

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

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Abstract. In the context of climate change and increasing anthropogenic pressures in Africa, understanding the interactions between atmospheric chemistry, regional climate, and biogeochemical cycles is critical. This study investigates the potential role of biogenic nitric oxide emissions from African soils (BioNO), particularly in arid and semi-arid ecosystems, as significant contributors to atmospheric $NO₂$ emissions and regional atmospheric chemistry. To this end, we rely on a modelling approach

- 5 based on the RegCM5 regional climate model, including an updated atmospheric chemistry module and, amongst other, a specific parametrization for BioNO emissions. Throughout the paper, the model performances are evaluated against various datasets including in-situ observations from the INDAAF network and chemical reanalyses. Sensitivity studies demonstrate that integrating BioNO emissions into the model enhances the accuracy of simulated NO_2 , HNO_3 , and O_3 seasonal cycles and surface concentration magnitudes, while reducing simulated biases against ground based observations. Large differences are
- 10 however still present regarding notably the simulated surface ozone concentration magnitude vs in situ measurements, while these biases are also observed for chemical reanalyses, and a state of the art chemistry transport model used for comparison. Beside outlining the impact and added value of BioNO flux representation for regional atmospheric chemistry, our findings also outline the suitability of RegCM5 coupled system for the study of regional climate, chemistry and nitrogen cycle interactions over Africa.

15 1 Introduction

Tropical Africa is a significant source of gas and particles affecting the regional and global atmospheric chemistry and climate. Beside large chemical sources linked to anthropogenic activities (biomass burning, megacities), there is also a significant amount of biogenic emissions which can significantly interact with regional tropospheric composition across various tropical locations (e.g., Aghedo et al., 2007). Nitrogen emissions originating from soil microbial processes are an important component

20 of these emissions (Fudjoe et al., 2023). Indeed, soil microbial processes, such as nitrification and denitrification involve the production of reactive gaseous compounds released to the atmosphere (Delmas et al., 1995; Medinets et al., 2015; Schreiber

et al., 2012). Soil NO (referred as BioNO) emissions dominate the global net nitrogen oxide exchange between ecosystems and the atmosphere (Ludwig et al., 2001) and have been estimated to approximate 4.7-26.7 TgN.yr⁻¹ above canopy (Davidson and Kingerlee, 1997; Ganzeveld et al., 2002; Hudman et al., 2012; Jaeglé et al., 2005; Müller, 1992; Steinkamp and Lawrence, 25 2011; Vinken et al., 2014; Yan et al., 2005; Yienger and Levy, 1995). BioNO emissions play a crucial role in the formation of particles and key atmospheric gaseous compounds, such as O_3 and HNO_3 (Liu et al., 2020; Mosier, 2001; Vinken et al., 2014; Williams et al., 2009). Soil microbial processes depend primarily on precipitation and soil moisture (Liu et al., 2009; Li et al., 2022). Over tropical Africa, which shows a marked rainfall seasonality, an accumulation of nitrogen in soils occurs during the dry season, leading to significant pulses of nitrogen emissions at the onset of the rainy season (Austin et al., 2004). Various 30 environmental and physical factors, including wind speed, floristic composition, nitrogen input (from organic and synthetic

fertilization and atmospheric deposition), plant cover, soil temperature and moisture, soil pH, and texture, influence BioNO emissions (Delon et al., 2007; Williams et al., 1992).

In tropical Africa, estimating BioNO emissions is however challenging due to a lack of observational data (e.g., Jaeglé et al., 2004; Van Der A et al., 2008). Nevertheless, global and regional modelling approaches, which consider these influences to

- 35 various degrees, have been proposed to quantify BioNO emissions (e.g., Hudman et al., 2012; Stohl et al., 1996; Stehfest and Bouwman, 2006; Yienger and Levy, 1995; Yan et al., 2005) and ensuing chemical impacts. For instance Williams et al. (2009) used an inventory of biogenic emissions derived from multi-annual simulations of the ORCHIDEE vegetation model (Lathiere et al., 2006)to examine the influence of NO and BVOC emissions from soils and vegetation on Equatorial Africa's tropospheric composition. The global chemistry-transport model simulations revealed that NO emissions from soils in Africa might con-
- 40 tribute to 2-45 % of tropospheric ozone production. Delon et al. (2008) used neural network-based parameterization coupled with a mesoscale model to investigate the impact of BioNO emissions on NQ_x and Q_3 production in the lower Equatorial Africa's troposphere on a specific period (6 August 2006) during the AMMA campaign in the Sahel. Their findings indicate an increase in tropospheric O_3 and NO_x concentrations in the lowest few kilometres of the atmosphere, in response to larger BioNO fluxes, compared to simulations using the prescribed inventory of Yienger and Levy (1995). Steinkamp et al. (2009)
- 45 used the ECHAM5/MESSy atmospheric chemistry model (EMAC) to examine the influence of BioNO on lower tropospheric NO_x , $O₃$, PAN, HNO₃, OH, and CH4 lifetime (τ CH4). Their results revealed that BioNO significantly contributes to NO_x levels in numerous regions, especially in the tropics. Moreover, BioNO notably raises OH concentrations, thereby increasing the global troposphere's oxidizing capacity and resulting in an 10% decrease in τCH4 due to soil NO emissions.
- From a modelling point of view, there is a strong interest in integrating interactive BioNO emission in regional climate 50 systems: it can allow for instance to study the impact of present and future climate change and variability, including temperature fluctuations, precipitation patterns, and soil moisture levels, on BioNO emissions. This understanding is crucial for predicting possible future emission trends, potential changes in chemical environment, and developing effective mitigation strategies. Additionally, BioNO emission can affect the formation of ozone and other secondary pollutants, which have implications for air quality and climate forcing and responses, which can be evaluated by means of coupled climate chemistry systems.
- 55 Furthermore, the regional climate modelling approach allows the study of regional environmental disturbances such as land use and agricultural changes in a flexible way. Finally, Since climatic and land use gradients are particularly important in west and

central Africa, the dynamical downscaling capabilities offered by the regional climate modelling approach are also particularly interesting to capture the regional contrasts in emissions and processes at play. One goal of the present study is to evaluate and extend such a system based on the ICTP RegCM5 model (Giorgi et al., 2023), while estimating and analysing the regional

- 60 impact of BioNO emissions (from soils and vegetation) on key tropospheric species relevant to the atmospheric nitrogen cycle. The latter plays an important role in the chemistry of the atmosphere as well as the functioning of aquatic and terrestrial ecosystems and agrosystems (McNeill and Unkovich, 2007; Vitousek et al., 1997). This N cycle is significantly disrupted by anthropogenic activities (agriculture, fossil fuel combustion, and biofuels) (Galloway et al., 2008). This issue is particularly concerning in tropical regions like Africa, where rapid population growth and seasonal cycles from natural and anthropogenic
- 65 sources contribute significantly to changes in N emissions (Adon et al., 2010). We focus on NO_2 , HNO_3 , and O_3 because these species are tightly interconnected and are involved in different environmental and health impacts. $NO₂$, a key reactive nitrogen species, contributes to the formation of $HNO₃$ and $O₃$. $HNO₃$ is formed when $NO₂$ reacts with other substances in the atmosphere, and is an important contributor of rainfall acidity deposition and nitrogen supply to ecosystems. O₃, a powerful oxidant, is influenced by the presence of $NO₂$ and has significant implications in HNO₃ formation. Understanding
- 70 the concentrations and fluxes of these species is essential for assessing nitrogen management and potential risks in Africa. To a large extent, this work shares scientific goals with, and is carried out as part of the INSA (Integrated Nitrogen Study in Africa; https://doi.org/10.3030/871944) and the INDAAF (International Network to study Depositions and Atmospheric chemistry in AFrica; https://indaaf.obs-mip.fr) projects.

The study is structured as follows: Sections 2 and 3 provide a description of the modelling context and developments, as 75 well as measurement sites and relevant databases used in this study. Section 4 will focus on evaluating model performances in simulating key regional climatic features affecting emissions and tropospheric chemistry. Section 5 will discuss simulated BioNO in more detail. Model results and limitations concerning their impact on key atmospheric chemistry components will be discussed in Section 6.

