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## Long-term atmospheric inorganic nitrogen deposition in West African savanna over 16 year period (Lamto, Côte d'Ivoire)

Money Ossohou<sup>1</sup> , Corinne Galy-Lacaux<sup>2</sup>, Véronique Yoboué<sup>1</sup>, Marcellin Adon<sup>3</sup>, Claire Delon<sup>2</sup>, Eric Gardrat<sup>2</sup>, Ismaël Konaté<sup>4</sup>, Adja Ki<sup>1</sup> and Raphaël Zouzou<sup>4</sup><sup>1</sup> Laboratoire de Physique de l'Atmosphère et de Mécanique des Fluides, Université Félix Houphouët-Boigny, Abidjan, Côte d'Ivoire<sup>2</sup> Laboratoire d'Aérodynamique, Université de Toulouse, CNRS, UPS, Toulouse, France<sup>3</sup> Laboratoire des Sciences et Techniques de l'Environnement, Université Jean Lorougnon Guédé, Daloa, Ivory Coast<sup>4</sup> Station Géophysique de Lamto, 01 BP 398 Abidjan 01, Ivory CoastE-mail: [ossohoumoney@gmail.com](mailto:ossohoumoney@gmail.com) and [corinne.galy-lacaux@aero.obs-mip.fr](mailto:corinne.galy-lacaux@aero.obs-mip.fr)**Keywords:** sub Saharan Africa, wet savanna ecosystem, inorganic nitrogen, wet and dry depositions, trendsSupplementary material for this article is available [online](#)**Abstract**

We present a long term assessment trend of atmospheric inorganic nitrogen deposition in Sub Saharan Africa (2000–2015) using observational and model data. This work proposes a compilation of International Network to study Deposition and Atmospheric chemistry in Africa wet and dry nitrogen deposition fluxes collected at the wet savanna site of Lamto (Côte d'Ivoire). Total deposition calculation takes in account: (a) gaseous ( $\text{NO}_2$ ,  $\text{NH}_3$ ,  $\text{HNO}_3$ ) dry deposition fluxes estimated by considering nitrogen compound concentrations at the monthly scale and modeling average monthly dry deposition velocities, (b) particulate  $\text{PM}_{10}$  ( $\text{pNO}_3^-$ ,  $\text{pNH}_4^+$ ) dry deposition fluxes calculated using the same inferential method and (c) wet deposition (WD) fluxes including ions concentration measurements ( $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ) in rainwater combined with rainfall amount. We demonstrate for the first time the monthly and annual decreasing trends for dry nitrogen deposition of  $\text{N-NO}_2$  ( $-2.33\%$  month<sup>-1</sup> and  $-2.54\%$  yr<sup>-1</sup>) and  $\text{N-NH}_3$  ( $-2.55\%$  month<sup>-1</sup> and  $-2.89\%$  yr<sup>-1</sup>), but increasing trends for dry deposition of  $\text{N-HNO}_3$  ( $+1.00\%$  month<sup>-1</sup>) and WD of  $\text{N-NO}_3^-$  ( $+1.67\%$  month<sup>-1</sup> and  $+2.13\%$  yr<sup>-1</sup>) and  $\text{N-NH}_4^+$  ( $+2.33\%$  month<sup>-1</sup> and  $+3.36\%$  yr<sup>-1</sup>). Dry season  $\text{N-NO}_2$  deposition flux decline shows agreement with long term trend in  $\text{NO}_x$  emissions by biomass burning. Increasing trends for wet N deposition signals a gradual increase of nitrogen fertilizers use in agricultural practices in the Lamto area. Results also show no significant trend in total N deposition over the 16 year study period explained by the compensation of decreasing and increasing trends for dry and wet N deposition, respectively. However, at the annual scale, the mean total N deposition flux is estimated to  $10.3 \pm 1.2 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  over the 16 year period, indicating an increase of 8% compared to the period 2000–2007.

**1. Introduction**

Reactive nitrogen (Nr) compounds in the oxidized ( $\text{NO}_y$ ) and the reduced ( $\text{NH}_x$ ) forms play a central role in the chemistry of the atmosphere as well as in the functioning of marine, freshwater and terrestrial ecosystems (Dentener *et al* 2014). Nr enters the environment through a number of processes related to atmospheric emissions, transport and deposition. The deposition of atmospheric nitrogen (N) is not only one of the major processes for removing N pollutants in the air, but also one of the most important

steps in the biogeochemical cycle of nitrogen and an important input to the ecosystem (Wei 2018). Currently both climate change and N deposition from air pollution may provoke biodiversity loss (Bobbink *et al* 2010, Rao *et al* 2010). The availability of nitrogen has also profound implications in determining the amount of atmospheric carbon dioxide ( $\text{CO}_2$ ) naturally sequestered through biological productivity (Zhang *et al* 2020). Global Nr deposition assessments are available to determine the critical levels of nitrogen in atmospheric deposition for temperate ecosystems (Bobbink *et al* 2010, Lamarque *et al* 2013,

Vet *et al* 2014, Ackerman *et al* 2019), but *in situ* measurements recorded on the African continent remain scarce, scattered (Reis *et al* 2016) and during relatively short periods of time (no more than 10 years).

