 2007)."*

Line-by-Line Technical Correction – Line 22: *"... (Referred to as BioNO) ..."*

We have corrected "referred as" to "referred to as" to adhere to proper English usage.

Revised text (Lines 23-24): *"Soil NO (referred to as BioNO) ... the atmosphere (Ludwig et al., 2001)."*

Line-by-Line Technical Correction – Line 23: *"... (Ludwig et al., 2001) and above-canopy emissions estimates range from 4.7 – 26.7 Tg N yr⁻¹..."*

To improve the clarity and structure of the sentence, we have reformulated it slightly to ensure smooth reading and clear attribution of the reference.

Revised text (Lines 24-26): *"... (Ludwig et al., 2001), and above-canopy ... (Davidson and Kingerlee, 1997; Ganzeveld et al., 2002; Hudman et al., 2012; Jaeglé et al., 2005; Müller, 1992; Steinkamp and Lawrence, 2011; Vinken et al., 2014; Yan et al., 2005; Yienger and Levy, 1995)."*

Line-by-Line Technical Correction – Lines 27-32: *I would reorganize these sentences, to first list the large variety of environmental factors that influence BioNO, and then following with the description of precipitation and soil moisture. While soil emissions are heavily influenced by precipitation and soil moisture, I don't find it accurate to lead with the idea that emissions "primarily" depend on soil moisture. This is just one of numerous important factors.*

We thank the reviewer for this valuable comment. We have reorganized the sentences as suggested, starting with a comprehensive list of environmental and physical factors that influence BioNO emissions, followed by a focused discussion on soil moisture and precipitation. Regarding the emphasis on soil moisture, we agree that it is one of several important factors influencing soil NO emissions. However, in tropical regions, such as those studied here, soil moisture plays a dominant role in controlling seasonal variations in BioNO

fluxes, primarily due to the well-documented pulse effect following the onset of rainfall (Johansson et al., 1988; Yienger and Levy, 1995; Meixner and Yang, 2004), essentially because soil temperature variation is low. To reflect this, we have nuanced the statement to highlight soil moisture as a key driver in tropical soils while acknowledging the influence of other environmental factors.

Revised text (Lines 28-37): *"BioNO emissions are influenced ... sharp increases in NO emissions."*

Line-by-Line Technical Correction – Line 33: *Remove "however"*

We have removed "however" from the sentence to improve the clarity and flow of the text.

Revised text (Line 38): "In tropical Africa, estimating BioNO emissions is challenging due to…"

Line-by-Line Technical Correction – Line 40: *"… used a neural network-based…"*

We have added the article "a" before "neural network-based" to maintain correct grammatical structure.

Revised text (Lines 45-46): *"... Delon et al. (2008) used a neural network-based parameterization coupled…"*

Line-by-Line Technical Correction – Lines 41–42: *Does "lower equatorial Africa's troposphere" refer to equatorial Africa's lower troposphere, or does this refer to Africa's troposphere at lower latitudes? The wording is confusing.*

"… troposphere on 6 August 2006 during the AMMA…" Please define the AMMA acronym."

We thank the reviewer for this helpful comment. Our intention was to refer to the lower troposphere over Equatorial Africa, focusing on the atmospheric layer closest to the surface where BioNO emissions influence NOx and O$_3$ production. We have revised the sentence to clarify this and avoid confusion. This revised phrasing aligns with the description provided in Delon et al. (2008). We have also defined the acronym AMMA upon its first mention as "African Monsoon Multidisciplinary Analysis" to ensure clarity.

Revised text (Lines 47–48): *"... production in the lower troposphere over Equatorial Africa for a specific day (6 August 2006) during the AMMA (African Monsoon Multidisciplinary Analysis) campaign..."*

Line-by-Line Technical Correction – Line 48: *"... resulting in a 10% decrease…"*

We have corrected "an 10%" to "a 10%" to ensure proper grammar.

Revised text (Line 53): *"... the global troposphere's oxidizing capacity and resulting in a 10% decrease ..."*

Line-by-Line Technical Correction – Lines 49–50: *"... regional climate systems to study the impact of present and future…."*

We have revised the sentence to clearly emphasize that the focus is on studying the impact of present and future climate change and variability on BioNO emissions.

Revised text (Lines 54–55): *"Integrating interactive BioNO emissions into models of regional climate systems study the impact of present and future climate change and variability, ..."*

Line-by-Line Technical Correction – Line 52: *"...in the chemical environment…"*

To improve clarity, we have modified "chemical environment" to "the chemical environment" to specify the context and avoid ambiguity.

Revised text (Line 56): *"... possible future emission trends, potential changes in the chemical environment ..."*

Line-by-Line Technical Correction – Line 58: *"I recommend starting a new paragraph with the sentence 'One goal of the present study…'"*

We have started a new paragraph with the sentence "One goal of the present study…" to improve the structure and readability of the text.