2 Model description

80 2.1 The Regional Climate-chemistry Model RegCM5-CHEM

The present work is based on the latest version of the ICTP RegCM5 discussed in detail in Giorgi et al. (2023). Compared to previous versions, a significant development has been the integration of the MOLOCH non-hydrostatic dynamical core (Davolio et al., 2020) improving model efficiency, notably in view of climate convection-permitting (CP) simulations (e.g., Ban et al., 2021; Coppola et al., 2020; Lucas-Picher et al., 2021; Pichelli et al., 2021; Prein et al., 2015).

85 This development is also important for atmospheric chemistry, which uses a large number of tracers, by reducing the numerical cost of advection in the context of long-term simulations. As for previous model versions, several options are available for the model physics. In the present study we run successive tests (not discussed further here) to retain a model configuration based on the the RRTM (Rapid Radiative Transfer Model) for shortwave and longwave radiation, the University of Washington turbulence scheme (UW, Bretherton et al., 2004), the Nogherotto et al. (2016) bulk microphysics scheme and the Tiedtke

Table 1. Summary of simulations performed for the analysis of regional climate and trace gas in this article.

- 90 convection scheme (Tiedtke, 1989). Continental surface processes are treated by the Community Land Model, version 4.5 (CLM4.5, Oleson et al., 2013) which also provides important coupling variables used through the atmospheric chemistry interface, such as surface resistances, soil humidity and temperature. The meteorological boundary condition fields are provided at a six-hourly frequency, from the ERA5 reanalysis (Hersbach et al., 2020). The sea surface temperature data is prescribed by the OI_WK every week.
- 95 Atmospheric chemistry processes are based on approaches initially developed in Solmon et al. (2006) and Solmon et al. (2021) for aerosol and Shalaby et al. (2012) and Ciarlo et al. (2021) for gas phases. The chemical reaction solver is here based on the CBM-Z photochemical mechanism module (Shalaby et al., 2012; Zaveri and Peters, 1999). It allows a comprehensive coverage of regionally to globally significant species and reactions involved in photo oxidant chemistry, while keeping a good precision and numerical efficiency. CBM-Z notably includes key prognostic species such as O_3 , NO_x, CO, VOCs. Aerosol gas
- 100 partition is treated here using a thermodynamic equilibrium approach and the ISORROPIA-II scheme (Fountoukis and Nenes, 2007; Nenes et al., 1998) mostly relevant for fine particles heterogeneous processes. Dry deposition processes and fluxes are parameterized according to Zhang et al. (2003), for 31 gas phase species in the model. Key input for Zhang deposition scheme include biophysical and physiological parameters, which are prescribed to the model using pre-defined land use categories and mapping (Dickinson, 1986; Zhang et al., 2002). Some parameters, such as LAI, roughness length, wind, surface temperature,
- 105 etc.are provided through the CLM4.5 interface. In the present study, slight modifications have been made in the deposition scheme to account for African regional specificities:
	- a. Default ground resistance (Rg) values for ozone are based on Zhang et al. (2003) were used but for the ocean domain, we adjusted these values by lowering them to obtain more realistic values of dry deposition velocity (e.g., Charusombat et al., 2010; Wu et al., 2011; Zhang et al., 2002) and so surface concentrations over the Ocean.
- 110 b. The friction velocity (u*) is a crucial parameter for calculating aerodynamic resistance (ra). According to Padro et al. (1991), the equation used to calculate ra requires that the Richardson number be maintained below 0.21 under stable conditions. This is particularly important in tropical forested areas with weak mean winds, where both u* and deposition velocity are often lower than those reported in the literature (e.g. Adon et al., 2013; Zhang et al., 2003). To ensure compliance with this criterion, we have set a lower threshold of 0.4 m/s for u* in forests and 0.1 m/s in savannas, based 115 on our statistical analysis.

The wet deposition flux is initially parameterized following the approach developed in the MOZART chemistry transport model (Emmons et al., 2010; Horowitz et al., 2003). 26 CBM-Z tracers are considered for wet removal through large-scale and convective precipitation processes. Compared to Shalaby et al. (2012) and Ciarlo et al. (2021), we significantly upgrade the wet deposition parameterizations by developing a new interface which uses directly cloud to rainwater production and precipitation 120 rate terms diagnosed from both the Nogherotto et al. (2016) stratiform and the Tiedtke (1989) convective rain rates.

Anthropogenic and biomass burning emissions are treated through a preprocessing interface designed for the regional interpolation and chemical aggregation of different possible inventories. In this study, the monthly, 0.1 deg resolution emission inventories from the Copernicus Atmosphere Monitoring Service (CAMS, version 6.2) is considered for non-biomass burning emissions (Soulie et al., 2023). For biomass burning, daily emissions from GFED4 (Global Fire Emissions Data, version 4)

- 125 for 0.25 deg as spatial resolution are considered (Randerson et al., 2018). For both inventories, the lumping of emitted VOC species to effective CBMZ species has been performed following a method similar to Huijnen et al. (2019). The biogenic VOC emissions are calculated on line as part of CLM45 using the embedded MEGAN (Model of Emissions of Gases and Aerosols from Nature; Guenther et al., 2006) scheme. Here only isoprene fluxes are passed to the atmospheric chemistry and transport interfaces (Strada et al., 2023).
- 130 Finally, an important development compared to Shalaby et al. (2012) and Ciarlo et al. (2021) concerns the chemical initial and lateral boundary conditions. The standard monthly climatology approach has been replaced here by a new interface using six-hourly CAMS chemical reanalysis (Inness et al., 2019; Wagner et al., 2021), consistent with the ERA5 dynamical forcing. For important chemical and aerosol species, this allows to represent more explicitly the influence and variability of long range chemical transport events possibly affecting the domain through the boundaries.

135 2.2 The BioNO emission parameterization

Interactive BioNO emissions are represented here following the empirical approach developed in Delon et al. (2007) (D2007) based on an Artificial Neural Network (ANN) supervised learning considering several databases. In some regions such as Africa, few in situ measurements of BioNO are available leading to inaccurate estimates of emissions affecting concentrations in the lower troposphere (Jaeglé et al., 2005). The main advantage in using this ANN algorithm is that it can be connected on

- 140 line and linked to varying environmental parameters of specific regions of interest, and can be used for calculating accurate BioNO emissions whatever the type of soil and/or climate (Delon et al., 2007). For now, this ANN algorithm has been used only in Tropical African climates (Delon et al., 2007, 2012, 2015). The main advantage in using this ANN algorithm is that it's linked to varying environmental parameters of specific regions of interest, and can be used for calculating accurate BioNO emissions whatever the type of soil and/or climate NO emissions are largely influenced by microbial activity, determined by
- 145 the soil physical properties (porosity, soil texture, soil moisture etc.) governing substrate diffusion and oxygen supply (Skopp et al., 1990). D2007's parameterization includes seven explicative variables, including wind speed, fertilisation rate, surface temperature, sand percentage, soil moisture, soil pH, and deep soil temperature (20-30 cm). Wind speed is used as an indicator of varying atmospheric conditions. Deep soil temperature relates to oxygen diffusion and nitrogen mineralization in the soil (Butterbach-Bahl et al., 2004). The sand percentage impacts water diffusion (Roelle et al., 2001). pH is a crucial factor due

- 150 to its impact on chemical or biological mechanisms (Serca et al., 1994). It can also influence NO emissions through chemodenitrification process (low pH) or biological activity (higher pH) (e.g., Ormeci et al., 1999; Serca et al., 1994). Finally the fertilisation rate is key to reflect the amount of externally introduced nitrogen (Sanhueza et al., 1990). The pH and fertilisation rates are prescribed using external databases as inputs: soil pH data are obtained from IGBP-DIS (International Geosphere Biosphere Programme-Data and Information System; Igbp-Dis, 1998), and fertilisation rates, including N fertilizer and N 155 manure, are sourced from Potter et al. (2010). The other variables are integrated on line within the model.
- In output, NO emission fluxes are finally calculated at each model time step, through the resulting equations:

$$
NOflux_{norm} = w_{24} + w_{25} \tanh(S_1) + w_{26} \tanh(S_2) + w_{27} \tanh(S_3)
$$
\n(1)

with

160
$$
S_1 = w_0 + \sum_{i=1}^{7} w_i x_{j,\text{norm}}
$$

\n $S_2 = w_8 + 15 \sum_{i=8}^{23} w_i x_{j,\text{norm}}$
\n $S_3 = w_{16} + \sum_{i=17}^{23} w_i x_{j,\text{norm}}$
\nwith $j = 1 \rightarrow 7$ (2)

NOflux_{norm} represents the normalised NO flux and the seven inputs mentioned above are represented by x_1 to x_7 (surface 165 WFPS, surface soil temperature, deep soil temperature, fertilisation rate, sand percentage, pH and wind speed respectively). Each input x_i is associated with a weight w_i , which represents the strength or influence of that input in determining the final NO flux.