Land use, climate, and nitrogen deposition as global change pressures are the three major drivers of changes in every biome for Africa as for the global scale (Sala *et al* 2000). Nitrogen critical loads are poorly assessed in Africa, and ecosystems may be damaged by an excess of nitrogen at a shorter scale than initially thought (Bobbink *et al* 2010, Bauters *et al* 2018) especially for unmanaged ecosystems and plant communities (Bassirad 2015). However, a contrast exists between 'too much' and 'too little' N supplied regions over the world and this paradox is specifically valid at the scale of the African continent (Sutton *et al* 2013). The supply in nitrogen in Sub Saharan Africa (SSA) is insufficient (Zhang *et al* 2015) to ensure food security. As a matter of consequence, increasing nitrogen supply without increasing (or ideally with decreasing) emissions of pollutants is a societal challenge to be faced (Hickman *et al* 2015). In Africa it is expected that nitrogen deposition on ecosystems, particularly in wet form, will increase by 50% by 2100 (Lamarque *et al* 2013) due to a strong increase of the population, traffic, biomass burning and fertilizer use in agriculture. The region is submitted to an intense demographic pressure (i.e. 2.7% increase in local human populations between 1998 and 2007), and to some pressure on cultivated surfaces as well as a strong climatic variability (Hulme *et al* 2001, Diawara *et al* 2014). In order to consider nitrogen mitigation solutions for food security, it is essential where possible to analyze the seasonal dynamic of various fraction of nitrogen deposition and their trends in Africa.

Several research projects conducted in SSA to date aim to develop and implement strategies to ensure food self-sufficiency (through the use of N fertilizers) in response to its growing population, sometimes neglecting the environmental consequences. Additionally, Elrys *et al* (2020) have noted a lack of interest and attention by policy makers regarding the organic inputs approach so far. It is therefore imperative to address the issue of nitrogen in SSA in order to control its use in agriculture, livestock and food processing and to limit the environmental impacts in rural, urban and peri-urban areas. This requires *in situ* measurements of surface atmosphere exchanges of N compounds, including better understanding of N transformations in the atmosphere, and an up-scaling of these processes at the SSA region at the annual/pluriannual scales.

Within this general context, the International Network to study Deposition and Atmospheric chemistry in Africa (INDAAF) project, operational since 1995, has provided the first consistent *in situ* measurements of wet and dry N deposition fluxes in rural SSA ecosystems in order to improve the