Revised text (Line 64): *"One goal of the present study ..."*

Line-by-Line Technical Correction – Line 59: *"Please define ICTP RegCM5"*

We have defined the acronym ICTP RegCM5 upon its first mention as "International Centre for Theoretical Physics Regional Climate Model version 5 (ICTP RegCM5)" to ensure clarity.

Revised text (Lines 64-65): *"... a system based on the International Centre for Theoretical Physics Regional Climate Model version 5 (ICTP RegCM5) (Giorgi et al., 2023) ..."*

Line-by-Line Technical Correction – Lines 70–73: *"This sentence referring to the shared scientific goals should not be in the introduction of the manuscript. This would be more appropriate to include in an acknowledgements section. Additionally, DOIs should not be included within the body of text in a manuscript. They should be included in the references section with a relevant citation."*

We thank the reviewer for this comment. We had already included detailed acknowledgements for both the INSA and INDAAF projects in the Acknowledgements section. To avoid redundancy, we have removed the sentence from the introduction and maintained the information in the Acknowledgements section. Additionally, we have ensured that all DOIs are correctly placed in the References section.

Line-by-Line Technical Correction – Line 74: *"... Sections 2 and 3 will provide", to be consistent with verb tense later on in the paragraph.*

To ensure consistency in verb tense throughout the paragraph, we have changed "provide" to "will provide".

Revised text (Line 77): *"The study is structured as follows: Sections 2 and 3 will provide a description ..."*

Line-by-Line Technical Correction – Line 80: *"Is the model referred to as RegCM5, or RegCM5-CHEM? I see references to both, without a clear description. I recommend choosing one for consistency."*

The model used in this study is RegCM5, which inherently includes the atmospheric chemistry module. To maintain clarity and consistency throughout the manuscript, we have specified the term "RegCM5" as the full model with online chemistry activated, rather than using the term "RegCM5-CHEM". We have clarified this in Section 2.1.

Revised text (Line 83): *"The Regional Climate-chemistry Model RegCM5"*

Line-by-Line Technical Correction – Lines 82 and 94 :

Line 82: "Please define MOLOCH"
Line 94: "Please define OI_WK"

We have now defined both MOLOCH and OI_WK upon their first mention in the manuscript to improve clarity.

- MOLOCH has been defined as *"MOdello LOCale in H coordinate (MOLOCH), a non-hydrostatic dynamical core developed by CNR-ISAC (Davolio et al., 2020)"*.
- OI_WK has been defined as *"Optimal Interpolation Weekly (OI_WK) sea surface temperature dataset"*.

Revised text (Line 85): *"... has been the inclusion of the MOdello LOCale in H coordinate (MOLOCH) ..."*

Revised text (Lines 97-98): *"... the Optimal Interpolated Weekly (OI_WK) dataset."*

Line-by-Line Technical Correction – Line 101: *"I believe this is supposed to say 'fine particle', not 'fine particles'."*

We agree with the suggestion and have corrected "fine particles" to "fine particle" to ensure grammatical accuracy and consistency.

Revised text (Line 120): *"... mostly relevant for fine particle heterogeneous processes."*

Line-by-Line Technical Correction – Lines 107–109: *"This sentence contains numerous grammatical errors."*

We have revised the sentence to correct grammatical errors and improve clarity.

Revised text (Lines 126–128): *"Default ground resistance (Rg) values ... over the ocean."*

Line-by-Line Technical Correction – Line 122: *"Use 'degree' instead of 'deg'."*

We have replaced "deg" with "degree" to maintain consistency with formal writing standards.

Revised text (Line 141): *"In this study, the monthly, 0.1 degree resolution emission…"*

Line-by-Line Technical Correction – Lines 142 & 144: *"This sentence appears to be repeated, the same as line 139." & "It appears that a new sentence begins in the middle of a different sentence here"*

We thank the reviewer for pointing out this redundancy. We have removed the repeated portion from line 142 to avoid duplication, and a new sentence begins correctly.

Revised text (Lines 158–163): *"The main advantage in using this ANN ... oxygen supply (Skopp et al., 1990)."*

Line-by-Line Technical Correction – Line 165: *"Please define WFPS."*

We have defined WFPS upon its first mention as "Water-Filled Pore Space (WFPS)" to ensure clarity.

Revised text (Lines 182-183): *"... (surface Water-Filled Pore Space (WFPS), surface soil temperature, ...)."*

Line-by-Line Technical Correction – Line 175: *"I suggest 'The model has a spatial resolution of 30 km x 30 km, with 35 vertical…'"*

We thank the reviewer for this suggestion. We have revised the sentence to improve clarity

Revised text (Line 206): *"The model has a spatial resolution of 30 km × 30 km, with 35 vertical levels ..."*

Line-by-Line Technical Correction – Line 179: *"Add subscripts for O3, NO2 and HNO3."*

We have added subscripts to O_3, NO_2, and HNO_3 to follow standard chemical notation.