2.3 Model experiments

To test the ability of RegCM5-CHEM to simulate African climate, BioNO emissions and to evaluate its effect on atmospheric 170 chemistry, we conducted two different simulations which are listed in Table 1. The simulations cover the period from January 2010 to February 2013, which includes a variety of climatic conditions and seasonal variations. The first two months are used as spin-up time for both experiments and are not considered in the analysis of the results.

- 1. BASE run: BASE run: With biomass burning and anthropogenic emissions, without BioNO emissions.
- 2. BIONO run: With biomass burning, anthropogenic emissions and BioNO emissions.
- 175 The model is 30 km * 30 km in longitude and latitude, with 35 vertical levels from the surface to 3.6 hPa. The corresponding model time step is 210 s.

3 Data and study sites

3.1 Study sites and ground-based observation of pollutants

- O3, NO2 and HNO3 concentrations are measured in the framework of the INDAAF long-term monitoring project. INDAAF, 180 part of the Aerosol Cloud and Trace gases Infrastructure (ACTRIS-Fr), represents a long term monitoring service to study evolution of atmospheric chemical composition and deposition in Africa. Gaseous concentrations are monthly measured on different sites in West and Central Africa and data are distributed at https://indaaf.obs-mip.fr. Datasets are referenced for each sites using DOIs (https://doi.org/10.25326/608; https://doi.org/10.25326/604; https://doi.org/10.25326/605; https://doi.org/10. 25326/275; https://doi.org/10.25326/607; https://doi.org/10.25326/603). We use observations from six sites representative of 185 the main african ecosystems (Figure 1, Table 2): Dry savannas (Banizoumbou, Katibougou), Wet savannas (Lamto, Djougou) and equatorial Forests (Bomassa, Zoétélé). Gas monitoring at Banizoumbou, Katibougou, Lamto, Bomassa and Zoétélé started
	- in 1998, with the Djougou site joining in 2005. Atmospheric gas concentrations (NO_2, HNO_3, O_3) are performed using passive sampling techniques based on the methodology outlined by Ferm et al. (1994). Developed by the Laboratory of Aerologie (LAERO) in Toulouse within the framework of the INDAAF project, these passive samplers underwent rigorous testing across
- 190 various tropical and subtropical regions (Adon et al., 2010; Carmichael et al., 2003; Ferm and Rodhe, 1997). Continuous measurements are ongoing at all INDAAF sites, and data are available for the entire period of our study from January 2010 to February 2013. Although there are some missing data for certain months in specific ecosystems, the overall dataset provides a comprehensive view for the analysis.

Detailed description and accuracy of INDAAF passive samplers are given in previous African studies, encompassing rural and 195 urban sites (Adon et al., 2010; Bahino et al., 2018; Carmichael et al., 2003; Ferm and Rodhe, 1997; Galy-Lacaux et al., 2009;

Figure 1. Annual anthropogenic and biomass burning NO emission (averaged over 2010-2013) and INDAAF measurement sites localization: Banizoumbou, Katibougou, Djougou, Lamto, Bomassa and Zoétélé.

Galy-Lacaux and Modi, 1998; Ossohou et al., 2019, 2023). Monthly mean concentrations (used here as reference) are derived from the duplicates' means. Upon completion of the exposure period, all samplers, including field blanks, undergo laboratory analysis using ionic chromatography. The LAERO bi-annualy participated in the WMO-GAW (World Meteorological Organization - Global Atmosphere Watch) quality assurance program since 1996, evaluating the precision of ionic chromatography 200 measurements for trace compounds twice yearly. Results, accessible under reference 700106 at http://qasac-americas.org/, consistently demonstrate analytical precision of 5% or better for all ions. Additionally, the measurement accuracy of passive samplers, assessed through covariance with duplicates, was estimated at 9.8% for NO_2 , 20% for HNO_3 , and 10% for O_3 (Adon et al., 2010). Detection limits, determined using the exposure period and field blanks for the studied duration, are reported as 0.2 ± 0.1 ppb for NO₂, 0.07 ± 0.03 ppb for HNO₃, and 0.1 ± 0.1 ppb for O₃.

205 3.2 Climatic and chemical evaluation datasets

For further model evaluation, we use a variety of sources, including data from meteorological stations and satellites, as well as reanalysis products. Table 3 summarises information about all variables used from each database. Precipitation data are obtained from the Tropical Rainfall Measuring Mission 3B42-version 7 (TRMM; Huffman et al., 2007). Temperature data are sourced from the Climatic Research Unit version TS4.03 (CRU; Harris et al., 2020). For circulation dynamics, we use data

210 derived from the European Environment Agency version 5 (ERA5; Hersbach et al., 2020). CRU data is exclusively based on

in situ observations, while TRMM data originates from satellite observations. As mentioned above, ERA5 reanalysis data is derived from a combination of in situ measurements and satellite observations assimilated in a Numerical Weather Prediction model simulation. By using different sources of observational data (in situ and satellite) as well as reanalysis estimates, for precipitation (Pr), wind field (U,V, and W), and 2 m surface temperature (T), we account for associated uncertainties, particularly 215 in West Africa.

For the chemical evaluation, the model outputs were compared with the INDAAF in-situ measurement database. To complement this local evaluation we also compare the model to CAMS chemical reanalysis (Inness et al., 2019; Wagner et al., 2021), and to outputs from the state of the art chemistry transport model GEOS-Chem (Goddard Earth Observing System-Chemistry; https://doi.org/10.5281/zenodo.3974569), and ground-level NO₂ concentrations derived from OMI and TROPOMI satellite 220 $NO₂$ observations (Cooper et al., 2022).

GEOS-Chem is a global 3D Chemical Transport Model (CTM) driven by assimilated meteorological observations from NASA's Goddard Earth Observing System (GEOS). It addresses atmospheric composition issues across local to global scales. CAMS (Copernicus Atmosphere Monitoring Service) provides chemical reanalysis data by assimilating diverse observational sources, including satellite and in-situ measurements, which improves the accuracy of the simulated chemical fields.

- 225 The ground-level NO₂ concentrations were derived from OMI and TROPOMI satellite observations (Cooper et al., 2022). Initially determined using OMI, these NO₂ column densities were downscaled using TROPOMI, then converted to surface concentrations with the GEOS-Chem model, constrained by ground monitoring data (Cooper et al., 2020, 2022). Both CAMS and GEOS-Chem are subject to their own model uncertainties, however with an added observational constraint in the case of CAMS reanalysis. TROPOMI-derived $NO₂$ data also carries uncertainties, particularly in the conversion from column densities
- 230 to surface concentrations, which may introduce discrepancies. These include potential errors in satellite retrievals and air mass factor calculations.

4 Regional Climate validation

The simulation of African climate by the model is evaluated in terms of seasonal and daily mean for the 3 simulated years. An exhaustive analysis of climate simulations is out of the scope of this paper. We report here model performance in terms 235 of temperature, precipitation and monsoon circulation, which are key features of the African climate, strongly impacting atmospheric chemistry.

Figure 2 compares the ERA5 reanalysis to simulated mean wind at 850 hPa i.e. in the monsoon layer. Overall the model manages to reproduce the main features of the monsoon circulation with a mean characteristic southwesterly flow up to about 17°N, and Harmattan-like circulation over northern Africa. However RegCM5 tends to underestimate the intensity of the

240 mean monsoon flow from the Gulf of Guinea to the Sahel. The monsoon front and the Saharan Heat Low (SHL) are also reasonably captured by the model, although the SHL amplitude is somehow underestimated (Figure 2). This could explain or contribute to the weaker Monsoon flux and the sahelian precipitation underestimation discussed further (Figure 3-c) following the connections described in Peyrillé et al. (2007), Lavaysse et al. (2009), Chauvin et al. (2010), Lavaysse et al. (2010) and Evan et al. (2015). Outside of the monsoon domain, the central equatorial Africa wind minimum is consistently captured.

Figure 2. JJA Monsoon wind speed at 875 hpa for ERA5 reanalysis and RegCM simulation.