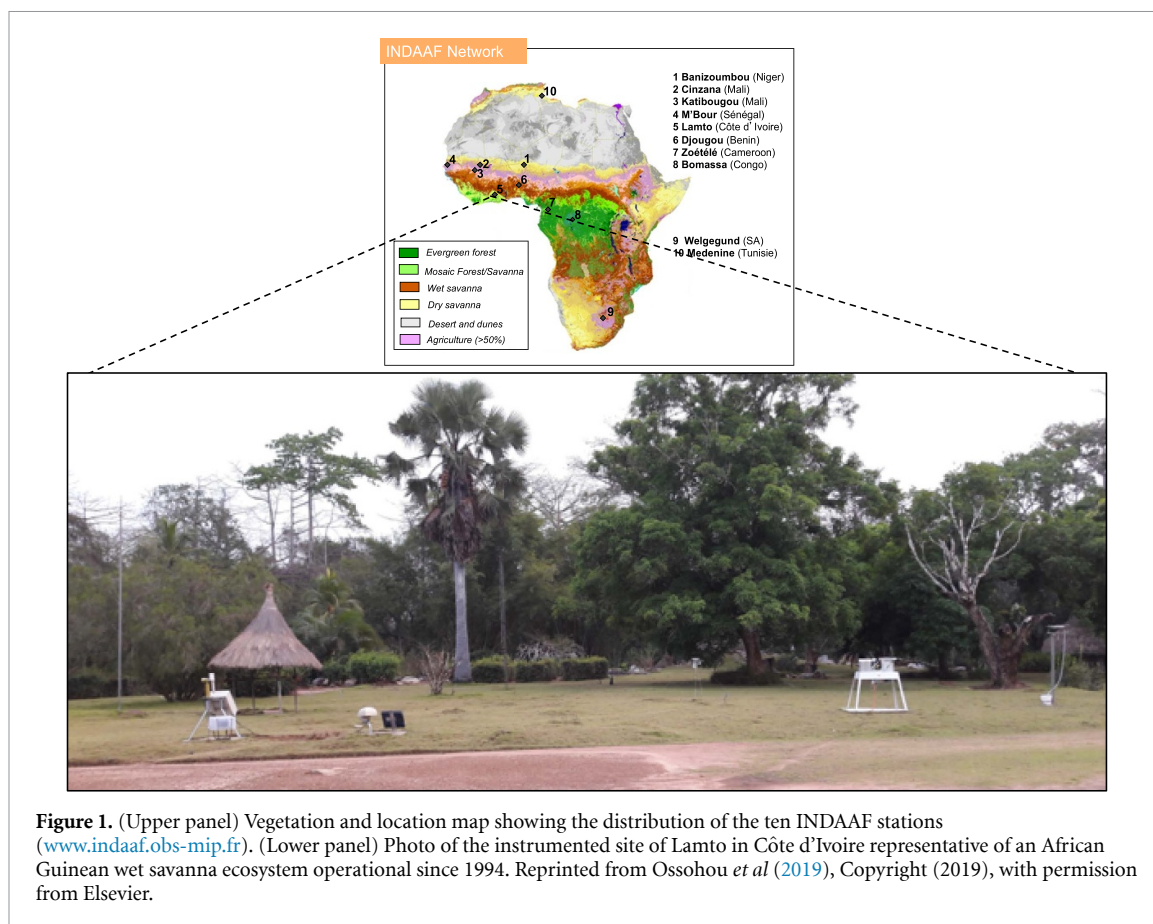
repository settings used in models. The INDAAF program is part of the European Aerosol Cloud and Trace Gases Research Infrastructure, is an official contributing network of the World Meteorological Organization-Global Atmospheric Watch (WMO-GAW) programme and also a component of the Deposition of Biogeochemically Important Trace Species activity labeled by International Global Atmospheric Chemistry. The INDAAF network measurement sites are representative of SSA ecosystems including dry savanna (Mali, Niger and Senegal), wet savanna (Côte d'Ivoire and Benin) and forest (Cameroon and Republic of Congo) (figure 1).

As part of the INDAAF programme, Galy-Lacaux and Delon (2014) have presented the contribution of major N compounds to establish the annual nitrogen atmospheric deposition budget regionally representative of a transect of dry savanna-wet savanna-forest in west and central Africa for the period 2000–2007. Nevertheless, this study did not take in account the long term trend assessment of atmospheric inorganic nitrogen deposition and its correlation with seasonal dynamic of various fractions of N deposition. Seasonal dynamics of different N deposition fractions would be helpful to understand the role of N supply and its interaction in African wet savannas. To address this gap, this study aims for the first time to assess long-term statistical trends of atmospheric and seasonal dynamic of N depositions in a regionally representative African wet savanna ecosystem, using a 16 year dataset representing the 2000–2015 period. This work presents a compilation of INDAAF wet and dry nitrogen deposition fluxes collected at the wet savanna site of Lamto (Côte d'Ivoire) over about two decades. The approach combines both experimental quality-controlled data performed by the INDAAF deposition program and modeling studies, especially developed for Lamto site to estimate the nitrogen atmospheric deposition budget. This work should contribute to the on-going International Nitrogen Assessment in the framework of the International Nitrogen Initiative.

## 2. Methods

### 2.1. Characterization of Lamto station

The Lamto geophysical and ecological station (06° 13' N, 05° 02' W) is a reserve located in the V-shaped Baoulé region (120 km North of Abidjan, Côte d'Ivoire). For the last four decades, seismological, ecological and climatological data from this natural reserve have been compiled. This site is used as a reference for a wet savanna ecosystem. The Lamto reserve covers approximately 2600 ha and is constituted essentially of the so called Guinean savanna along the Bandama River (Yoboué *et al* 2005). The vegetation is a mosaic of savannas with various tree densities, and gallery forests (Villocourt and Roose



**Figure 1.** (Upper panel) Vegetation and location map showing the distribution of the ten INDAAF stations ([www.indaaaf.obs-mip.fr](http://www.indaaaf.obs-mip.fr)). (Lower panel) Photo of the instrumented site of Lamto in Côte d'Ivoire representative of an African Guinean wet savanna ecosystem operational since 1994. Reprinted from Ossohou *et al* (2019), Copyright (2019), with permission from Elsevier.