Revised text (Lins 215-216): *"O_3, NO_2, and HNO_3 concentrations are measured…"*

Line-by-Line Technical Correction – Line 196: *"This line says 'used here as reference'. What is this referring to? Should there be a reference to a figure perhaps? Also, it is not clear what 'duplicates means' is referring to."*

We thank the reviewer for highlighting this ambiguity. We have clarified the phrase "used here as reference" by specifying that it refers to the baseline dataset used for model evaluation. We have also revised "duplicates' means" to "average of these duplicate samples"

for clarity. The duplicate samples refer to the two passive samplers exposed at each INDAAF site to ensure reproducibility, as described in Ossohou et al. (2023), with monthly concentrations calculated from the arithmetic mean of these duplicates.

Revised text (Line 130-235): *"... provides a comprehensive overview and is used as a reference dataset for model ... the average of these duplicate samples (Ossohou et al., 2023)."*

Line-by-Line Technical Correction – Line 198: *"The sentence states that LAERO participated bi-annually, and then later specifies twice yearly, which is redundant."*

We have revised the sentence to remove the repetition and improve clarity.

Revised text (Lines 236-238): *"LAERO has participated in the bi-annualy ... for trace compounds."*

Line-by-Line Technical Correction – Line 200: *"References to other work/studies should not be done by including a link within the body of the text. Same for DOIs."*

We have removed all direct links from the body of the text and included them in the references section following the journal's guidelines.

Line-by-Line Technical Correction – Line 210: *"ERA5 does not stand for 'European Environment Agency version 5'."*

We thank the reviewer for this comment. We have corrected the definition of ERA5, which stands for "ECMWF Reanalysis v5" (produced by the European Centre for Medium-Range Weather Forecasts), and updated the text accordingly.

Revised text (Lines 247-249): *"... we use data derived from the ECMWF (European Centre for Medium-Range Weather Forecasts) Reanalysis version 5 (ERA5; Hersbach et al., 2020)."*

Line-by-Line Technical Correction – Line 241: *"Remove 'somehow'."*

We have removed "somehow" to improve clarity and maintain a more objective tone.

Revised text (Line 281): *"... although the SHL amplitude is underestimated (Figure 2)."*

Figure Comment: *"I recommend removing the underlying emissions data from this figure, as those results do not get discussed at any point. I recommend modifying this figure to be a simpler introduction to the model domain, with a rectangle denoting the extent of the model domain, as well as the INDAAF points, to show where measurements were taken. Otherwise, there is no introduction to the model domain."*

We thank the reviewer for this insightful comment. We understand the importance of providing a clear introduction to the model domain and have addressed this concern in the following ways:

1. **Clarifying the Model Domain in Figure 1:**
   The current Figure 1 already represents the full extent of the model domain used for the simulations, without any cropping during post-processing. To make this clearer, we have updated the figure legend to explicitly state that the displayed area corresponds to the entire model domain. Details about study are already provided in Section 2.3. The INDAAF measurement sites are also shown to provide context for the locations of observational data used in the study.

2. **Retention of Emissions Data in the Figure 1:**
   While we understand the suggestion to simplify the figure, we believe that retaining the emissions data is important for contextualizing the spatial distribution of anthropogenic and biomass burning NO emissions within the model domain. These emissions are directly relevant to the discussions in the manuscript, particularly when interpreting model results over regions where biomass burning or anthropogenic activities predominate. For instance, statements such as *"In the biomass burning regions, these differences are less visible"* and *"Both the model and the satellite data show high concentrations of NO_2 in areas such as the Sahel and forest regions, where biomass burning plays a significant role"* rely on the spatial distribution of these emissions.

3. **Updated Figure and Legend:**
   We have therefore chosen to retain the emissions data in Figure 1 while ensuring that the figure serves both purposes: Introducing the model domain and measurement sites and Showing the spatial distribution of emissions that are discussed later in the manuscript.

Revised text: *"Figure 1. Annual anthropogenic and biomass burning NO emissions (averaged over 2010–2013) and INDAAF measurement site locations, showing the full extent of the model domain used in the simulations. Measurement sites include Banizoumbou (Ba), Katibougou (Ka), Djougou (Dj), Lamto (Lam), Bomassa (Bom), and Zoétélé (Zoe)."*

Lines 399-400: " The variability of simulated NO_2 concentration over the domain of interest is primarily driven by regional biomass burning emissions (see Figure 1)".

Line 405: "In the Biomass burning regions (see Figure 1) …"

Lines 435-437: "Model results and satellite data both show high concentrations of NO 2 in areas such as Sahel and forested regions, where biomass burning plays a preponderant role (see Figure 1)"

Line 491-492: "In the DJF season, strong ozone production occurs between 5°N and 15°N as a result of biomass burning activities (Figure 1)."