245 RegCM5 captures the patterns and spatial gradients of the 2 m surface temperature from hot Sahara regions to colder tropical forests, but with a cold bias over the northern Sahel/Southern Sahara (range -5 to -1 °C) during the monsoon season

(JJA) (Figure 3-f). Attributing surface temperature bias to specific causes is uneasy due to temperature surface-atmosphere interactions and feedbacks (Sylla et al., 2012; Tadross et al., 2006). For Sahelian and Sahara regions RegCM shows a negative bias which is likely linked to a bias in surface radiative budget, depending on simulated surface SW and LW net radiation 250 related to surface radiative parameters, possible excessive high level cloudiness (e.g., Sylla et al., 2012; Zittis et al., 2016) or/and aerosol for instance (e.g., Lavaysse et al., 2011; Wang et al., 2015). In the heat low region, this cold bias is likely consistent with weaker monsoon flux and sahelian precipitation. In the equatorial region the temperature bias could be linked, on the contrary, to excessive cooling induced by overestimated precipitations. Sylla et al. (2012) showed that cold bias in surface temperature is generally consistent with positive rainfall bias. Locally the surface temperature overestimation over coastal central Africa 255 has also been noticed by Mbienda et al. (2023). This bias in Central Africa might stem from an inadequate modelling of the low-level cloud cover that is typical of this area (Philippon et al., 2019). For precipitation, simulated values vary from 0 to more than 13 mm/day over the study domain, with a consistent spatial/seasonal (Figure 3a-b) pattern of precipitation compared to TRMM observation. In general, the most predominant biases in our simulation are an underestimation in Sahel and Central Africa and an overestimation closer to equatorial regions (about from -5 to -0.5 in Sahel/Central Africa and 1 to 260 5 mm/day in Cameroon Highlands). We also observe the rain belt over the Sahelian region, associated with the InterTropical Convergence Zone (ITCZ) stretching from the mountains of Darfur in East Africa to the Guinea Highlands and downstream into the Atlantic. In some subregions, observation data sets do not fully agree. For instance, in comparison to GPCP data, which is consistent with gauge-based precipitation datasets in Africa (Sylla et al., 2013b), TRMM data shows weaker precipitations

265 This variability amongst observations should be kept in mind while evaluating the model results. As outlined in many studies, the model precipitation is extremely sensitive to the choice of parameter combinations used in the physics configurations, such as convection, land surface scheme, boundary layer, etc. The number of parameter combinations is large and model optimization is a complex and often time-consuming issue (e.g., KhayatianYazdi et al., 2021). With the present set up, the main precipitation and temperature biases remain reasonable, especially considering the range of bias shown by state of the

over East Africa and the Guinea Highlands, and over the Cameroon Highlands (Nikulin[∗] et al., 2011; Nikulin et al., 2012).

- 270 art CMIP6 (Coupled Model Intercomparison Project phase 6) Global Climate Models and CORDEX (COrdinated Regional Downscaling EXperiments) RCMs for the African climate simulations (e.g., Bucchignani et al., 2018; Zittis et al., 2016). For the west African region, Figure 4 shows a time-latitude Hovmoeller diagram of precipitation zonally averaged between 10°W and 10°E for the RegCM5 model and TRMM observational data. Both in model and observations, the three characteristic phases of the African monsoon (Hourdin et al., 2010; Koné et al., 2022; Sultan et al., 2003) are highlighted. For TRMM, the
- 275 onset of the rainy season occurs in mid-April and extends until mid-June, as evidenced by the core of the rainfall band along the Guinea coast between approximately 4° and 7°N, while the simulations show a delayed onset from mid-May to June with lower intensity rainfall. In TRMM observations, the monsoon phase itself is characterised by a shift of the rainfall maximum band between July and September reaching 15°N. For this phase, the simulations show a consistent northward precipitation shift, but the maximum does not penetrate as far north as in the observation, in relation with the Sahelian precipitation underestimation
- 280 pointed out earlier. The monsoon withdrawal is observed with precipitation gradually shifting towards the coast. The late season precipitation rainfall is more intense in our simulations compared to TRMM. Again these characteristic seasonal patterns are

Figure 3. Summer (JJA) means Precipitation and Surface Temperature Biases (TRMM and CRU vs. RegCM5), and Frequency of Daily Temperature and Precipitation Events (ERA5, TRMM, and RegCM5). Units: Precipitation in mm/day, Temperature in °C.

rather satisfactorily captured by the model in reference to state of the art climate models.

Beyond seasonal means, simulated daily temperature and precipitation intensity variability are also analysed using Probability Density Functions (PDFs) (Giorgi et al., 2023) and compared to ERA5 and TRMM respectively. These are presented here, for 285 June-July-August of the study period, for the model and the observations in Figures 3-g and 3-h on the Sahel region, which has a particular meteorological regime for precipitation and where the relevant impact of BioNO occurs: Sahel (10°W-10°E; 10°N-16°N).

For the daily temperature PDFs (Figure 3-h), the model simulation captures the observed distribution, although over the region, RegCM5 peaks are slightly shifted compared to the ERA5 data. Overall we observe that simulated values between 20 and 30 290 °C appear with a high frequency of occurrence (more than 18 %) while we have less than 15 % for temperatures above 30°C. However, ERA5 presents the higher frequency of occurrence for high temperatures (25-35 °C) over the region with 2-16 %, against 1-13 % for simulated values. This can also be linked to the underestimation shown by RegCM5 compared to the observation (Figure 3-f).

Figure 4. JJA Hovmoller diagram of monthly precipitation (mm.day⁻¹) averaged between longitudes 10°W and 10°E of the study period.

The corresponding precipitation PDFs are intercompared in Figure 3-g. Overall, RegCM5 is close to TRMM in low (less than 295 10 mm/day) and high (more than 100 mm/day) precipitation intensities. Over the domain, RegCM5 is characterised by higher frequencies of mid precipitation intensities (between 10 and 60 mm/day) than TRMM. For high precipitation intensities (more than 70 mm/day), simulated precipitation intensity frequencies remain lower than the observed ones, consistently with the mean precipitation bias shown in Figure 4 (cold bias for sahel region).

The simulation quality is considered at this stage good enough for supporting further analysis focusing on atmospheric chem-300 istry.

5 BioNO fluxes

Since soil moisture is an important driver of microbial activity and BioNO emissions (Skopp et al., 1990), we evaluated simulated soil moisture compared to the Famine Early Warning Systems Network Land Data Assimilation System FLDAS

- (McNally et al., 2017, 2018) (Figure 5). As shown in the figure, JJA soil moisture is larger in subregions dominated by dense 305 vegetation (Forests regions: -7°N-4°N) and where precipitation events are more intense. Despite the overall slight underestimation of simulated soil moisture compared to FLDAS observed data, especially in Saharan regions, RegCM captures the spatial distribution of soil moisture both in DJF and JJA. FLDAS integrates various observational datasets and uses advanced modelling techniques to provide soil moisture data, but its accuracy in arid regions like the Sahara can be challenging due to sparse in-situ data and the extreme dryness of the environment. Soil moisture data in such regions are often derived from
- 310 remote sensing sources like microwave satellites, which can struggle with accuracy in arid zones where ground measurements are extremely rare (Rao et al., 2022). The BioNO emission spatial distribution reflects the influence of the different explicative variables considered in the D2007 ANN. Overall, in both seasons, weak BioNO emissions in Saharan regions (above 16°N), are associated with low N content, no N input, low soil moisture and a high sand percentage. A high sand percentage leads to increased evaporation and drainage rates (Delon et al., 2008), which prevents the soil from retaining enough water to support
- 315 the microbial processes responsible for NO emissions. Between 8 and 16°N (Sahel), higher BioNO emission spots result in a combination of large soil moisture, latitudinal distribution of soil pH (less high), and significant nitrogen input (shown in Potter et al., 2010). The seasonal variability of Sahelian emission hot spots is mostly driven by soil humidity as illustrated on Figure 5. There is also a significant canopy inhibition factor in the region of large LAI which for example reduces forest emission to the atmosphere. As outlined in Delon et al. (2008), the Artificial Neural Network (ANN) algorithm used in this study tends to be
- 320 more suitable for the Sahel region because it is primarily trained on data from semi-arid regions and temperate zones, making it more uncertain in forested regions where the environmental conditions and soil characteristics differ significantly. In forested areas, factors such as dense canopy cover (affecting soil temperature and moisture), higher organic matter content (affecting nitrogen cycling dynamics), and different microbial communities influence soil processes and NO emissions (Davidson et al., 2000; Pilegaard, 2013) differently compared to semi-arid and temperate zones. This discrepancy highlights the need for further
- 325 region-specific training data to improve the model's accuracy in diverse ecosystems. The limited flux measurements for BioNO emissions in Africa make a systematic evaluation of the model challenging. In this study, simulated BioNO range from 0.02 to 7 mg.m⁻².day⁻¹, i.e from 0.1 to 37.52 ngN.m⁻².s⁻¹ as seasonal averaged flux over the domain. This is within the range given by Delon et al. (2008) (from 0.43 to 6.52 mg.m⁻².day⁻¹) and Davidson and Kingerlee (1997) (from 0.5 to 28 ngN m⁻².s⁻¹). The simulated BioNO are also rather consistent with the measured fluxes
- 330 from the flight B227 observed data of the British Aerospace 146 (BAe-146) under the Facility for Airborne Atmospheric Measurements (FAAM) program: 0.8 to 35 ngN m⁻².s⁻¹, but remain greater than the Ganzeveld et al. (2002) estimation : 2.32 to 11.6 ng N.m⁻².s⁻¹, for the Sahel region. Our simulated BioNO are in better agreement with NO fluxes from soil measured during the DACCIWA field campaign West Africa) in June and July 2016, which ranged from 0 to 48.39 ngN m⁻².s⁻¹ (Pacifico et al., 2019). Feig et al. (2008) also obtained NO flux fields from 4.7 to 27.01 ngN m^{−2} s¹, but for South AFrica. The summary
- 335 of all these estimates can be found in table 4 and additional measurements of BioNO emissions from wet African savannas can be found in table 7 of Delon et al. (2012).