1978, Diawara *et al* 2014). The soils are composed of granites and derived sands and classified as tropical ferruginous soils with a superficial gravelly horizon (Lata 1999), with particularly low total N concentrations and very low mineral N concentrations (Le Roux *et al* 2006), leading to a limitation of the efficiency of chemical fertilization (Assémien *et al* 2017, Srikanthasamy *et al* 2018). The people living in the vicinity of the station mainly practice agriculture, and to a lesser extent livestock farming.

From 1998 to 2015, total annual rainfall ranged from 991.9 mm in 2014 to 1586.2 mm in 2007 with an average rainfall of  $1240.9 \pm 133.5$  mm. The wet season extends from April to October and the dry season from November to March. The rainfall profile at Lamto is bimodal, with maximum rainfall in June and October (figure S1, which is available online at [stacks.iop.org/ERL/16/015004/mmedia](https://stacks.iop.org/ERL/16/015004/mmedia)). During the dry and wet seasons, the predominant air masses are harmattan and monsoon, respectively. Relative humidity at Lamto remains high most of the year, with a maximum of 82.8% in June and a minimum of 65.6% in January. Air temperatures are relatively constant throughout the year ( $27^\circ\text{C}$  on average). The Leaf Area Index (LAI) broadly follows the same pattern as rainfall, with high values from April to June and from August to November.

## 2.2. Wet and dry deposition calculations

### 2.2.1. Wet deposition (WD) calculation

The Lamto station is equipped with a semi-automatic rain collector specially designed for the INDAAF network (<http://indaaaf.obs-mip.fr>). A local operator collects water from each rainfall event in a Greiner tube (50 ml). Rain samples are stored just after collection in a deep freezer at  $-18^\circ\text{C}$ . The instrumentation and the collection protocol follow the WMO-GAW recommendations (report n°160). INDAAF experimental protocols have been widely used in Africa and are well described in Galy-Lacaux and Modi (1998), Sigha *et al* (2003), Yoboué *et al* (2005), Galy-Lacaux *et al* (2009), Laouali *et al* (2012), Akpo *et al* (2015). All rain samples were analyzed by ion chromatography at the Laboratoire d'Aérodologie (LAERO) to determine nitrate ( $\text{NO}_3^-$ ) and ammonium ( $\text{NH}_4^+$ ) concentrations. From 1994 to 2015, 1787 rain samples collected at Lamto were analyzed. International intercomparison studies are carried out bi-annually to ensure the quality of measurements. The LAERO is involved in the Quality Assurance/Quality Control WMO-GAW intercomparison programme since 1996 under the reference 700 106 ([www.qasac-america.org/](http://www.qasac-america.org/)). The analytical precision is 5% or better for mineral ions. Galy-Lacaux and Delon (2014) estimated a 25% uncertainty on WD fluxes.

Mean annual  $\text{NO}_3^-$  and  $\text{NH}_4^+$  volume weighed mean (VWM) concentrations in precipitation and mean annual rainfall were compiled to calculate the wet nitrogen deposition budget over the 22 years of measurement at Lamto. WD flux is the product of VWM concentrations by the average rainfall amount (Akpo *et al* 2015).

### 2.2.2. Dry deposition calculation

Dry deposition fluxes are estimated using the inferential method, which combines measured air concentrations and modeled deposition velocities. Atmospheric concentrations of  $\text{NO}_2$ ,  $\text{NH}_3$  and  $\text{HNO}_3$  were measured by passive samplers on a monthly basis (Osohou *et al* 2019). Particulate matter 10 ( $\text{PM}_{10} < 10 \mu\text{m}$ ) measurements were performed using the INDAAF aerosol sampler described by Ouafu-Leumbe *et al* (2018). Sampling of  $\text{PM}_{10}$  aerosol were collected on Teflon filters (Pall Zefluor, 47 mm diameter and  $0.5 \mu\text{m}$  of porosity). To determine the mineral water-soluble components, each sampled filters have been extracted in 10 ml extra pure water (resistivity  $\sim 18.2 \text{ M}\Omega$ ) by ultrasonic stirring for 15 min. Two Dionex ion chromatographs (ICS 1000 and ICS 1100) equipped with ion exchange column CG12A + CS12A and AG4A-SC + AS4A-SC were used to separate  $\text{NO}_3^-$  and  $\text{NH}_4^+$ , respectively.

$\text{NO}_2$  and  $\text{HNO}_3$  concentrations were sampled from 1998 to 2015, and  $\text{NH}_3$  from 2000 to 2015.  $\text{PM}_{10}$  were collected on a weekly basis from 2005 to 2015 with missing years for 2008–2010. The number of gas and aerosol samples collected at Lamto ranged from 181 to 205 and 273–319, respectively.

In this study, we used the averaged monthly dry deposition velocities ( $V_d$ ) of gases ( $\text{NO}_2$ ,  $\text{NH}_3$  and  $\text{HNO}_3$ ) simulated from 2002 to 2007 for the Lamto wet savanna by Adon *et al* (2013). In addition, to take into account the bidirectional exchange of  $\text{NH}_3$ , we used the  $\text{NH}_3$  averaged monthly canopy compensation point ( $X_{cp}$ ) estimated for Lamto (Adon *et al* 2013). Monthly dry deposition velocities ranges were  $0.21\text{--}0.33 \text{ cm s}^{-1}$  for  $\text{NO}_2$ ,  $0.35\text{--}0.60 \text{ cm s}^{-1}$  for  $\text{NH}_3$ , and  $0.93\text{--}1.30 \text{ cm s}^{-1}$  for  $\text{HNO}_3$  over the period 2002–2007 (Adon *et al* 2013). The average monthly  $\text{NH}_3$   $X_{cp}$  point ranged from  $0.36 \pm 0.01$  to  $1.15 \pm 0.30 \mu\text{g m}^{-3}$ . Note that for dry deposition of gases, the total rate of uncertainty applied for deposition fluxes is 70% for  $\text{NO}_2$ , 31% for  $\text{NH}_3$  and 38% for  $\text{HNO}_3$  (Galy-Lacaux and Delon 2014).

Several models for particle dry deposition velocity calculations are available in the literature (Zhang *et al* 2001, Kouznetsov and Sofiev 2012, Zhang and He 2014). Using a literature review, at the Lamto station, considering low wind speed less than  $1 \text{ m s}^{-1}$  (Roux 2006) and LAI about  $6 \text{ m}^2 \text{ m}^{-2}$  during the wet season, it was decided to consider a  $\text{PM}_{10}$  dry deposition velocity of  $0.25 \text{ cm s}^{-1}$  (Hirabayashi *et al* 2015).

## 2.3. Statistical analysis

Statistical tests were applied to determine if the long-term N depositions exhibit increasing or decreasing trends. The Mann–Kendall (MK) non-parametric test was used to detect trends for seasonal and annual N deposition fluxes (Mann 1945, Kendall 1975). Assuming that time series was formed by  $n$  observations  $(x_1, y_1), (x_2, y_2), (x_3, y_3), \dots (x_n, y_n)$  where  $x$  is time and  $y$  is the object variable, the Kendall S statistic can be computed from each data pair as  $S = P - N$ , where  $P$  is the number of  $y_i < y_j$  for all  $i < j$  and  $N$  is the number of  $y_i > y_j$  for all  $i < j$ . We only assume a trend if  $S$  is significantly different from zero. To infer monthly N deposition flux trends, the Seasonal MK test (SMK) (Hirsch *et al* 1982) was applied. SMK test combines the MK test computed on each month separately. The magnitudes of the MK and SMK trend tests are calculated using the method given by Sen's Slope (Sen 1968) and seasonal Kendall slope estimator which reports the median change in value versus time (slope) of all the possible pairs in the dataset including zero differences (Du *et al* 2014). The slope obtained was statistically different from zero at the 90% confidence level ( $p$ -value  $> 0.1$ ). These tests are 'distribution free' and not sensitive to outliers. All statistical analyses were performed using XLSTAT2018 and R version 3.5.0 (Core Team 2018).

## 3. Results and discussion

### 3.1. Nitrogen deposition fluxes

#### 3.1.1. Dry nitrogen deposition

##### 3.1.1.1. Gaseous dry deposition fluxes

Mean monthly dry deposition fluxes were  $\sim 0.0\text{--}0.1 \text{ kgN ha}^{-1} \text{ month}^{-1}$  for  $\text{NO}_2$  and  $\text{HNO}_3$ , and  $0.1\text{--}0.3 \text{ kgN ha}^{-1} \text{ month}^{-1}$  for  $\text{NH}_3$  (figure S2). The annual mean dry N gaseous deposition fluxes in  $\text{kgN ha}^{-1} \text{ yr}^{-1}$  in Lamto were  $2.0 \pm 0.6$  ( $\text{NH}_3$ )  $> 0.7 \pm 0.1$  ( $\text{HNO}_3$ )  $> 0.4 \pm 0.1$  ( $\text{NO}_2$ ).