As for the total amount of nitrogen emitted from BioNO on the whole domain, simulated values range from 0.01 to 4.4 TgN.month⁻¹. If we downscale for the Sahel region (10W 10E, 10N 20N, and area of 2.3 10⁶ km²) as done in Stewart et al.

Figure 5. Simulated BioNO emissions (in mg.m⁻².day⁻¹) and Soil Moisture Comparison for DJF and JJA Seasons: Analysis Using FLDAS Noah Land Surface Model L4 for 10-40 cm Soil Depth (in %).

(2008), the simulated emissions amount ranged from 0.0006 to 0.23 TgN. month−¹ . This value is consistent with Stewart et al. 340 (2008) estimates of 0.03 to 0.3 TgN for 2 months (July and August) and Yan et al. (2005) estimates of 1.373 TgN.yr⁻¹ for the entire Africa. Delon et al. (2010) also showed an annual estimate of 0.35 TgN. yr−¹ over the Sahel region. Vinken et al. (2014) showed on the same domain (Sahel) an annual total of 0.52 TgN. yr⁻¹ with a top-down soil NO_x emission inventory for 2005 based on retrieved tropospheric NO₂ column from the Ozone Monitoring Instrument (OMI).

Our total estimated amount is consistent with Williams et al. (2009) estimated BioNO in the same Sahel region (0.575 TgN. 345 yr⁻¹) which is based on biogenic emission inventories provided by Granier et al. (2000) and Lathiere et al. (2006). It's worth

Regionc/Biomes Range of fluxes (ngN.m⁻².s⁻¹) Period Citation Sahel 2.32 - 35.29 August 2006 Delon et al. (2008) Sahel 2.32 - 11.6 - Ganzeveld et al. (2002) Niger 0.8 - 35 August 2006 Flight B227 (https://www.faam.ac.uk) West Africa $0 - 48.39$ June-July 2016 Pacifico et al. (2019)) Semi-arid savanna (South Africa) 4.7 - 27.01 June 2003-October 2005 Feig et al. (2008) Semi arid sahelian rangeland (Dahra, Sénégal) 2 - 10 July 2012, July 2013, November 2013 Delon et al. (2017)

Table 4. Summary of some BioNO emissions estimates from litterature

noting that our estimated emission fluxes, particularly in the Sahel region, are higher than those reported by Simpson and Darras (2021) using the EMEP MSC-W model (Meteorological Synthesising Centre – West of the European Monitoring and Evaluation Programme), part of the Copernicus Atmosphere Monitoring Service (CAMS) project. Their estimates ranged from 0 to 3.5 mg m−² day−¹ over the study period and region. The ANN algorithm incorporates, here additional surface controlling 350 parameters, which may explain the higher emission fluxes obtained in our study.

6 RegCM simulations and impact of BioNO emissions

6.1 Regional and local nitrogen

6.1.1 *NO*² *concentration*

- 355 We first analyse the simulated seasonal surface nitrogen $(NO₂$ and $HNO₃)$ concentrations for the BASE run over the domain, in comparison with the CAMS reanalysis (Inness et al., 2019; Wagner et al., 2021) and outputs coming from Geos-Chem model. Simulated $NO₂$ concentration's variability over the domain is primarily driven by regional biomass burning emissions. We thus note consistent spatial and seasonal patterns between RegCM simulations, CAMS and the Geos-Chem (Figure 6) reflecting the broad spatial agreement between the different biomass burning emission inventories considered in these models.
- 360 However sub-regional details, for instance over west Africa, can be clearly distinguished in higher resolution models against coarse Geos-Chem. Compared to the CAMS reanalysis and to Geos-Chem, the RegCM-BASE shows lower surface $NO₂$ concentrations (about 0.7-0.9 ppb below), especially in the Sahel regions during the summer. In the Biomass burning regions, these differences are less visible. The biomass burning regions, which are primarily located in the central (in DJF) and southern (in JJA) parts of Africa, are areas where extensive burning occurs.

- 365 This difference in NO² concentration magnitude can be attributed to possible differences in NO emissions (biomass burning, anthropogenic inventories and BioNO) as BioNO emissions were not accounted for in the BASE simulation (cf section 2.3). Differences in biomass burning injection heights and nitrogen fluxes considered in RegCM versus CAMS and Geos-Chem could also explain differences in surface concentrations. The introduction of BioNO fills this gap to some extent, by increasing surface NO₂ concentration, bringing it closer to, or even exceeding CAMS values (Figure 6b,f). This is especially apparent 370 in transitional ecosystems such as savannas and grasslands. To assess the potential importance and to quantify the impact of BioNO emission on lower troposphere NO2, Figure 9 shows the difference between the BioNO and the BASE simulations previously described. The plots are shown on a vertical cross section averaged between -10 and 10 °E, from 4 to 21 °N.
- When comparing RegCM5 results to the TROPOMI-derived annual mean ground-level $NO₂$ concentrations (7) over the period 375 2010-2013, it becomes apparent that the model generally performs well in capturing the spatial patterns of NO2 across the region, particularly in transitional ecosystems where the impact of BioNO is significant. However, the model tends to overestimate NO₂ concentrations compared to the TROPOMI observations. This overestimation could be attributed to several factors, including the representation of biogenic and biomass burning emissions in the model, as well as the inherent uncertainties in the satellite-derived data. As detailed by Cooper et al. (2022), uncertainties in the conversion of satellite-observed $NO₂$ 380 column densities into surface concentrations can lead to discrepancies. These uncertainties include potential errors of around 10% in the retrieval of slant columns from satellite radiances and errors ranging from 23 to 37% in the calculation of air mass factors. Moreover, the use of ground-monitor data to constrain the model may introduce biases, particularly in regions with sparse monitoring data. These factors suggest that while RegCM provides a reasonable representation of NO2 distribution, the apparent overestimation relative to TROPOMI may also be partly due to these uncertainties in the satellite-model processed
- 385 data.