$\text{NH}_3$  was the predominant N species and accounted for 67% of the total gaseous N deposition, combining the highest concentrations and deposition velocities among all compounds.  $\text{HNO}_3$  and  $\text{NO}_2$  represented 22% and 11%, respectively. The significant dry deposition fluxes of  $\text{HNO}_3$  relied on large dry deposition velocities 3–4 times higher than those of  $\text{NO}_2$ . A significant source of  $\text{NH}_3$  at Lamto is represented by savanna fires and domestic fuelwood burning (Delmas *et al* 1995). Agriculture also contributes undeniably to the deposition of large quantities of  $\text{NH}_3$ , by using significant amounts of N fertilizers and phytosanitary products to improve yields of rubber and cacao crops (Suzanne 2016), a space consuming agriculture. Osohou *et al* (2019) showed that biomass-burning was the most important source of ground-based  $\text{NO}_2$  at Lamto during the dry season. Increased  $\text{NO}_x$  oxidation rate under warm conditions

is an important source of HNO<sub>3</sub>. Thus, the chemical reaction between NO<sub>2</sub> and radical hydroxyl explained the large HNO<sub>3</sub> concentrations measured at Lamto (Osohou *et al* 2019).

The total mean annual N gaseous dry deposition flux was  $3.1 \pm 0.7$  kgN ha<sup>-1</sup> yr<sup>-1</sup> over the 16 years. This result is in the lower end of values ( $3.4$ – $4.6$  kgN ha<sup>-1</sup> yr<sup>-1</sup>) given by Galy-Lacaux and Delon (2014) for wet savannas over the period 2000–2007, probably due to the consideration of the bidirectional NH<sub>3</sub> exchange in our study.

### 3.1.1.2. Dry deposition of nitrogen from PM10

Monthly and seasonal PM10 dry deposition patterns clearly showed that pNO<sub>3</sub><sup>-</sup> and pNH<sub>4</sub><sup>+</sup> were enhanced in the dry season compared to the wet season (figure S3).

The mean annual particulate N deposition fluxes were  $0.1 \pm 0.1$  and  $0.1 \pm 0.0$  kgN ha<sup>-1</sup> yr<sup>-1</sup> for N-pNO<sub>3</sub><sup>-</sup> and N-pNH<sub>4</sub><sup>+</sup>, respectively. This result showed that pNO<sub>3</sub><sup>-</sup> dominates in the aerosols, and highlighted the importance of the biomass burning source at Lamto (Osohou *et al* 2019). The total (N-pNO<sub>3</sub><sup>-</sup> + N-pNH<sub>4</sub><sup>+</sup>) annual particulate N dry deposition flux over the 8 years of study was  $0.2 \pm 0.1$  kgN ha<sup>-1</sup> yr<sup>-1</sup>, with contributions of 57 and 43% for pNO<sub>3</sub><sup>-</sup> and pNH<sub>4</sub><sup>+</sup>, respectively. In addition, results showed that the dry season (November to March) contributed to 58% of the annual total dry N particulate deposition.

### 3.1.2. Wet nitrogen deposition

The monthly WD fluxes for rainy months ranged from 0.004 to 0.72 kgN ha<sup>-1</sup> month<sup>-1</sup> for N-NO<sub>3</sub><sup>-</sup> and from 0.004 to 1.66 kgN ha<sup>-1</sup> month<sup>-1</sup> for N-NH<sub>4</sub><sup>+</sup>. The largest fluxes for both N-NO<sub>3</sub><sup>-</sup> and N-NH<sub>4</sub><sup>+</sup> occurred in February, March, April and May (from the end of the dry season to the beginning of the wet season) (figure S4).

On a seasonal basis over the period 1994–2015, during the wet season, WD fluxes were  $1.2 \pm 0.2$ ,  $2.6 \pm 0.5$  kgN ha<sup>-1</sup> for N-NO<sub>3</sub><sup>-</sup>, N-NH<sub>4</sub><sup>+</sup> respectively with a total of  $3.7 \pm 0.7$  kgN ha<sup>-1</sup>. In the dry season, WD fluxes were  $0.8 \pm 0.2$  kgN ha<sup>-1</sup> for N-NO<sub>3</sub><sup>-</sup> and  $1.8 \pm 0.6$  kgN ha<sup>-1</sup> for N-NH<sub>4</sub><sup>+</sup>, with a total of  $2.7 \pm 0.7$  kgN ha<sup>-1</sup>. At the annual scale, WD fluxes were  $2.0 \pm 0.4$  kgN-NO<sub>3</sub><sup>-</sup> ha<sup>-1</sup> yr<sup>-1</sup> and  $4.3 \pm 1.0$  kgN-NH<sub>4</sub><sup>+</sup> ha<sup>-1</sup> yr<sup>-1</sup> giving a total N WD of  $6.3 \pm 1.3$  kgN ha<sup>-1</sup> yr<sup>-1</sup> over the 22 year period with an uncertainty within a 10% margin (Galy-Lacaux *et al* 2014). NO<sub>3</sub><sup>-</sup> (oxidized form) accounted for 32% of the annual dissolved inorganic nitrogen (DIN) WD and NH<sub>4</sub><sup>+</sup> (reduced form) for 68%. This result is higher than previous annual DIN WD fluxes estimated by Laouali *et al* (2012) and Galy-Lacaux and Delon (2014) ranging from 1.8 to 3.3 kgN ha<sup>-1</sup> yr<sup>-1</sup> in dry savannas, around 4.4 kgN ha<sup>-1</sup> yr<sup>-1</sup> in wet savannas, and 5.0 kgN ha<sup>-1</sup> yr<sup>-1</sup> in forest ecosystems.

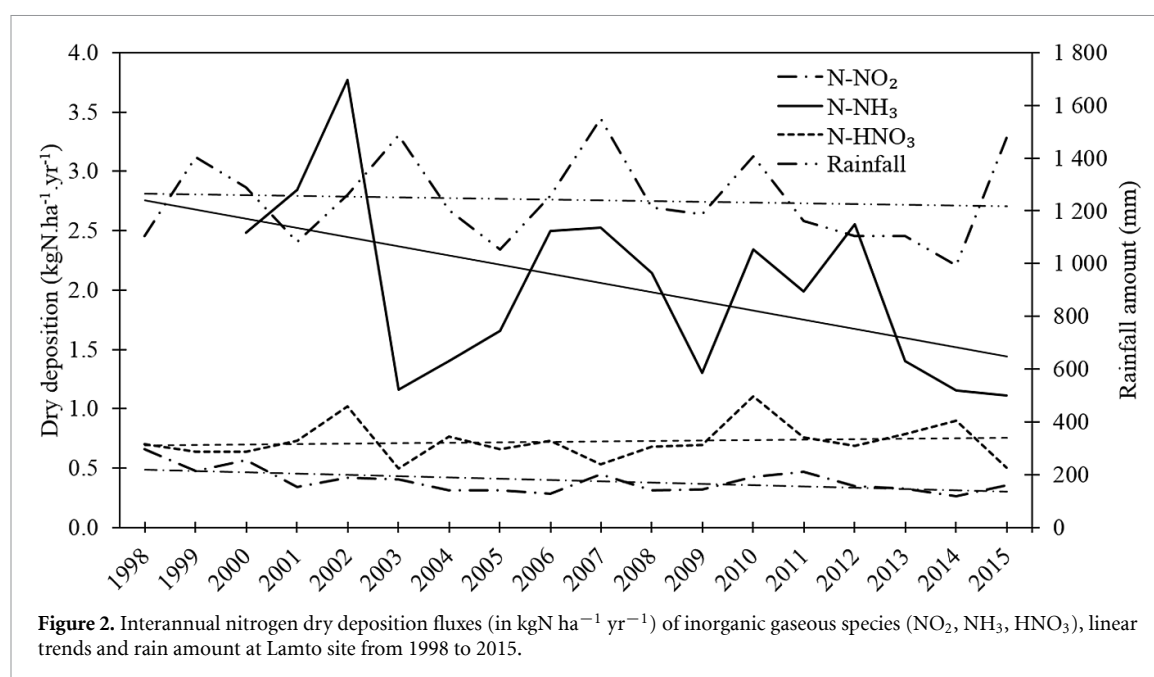
Gaseous NO<sub>2</sub>, particulate NO<sub>3</sub><sup>-</sup> on one hand, and gaseous NH<sub>3</sub>, particulate NH<sub>4</sub><sup>+</sup> on the other hand are significant contributors to the NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> content respectively in the precipitation. Nitrogenous gases and particles generally have higher concentration levels during the 5 months of the dry season (November–March) at Lamto (Adon *et al* 2010). These N compounds are further washed out by rainfall during the seven following months (April–October), with higher WD at the beginning of the wet season due to increasing rainfall amount at that period, and due to the strong influence of biomass burning source (Osohou *et al* 2019) and field preparation during the dry season (Suzanne 2016).