In this study, we also compare the RegCM5 simulations to the OMI/Aura-derived tropospheric NO2 columns (https://doi. org/10.5067/Aura/OMI/DATA3007) over the period 2010-2013 (Figure 8). The satellite data come from the Level-3 daily global gridded $0.25x0.25$ degree OMI NO₂ product (OMNO2d), which provides total and tropospheric NO₂ columns for all atmospheric conditions and is cloud-screened for sky conditions where cloud cover is less than 30%. With its high spatial

- 390 resolution (0.25° x 0.25°), this dataset is well-suited for studying large-scale $NO₂$ distributions in the troposphere. It becomes clear that the model captures the general spatial distribution of $NO₂$ across the region. Both the model and the satellite data show high concentrations of $NO₂$ in areas such as Sahel and forests regions, where biomass burning plays a significant role. The spatial correlation analysis shows moderate agreement between the RegCM5 simulations and the satellite-derived data. Specifically, the Pearson Correlation Coefficient (PCC) indicates a correlation of 0.32 when excluding BioNO emissions (No
- 395 BioNO) and a correlation of 0.22 when BioNO emissions are included in the simulation. This suggests that the addition of BioNO emissions reduces the overall correlation with the OMI observations. One potential explanation for this reduction could be related to the fact that OMI measures the entire tropospheric column, which may not fully capture diffuse biogenic emissions or localized sources (e.g., Ossohou et al., 2019) that contribute to near-surface concentrations represented in the model. Despite

some discrepancies in specific regions, the spatial correlation suggests a reasonable alignment of $NO₂$ patterns between the 400 model and the satellite observations, particularly in regions where biomass burning and other large-scale processes dominate.

The effect of BioNO emissions from the ANN algorithm at the regional scale, leads to an overall increase in $NO₂$ seasonal mean concentrations ranging from 0 to 2-4 ppb. This increase also extends to the lower troposphere, as illustrated in Figure 9. The maximum increase occurs over the Sahel region (especially 10-21°N) and can reach up to 3 ppb in JJA, consistently with increased BioNO fluxes in this region. We can note a general positive correlation between the BioNO fluxes emissions

- 405 (Figure 5a,d) and the difference [BIONO BASE] simulations at the surface level (Figure 16a,d). This increase in surface $NO₂$ concentrations over the domain is consistent with Delon et al. (2008) results who report that a local BioNO induced NO₂ increase by up to 0.9 ppb at 4°E and between 7°N-21°N for August. Based on global simulations, Steinkamp et al. (2009) found an increase in global NO_x mean mixing ratio in the lower troposphere reaching 7 and 17 % for DJF and JJA respectively. To further examine the simulated $NO₂$ concentrations, we compare the results with monthly averaged surface concentrations
- 410 from the INDAAF stations. Simulated outputs at the lower model level (around 40 m above the ground level) are interpolated to the site locations for the simulated period. BIONO and BASE biases (based on the 3-years monthly-averaged mass concentration) (Table 5) and correlation with observations (Figure 11) were calculated for the pollutant at the corresponding stations.
- Figures 10 and 11 show that the BASE model experiment tends to generally underestimate the $NO₂$ concentrations except for 415 december-january in wet savannas (Lamto, Djougou). This is also generally the case for the CAMS reanalysis (Figure 10). RegCM5-BASE NO² concentrations tend to be underestimated especially over dry savannas (Banizoumbou, Katibougou) in the wet season, where the maximum negative bias recorded could reach ≈ -4 ppb in June and October at the Banizoumbou site. Mostapha et al. (2019) used the RegCM4-CHEM and also showed that the model mostly underestimates, compared to the Greater Cairo observation (Egypt), the monthly averages of $NO₂$ concentrations at four representative sites with maximal
- 420 underestimation in April. Taking into account BioNO emissions has a very significant impact on reducing the dry savanna's wet season bias, as illustrated on figures 10 and 11. Figure 10 suggests that this reduction can bring for instance the model maximum negative bias to ≈ -3.94 ppb when BioNO are accounted for, vs -4.62 ppb in the BASE run, in July at Banizoumbou. We can however notice on figure 9 that the model (BIONO run) tends to produce maximum concentration in the middle of the rainy season, while the observations tend to show that maximum NO2 concentrations occur at the beginning and end of the wet
- 425 season (in a sort of bi-modal pattern). This could be due to nitrogen pool limitation not accounted for by the ANN approach which reacts only to environmental conditions. Indeed, soil N content in the Sahel shows a maximum at the end of the rain season when senescent herbaceous biomass begins to decompose, leading to increased BioNO fluxes (not represented by the model). The temporal distribution of rain events might be also at play here, with peak of emissions occurring preferentially for rainfall events consecutive to a dry period, which are more likely at the beginning and end of the rainy season (Gasche and
- 430 Papen, 1999; Hickman et al., 2018; Johansson et al., 1988; Jaeglé et al., 2005; Yienger and Levy, 1995). For wet savannas, the model and measurements are rather consistent in the BASE run. During the dry season, positive biases are present in the BASE run for wet savannas (unlike dry savannas), ranging between 0.2 and 1.6 ppb. The highest $NO₂$

surface concentrations in the dry season are linked to biomass burning emissions (Oppenheimer et al., 2004; Van Marle et al.,

2017). Moreover, Ossohou et al. (2019) suggested that NO₂ concentrations in the dry season could be mainly attributed to the 435 intensity of biomass burning sources in all the six sites except Banizoumbou and Katibougou. This context helps to explain the observed positive biases, which might be enhanced by the addition of BioNO emissions. While BioNO emissions lead to a small improvement in simulations, especially for the Lamto station during both wet and dry seasons, they tend to worsen these biases. For instance, the maximum positive bias is increased by 1.2 ppb in January at the Djougou site (79.9% increase in bias). The BioNO emissions are possibly overestimated due to larger, and excessive, response of the ANN to soil moisture 440 in wet savanna compared to dry savanna in both seasons (5), which can also combine with a smaller canopy reduction factor when compared to forested regions.

Table 5. BASE and BIONO Biases (ppb) for NO₂. The Reduced/Increased (Red/Inc) biases given by BIONO run is in %. Ba: Banizoumbou, Ka: Katibougou, La: Lamto, Dj: Djougou, Bo: Bomassa, Zoétélé: Zo

^a Bias with BASE simulation

 \real^b Bias with BIONO simulation

 $\,^c$ Reduction/Increase Bias by BioNO emissions

For tropical/transition forest ecosystems, both the BASE simulation and CAMS reanalysis show a significant $NO₂$ underestimation compared to INDAAF measurements. The discrepancies observed in the BASE and CAMS simulations could be influenced by several factors, including the representation of NO_x sources such as anthropogenic emissions near the surface 445 and regional chemical processes. Furthermore, INDAAF stations are often located in areas with strong simulated $NO₂$ gradients (BIONO run), particularly in DJF, where even slight spatial discrepancies could lead to significant differences between simulated and observed levels (challenge of regional representativity of the INDAAF stations). The introduction of BioNO emissions in the simulation helps to reduce these discrepancies by enhancing NO_x concentrations, which in turn brings the simulated ozone levels closer to those observed (Figures 10 and 11).

450 6.1.2 *HNO*³ *concentration*

On Figure 12, simulations exhibit in general a consistent representation of $HNO₃$ spatial distributions between the different models. While RegCM concentrations are rather close to CAMS in magnitude, both are significantly smaller than Geos-Chem, in relation to the O_3 fields as discussed later. Over the Sahel, the lower JJA simulated HNO₃ concentrations (BASE run) are likely associated with the previously discussed underestimation of modelled NO_2 in the regions (5° -20°N), since HNO₃ is a

- 455 product of $NO₂$ oxidation. In general for both seasons, BioNO contribute to increased spatial concentrations of $HNO₃$, bringing RegCM simulations closer to reanalysis and Geos-Chem as shown by Figure 12. According to Figure 9, the lower troposphere $HNO₃$ increase can reach 0.3 ppb with maximum increases correlated with strong BioNO emissions (Figures 5a, 5d), as for $NO₂$. The effect of BioNO on $HNO₃$ is somehow less sensitive compared to $NO₂$. This sensitivity difference is likely due to chemical controls occurring through OH and O_3 formation (Steinkamp et al., 2009).
- 460 HNO₃ formation pathways from NO₂ involve the presence of oxidants such as OH and O_3 , which are controlled by regional photo-oxidant chemistry and emissions. As a result, the impact of BioNO on simulated $HNO₃$ is not as straightforward as for $NO₂$.

As for NO₂, both RegCM5 and CAMS show an overall large underestimation of HNO₃ concentrations compared to available measurements over nearly all INDAAF stations (Figure 13). The corresponding biases are large in the wet season and smaller

- 465 in the dry season when the contribution of biomass burning is more important relative to BioNO (Figure 13/Table 6). The maximum bias in late June for wet season over dry savannas is likely to be partially due to the same reasons as discussed previously for the monthly-averaged NO_2 concentrations. Lin et al. (2013) argue that enhanced soil emissions and higher NO_x oxidation rate under warmer conditions can generate high atmospheric HNO₃. Despite the large remaining underestimation, due to the less sensitive improvement across all ecosystems, the addition of BioNO in RegCM5 helps reduce the biases and
- 470 brings the model closer to the observations (Figure 13). Figure 13 indicates an overall increase at all the study sites and a reduction in negative bias: a maximum of -1.67 vs -1.73 ppb in June (3.51% reduction in negative bias) in dry savannas, -0.77 vs -0.8 ppb in April (4.56% reduction in negative bias) in wet savannas, and around 2.27% reduction in negative bias over forests.

In addition to improving the amplitude of surface concentrations at the six remote sites and across the region, the introduction 475 of ANN on-line emissions also improves the spatial correlation between the simulated (RegCM) and observed (INDAAF)

Table 6. BASE and BIONO Biases (ppb) for HNO₃. The Reduced/Increased (Red/Inc) biases given by BIONO run is in %. Ba: Banizoumbou, Ka: Katibougou, La: Lamto, Dj: Djougou, Bo: Bomassa, Zoétélé: Zo

 a Bias with BASE simulation

 b Bias with BIONO simulation</sup>

^c Reduction/Increase Bias by BioNO emissions

concentrations. The BioNO induced enhancement is also associated with a more realistic seasonal evolution of $NO₂$ and HNO³ surface levels when compared to INDAAF observations (Figures 11 and 14).

6.2 Regional and local Ozone

Together with transport, emission and deposition processes, ozone photo-chemistry regulates the content of nitrogen com-480 pounds in the atmosphere. Tropospheric Ozone simulation is in general very challenging due to numerous sources of variability and uncertainties (Young et al., 2018). Such simulations involve complex and interrelated factors, including precursor emissions, meteorological variability, ozone photochemical production and loss, surface deposition, long-range transport influence

and stratosphere-troposphere exchange (Lelieveld and Dentener, 2000). In this section we discuss the ability of the model to represent regional ozone and the subsequent impact of BioNO emissions on regional ozone production. Figure 15 displays the 485 regional surface ozone simulated by RegCM5-CHEM for BASE and BioNO runs, compared to the CAMS chemical reanalysis and the Geos-Chem model. The strong seasonality of surface ozone concentrations (winter vs summer) is first highlighted.

In the DJF season, strong ozone production occurs between 5°N and 15°N as a result of biomass burning activities. RegCM5-CHEM shows a consistent spatial pattern with CAMS and Geos-Chem in terms of simulated surface concentrations, but with lower values in the source zones (Geos-Chem showing the larger concentrations). In areas where local chemical production is

- 490 low such as over the Sahara, long range and vertical ozone transport essentially determines the background ozone level (e.g., Sauvage et al., 2005). We can outline here the added values of improved chemical boundary conditions which set up a realistic and climatically relevant seasonal ozone background compared to CAMS reanalysis, while also accounting for long range transport events at shorter time scale.
- During summer months (JJA), we also observe consistency between the continental-scale surface ozone gradients simulated 495 by RegCM5 and those from CAMS and Geos-Chem. In northern Africa, there is a only a slight overestimation of ozone, which can be attributed to enhanced vertical transport and mixing during the African monsoon (the south-to-north transport more effective, Sauvage et al., 2007), potentially combined with an overrepresentation of stratosphere-troposphere exchange and local photochemical production under strong solar radiation (Li et al., 2019). However, in the southern biomass burning regions ozone "hot-spots" (Sauvage et al., 2007), RegCM5 tends to simulate lower surface ozone concentrations compared to 500 CAMS and more noticeably, to Geos-Chem.
-

Focusing here on surface concentration only offers a limited view of actual model to model differences, and a deeper tropospheric ozone budget assessment would be required for a more systematic quantitative analysis, which is beyond the scope of this paper. Studies have shown that discrepancies in model ozone simulations are in general large in tropical regions. For example, a comprehensive intercomparison between Geos-Chem and CAM-chem (e.g., Lin et al., 2024) highlighted significant

- 505 differences in ozone budgets and vertical profiles due to variations in photolysis schemes, aerosol interactions, and convective transport processes. These differences can lead to variations in how ozone is transported vertically, impacting surface concentrations (e.g., Li et al., 2019). Moreover, recent intercomparison studies focusing on tropospheric ozone in various tropical regions, including Africa, confirm that such discrepancies are common and often linked to model-specific handling of emissions, injection heights and vertical dynamics (Huijnen et al., 2020; Lin et al., 2024). For instance, Tsivlidou et al. (2023) argue
- 510 that it is essential to consider the combination of injection height of ozone precursors and the strong vertical mixing in the tropics which largely determine the surface ozone values. These factors, along with the non-linear interactions between NO_x and VOCs, including the uncertain biogenic emissions, must be carefully analysed when assessing model outputs. Figures 16c and 16f illustrate the influence of BioNO emissions on the RegCM5 simulated surface O_3 field. The consecutive

production or depletion of O_3 is not solely dependent on NO_x concentrations but also on the $NO_x/VOCs$ ratio, which deter-515 mines the ozone chemical regime variable in different subregions of the domain. At the regional to continental scale and for both seasons, the introduction of BioNO leads to both an increase and a decrease in surface ozone production, with a predomi-

nantly increasing effect in the lower troposphere (Figures 9c,f and 16c,f). In regions coinciding with intense BioNO emissions

and for both seasons, there is however a notable negative impact on surface O_3 . This decrease in ozone levels, which can reach up to 2 ppb, is likely due to ozone titration processes, more characteristic of VOC-limited conditions. In areas with higher 520 proportion of NO_x emissions (here BioNO source areas), O_3 formation can be VOC-limited or may shift between chemical regimes (Kleinman, 1994; Sillman and He, 2002) e.g depending on the time of the day.

As one moves away from these intense sources towards the regional scale, the ozone average response shifts to positive, reflecting the classical change in chemical regime downwind of the sources. An illustration of this process can be seen in JJA in the vertical wind (wa) monsoon region where intense Sahelian BioNO sources decrease locally surface ozone but contribute to

- 525 a relative downwind surface ozone increase in northern Sahel/southern Sahara (dipole pattern on Figure 16f). For this situation, NO_x/VOC ratio decreases and chemical regime becomes more NO_x -limited (Delon et al., 2008; Stewart et al., 2008). In DJF, the surface ozone increase can reach approximately 4 ppb in the southern part of the domain, while in JJA, we observe increases of up to 3 ppb over the Sahel region and in eastern Africa. Over West Africa, this influence extends into the lower troposphere, where an average increase in ozone concentration of up to 4 ppb is noted (Figure 9c,f). This pattern aligns with findings by De-
- 530 lon et al. (2008), who observed that a moderate increase in NO_x concentrations leads to a sensitive increase in simulated ozone across all altitudes (0-15 km), characteristic of a NO_x -limited regime. Several studies have shown that throughout much of the troposphere in tropical regions, including Africa, O_3 formation is predominantly NO_x -limited, meaning it largely depends on the availability of NO_x (e.g., Li et al., 2021; Tadic et al., 2021).

Regarding comparison with the INDAAF ozone ground measurements, Figure 17 indicates that, unlike the improvements

- 535 observed for other species, the inclusion of BioNO emissions results in minimal to no enhancement in simulated ozone concentrations at the local level. However, a general reduction in bias, leading to better alignment between simulated and observed values, is observed for nearly all the sites during the period from June to August. In dry savannas the maximum negative bias is for instance reduced by 83.38 % in June over Banizoumbou (Table 7) while the ozone increase can also contribute to worsen the positive bias (+2.4 $\%$ in march). We observe an overall slight decrease in O_3 amplitude induced by BioNO over wet savannas
- 540 and equatorial forests from June to August, which tends to only very moderately improve simulations. Over tropical forest surface ozone concentrations are influenced by regional transport from burning areas, and local vertical exchanges between surface and lower troposphere. Model-measurement O_3 comparisons are not straightforward due to the presence of an important tree canopy on measurement sites (https://doi.org/10.25326/607; https://doi.org/10.25326/603) which can potentially affects both local dynamics and chemistry (e.g., Bryan and Steiner, 2013). Big leaf dry deposition schemes for ozone over tropical forests
- 545 can only crudely represent deposition processes, while extra chemical sinks within the canopy layer are simply not properly parametrized (Ganzeveld and Lelieveld, 2004; Sun et al., 2022). These deposition and chemical processes in the canopy should potentially be accounted for to better explain the discrepancy between the simulations and the in situ observations, and notably the fact that ozone observations are systematically much lower than model results in general (Figures 17 and 18).

 a Bias with BASE simulation

^b Bias with BIONO simulation

 $\,^c$ Reduction/Increase Bias by BioNO emissions

7 Conclusion

- 550 RegCM5 is the latest released version of the ICTP regional climate model designed to conduct high-resolution regional climate simulations. In the broader context of on-going programs targeting the regional nitrogen cycle over Africa, we more specifically use and assess the related atmospheric chemistry module (RegCM5-Chem), which has been substantially updated relative to previous versions. Doing so, we also conduct a specific study targeting the impact of BioNO emissions on regional chemistry over Africa. The sensitivity study consists of two experiments of 3-years coupled climate chemistry runs considering or not
- 555 BioNO emissions, simulated through an original parameterization based on ANN. These runs, and the impact of BioNO are

evaluated in the light of various data including satellite observations for climate, ground-based observations, reanalysis and alternative state of the art model outputs for key atmospheric compounds.

Obtained results show evidence that RegCM5-CHEM can capture the main features of the regional climate over the region, for instance in terms of seasonal and daily mean of temperature, precipitation and wind circulation relevant for regional 560 atmospheric chemistry and emission processes.

Simulated NO_2 , HNO_3 and O_3 fields show consistency with reanalysis and Geos-Chem simulations in terms of spatiotemporal distribution and gradients. Local comparison with surface concentrations measured over the six INDAAF sites indicates that the coupled chemistry-climate model can reproduce the seasonal cycle of all species over all sites. However, these comparisons strikingly outline an overall overestimation of simulated O_3 and, to a lesser extent, an underestimation of NO₂ and

- 565 HNO³ especially in the wet season over dry savannas stations. These large biases are present not only in RegCM simulations but also in reanalyses, and Geos-Chem outputs. In BASE simulation, the O_3 biases range from -2.64 to 34.2 ppb (dry savannas), -2.63 to 28.72 ppb (wet savannas) and 1.96 to 23.05 ppb (forests) while for $NO₂$, we obtained biases from -4.97 to -0.41 ppb (dry savannas), -0.84 to 1.59 ppb (wet savannas), -1.64 to 1.14 ppb (forests), and -1.73 to 0.02 ppb (dry savannas), -0.8 to 0.26 (wet savannas) and -0.54 to 0.05 ppb (forests) for $HNO₃$. These differences are attributed to potential deficiencies in chemical
- 570 emissions and mechanisms, deposition, boundary layer dynamics and transport from the upper layer which are particularly challenging to be reproduced in tropical regions. It is well known that regional models often struggle to accurately capture local-scale emissions and processes due to the coarse resolution (Valari and Menut, 2008; Wang et al., 2023) of their grids (in this case, 30 km x 30 km), which can lead to discrepancies when comparing with point measurements. Nevertheless, despite a substantial room for potential improvements, our conclusion is that RegCM5-Chem performance turns out to be consistent
- 575 with state of the art chemical reanalysis and chemistry transport model regarding regional photo-oxidant chemistry. When integrating BioNO emissions, we estimate seasonal averaged BioNO flux to be ranging from 0.02 to 7 mg m⁻² day⁻¹ and a total amount of nitrogen emitted from BioNO ranging from 0.01 to 4.4 TgN. month−¹ on the whole domain. The regional distribution of BioNO is essentially determined by the environmental predictors considered in the ANN based parameterization,
- with an important role of soil moisture variability. When taken into account, BioNO emissions lead to increased concentration 580 levels of surface NO_2 (ranging from 0.05 to 4 ppb) and HNO_3 (from 0.05 to 0.3). A decrease in surface ozone (until 2 ppb) likely in the Sahel) is also obtained as a likely result of predominant NO induced titration effect in the surface layer, while the O³ concentrations relatively increase in altitude and downwind from BioNO sources and towards the regional scale (up to 4 ppb).

In terms of model performance compared to INDAAF stations, the inclusion of BioNO emissions improves the representation 585 of O_3 , NO₂, and HNO₃ seasonal cycles and concentration magnitudes, particularly by reducing biases in some cases. However, in certain conditions, it can also lead to an increase in biases, highlighting the complex interactions at play across different regions and ecosystems.

Overall, our study points out an added value of including interactive BioNO emission representations, especially over the dry savannas of northern Sahel, where atmospheric nitrogen cycle and deposition inputs are particularly important for these 590 ecosystems where N content is low and sentitive to small variations. One limitation of the ANN approach is that it does not

account for limitations in the nitrogen pool ready to be emitted, which could be at play especially in dryer and unfertilized ecosystems. A deeper look at such limitations, using for example constraints from explicit soil nitrogen modules, is foreseen. Perspective to this work also include improving the representation of atmospheric chemistry processes important for the regional nitrogen budgets, such as LiNOx emissions and relevant heterogeneous chemistry processes (e.g. dust - HNO3), which 595 will have a significant impact on deposition processes, while keeping numerical efficiency required by climate scale simulation. Extended multi-decadal simulations aiming at investigating the impacts of regional climate variability and direct anthropogenic perturbations on the regional nitrogen cycle over Africa, which may bring a more thorough insight in future trends of these processes in Africa.

Code and data availability. The RegCM5 model code can be accessed at the web site: https://zenodo.org/record/7548172#.Y8gVV7TM-KUk.INDAAF 600 measurement network data is available at https://indaaf.obs-mip.fr. OMI TROPOMI-inferred ground-level NO² concentrations from 2010 to 2013 used in this study are available at https://doi.org/10.5281/zenodo.5424752. The GEOS-Chem model version 12.9.3 used in this work is available at https://doi.org/10.5281/zenodo.3974569. Data from these simulations can be freely shared upon request via email to fabien.solmon@aero.obs-mip.fr

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Competing interests. The authors declare that they have no financial or personal conflicts that could have affected the integrity of the work presented in this paper.

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Figure 6. Comparison of BASE and BioNO simulations of surface NO₂ against the CAMS reanalysis and the Geos-Chem model for DJF and JJA seasons. The INDAAF measurement values are overplotted and represented by small circles on the map.

Figure 7. Comparison of BASE and BioNO simulations of annual mean surface NO2 concentrations (ppb) against OMI and TROPOMIderived surface-level concentrations over 2010-2013, the CAMS reanalysis and the GEOS-Chem model. The INDAAF measurement values are overplotted and represented by small circles on the map.

Figure 8. Comparison of BASE and BIONO simulations of annual mean surface NO₂ concentrations (ppb) against OMI and OMI/Auraderived tropospheric NO₂ columns over 2010-2013, and associated Pearson spatial correlation.

Figure 9. DJF and JJA differences (BIONO - BASE) in NO₂ (a, d), HNO₃ (d, e) and O₃ (c, f) concentrations, on the transect 4-21°N averaged between 10°W-10°E. Units are in ppb.

Figure 10. Simulated monthly-averaged concentrations of NO₂ by BASE, BIONO runs and the CAMS reanalysis in comparison with INDAAF observation at representative remote sites.

Figure 11. Surface observed NO² concentrations (INDAAF) vs simulated with RegCM5. BioNO emissions are considered in the right panel.

Figure 12. Comparison of BASE and BioNO simulations of surface HNO₃ concentrations (in ppb) against the CAMS reanalysis and the model Geos-Chem for DJF and JJA season. The INDAAF measurement values are overplotted and represented by small circles on the map

Figure 13. Simulated monthly-averaged concentrations of HNO₃ by BASE, BIONO runs and the CAMS reanalysis in comparison with INDAAF observation at its representative remote sites.

Figure 14. Surface observed HNO₃ concentrations (INDAAF) vs simulated with RegCM5. BioNO emissions are considered in the right panel.

Figure 15. Comparison of BASE and BioNO simulations of surface O₃ concentrations (in ppb) against the CAMS reanalysis and the model Geos-Chem for DJF and JJA season. The INDAAF measurement values are overplotted and represented by small circles on the map.

Figure 16. DJA and JJA differences (BIONO - BASE) in surface NO₂ (left), HNO₃ (middle) and O₃ (right) concentrations. Units are in ppb.

Figure 17. Simulated monthly-averaged concentrations of O₃ by BASE, BIONO runs and the CAMS reanalysis in comparison with INDAAF observation at its representative remote sites.

Figure 18. Surface observed O₃ concentrations (INDAAF) vs simulated with RegCM5. BioNO emissions are considered in the right panel.