Results based on 10 years of study has shown that atmospheric concentrations NH<sub>3</sub> are higher year-round than that of NO<sub>2</sub> (Adon *et al* 2010) leading to generally acid rain in Lamto (Yoboué *et al* 2005). In addition, another study has highlighted the predominance of NH<sub>4</sub><sup>+</sup> in the fine fraction of aerosols, which is efficiently washed out by rainfall (Lovett and Lindberg 1986), coarse NO<sub>3</sub><sup>-</sup> particles undergo greater dry deposition (Pakkanen 1996). At the scale of the wet savanna of Lamto, NH<sub>4</sub><sup>+</sup> presents the highest WD due to the signature of a combination of sources: domestic animals, agro-systems, domestic combustion and biomass burning (Yoboué *et al* 2005). According to Serça *et al* (1998), the lower NO<sub>3</sub><sup>-</sup> deposition can be explained by the biogenic low NOx emissions in the Lamto station. In addition, decreasing trends in NOx emissions from biomass burning has been established at the regional scale of the studied site (Osohou *et al* 2019).

### 3.1.3. Total nitrogen deposition

To assess the total nitrogen deposition representative of the wet savanna of Lamto, monthly N dry and wet fluxes were averaged from 2000 to 2015. The monthly total atmospheric N deposition fluxes ranged from 0.5 (August) to 1.6 (March) kgN ha<sup>-1</sup> month<sup>-1</sup> (figure S5).

Total N deposition appeared to be the highest at the end of the dry season (February and March)—beginning of the wet season (April and May). Seasonal results are calculated considering the sum of dry months flux values and wet months flux values. The wet and dry season total N fluxes were  $5.4 \pm 0.9$  kgN ha<sup>-1</sup> and  $3.9 \pm 0.9$  kgN ha<sup>-1</sup> respectively. At the annual scale, the mean total N deposition flux was  $10.3 \pm 1.2$  kgN ha<sup>-1</sup> yr<sup>-1</sup> over the 16 year period. This result shows an increase of 8% compared to the period 2000–2007 studied by Galy-Lacaux *et al* (2014), and is close to N compound emissions calculated in Lamto for the period 2002–2007. Indeed, Delon *et al* (2012) showed that N emissions in Lamto reach  $10.1 \pm 4.1$  kgN ha<sup>-1</sup> yr<sup>-1</sup>, with the largest contribution due to N compound emissions from biomass burning.



**Figure 2.** Interannual nitrogen dry deposition fluxes (in  $\text{kgN ha}^{-1} \text{yr}^{-1}$ ) of inorganic gaseous species ( $\text{NO}_2$ ,  $\text{NH}_3$ ,  $\text{HNO}_3$ ), linear trends and rain amount at Lamto site from 1998 to 2015.

The total annual deposition of N at the wet savanna site of Lamto highlighted the importance of wet nitrogen deposition processes compared to dry deposition with a contribution of 68% and 32% respectively (figure S6). The contribution of particulate N species to the total N deposition was negligible ( $\sim 2\%$ ). Annual mean reduced ( $\text{NH}_3$ ,  $\text{pNH}_4^+$ ,  $\text{NH}_4^+$ ) and oxidized ( $\text{NO}_2$ ,  $\text{HNO}_3$ ,  $\text{pNH}_4^+$ ,  $\text{NO}_3^-$ ) contributions to the N deposition flux were  $7.0 \pm 1.0 \text{ kgN ha}^{-1} \text{yr}^{-1}$  and  $3.3 \pm 0.3 \text{ kgN ha}^{-1} \text{yr}^{-1}$ , respectively. In the total deposition budget, reduced N is predominant with 48% of  $\text{NH}_4^+$  WD and 19% of ammonia dry deposition.

These results need to be considered as a lower value of the total atmospheric N deposition since atmospheric deposition budgets based solely on inorganic nitrogen are likely to be underestimating by about a third (Cornell *et al* 2003).

### 3.2. Nitrogen deposition trends

#### 3.2.1. Dry nitrogen deposition trends

Monthly, seasonal and annual trends of gaseous dry N deposition fluxes were calculated over the 1998–2015 period for N- $\text{NO}_2$  and N- $\text{HNO}_3$ , and from 2000 to 2015 for N- $\text{NH}_3$  and total N. Trends of particulate dry N deposition fluxes were not calculated because of shorter time series (8 years of measurements only). Generally, a significant decrease of the gaseous dry N deposition fluxes of  $\text{NO}_2$ ,  $\text{NH}_3$  and consequently of the total N was observed (figure 2). To the opposite, a slight increase in the dry N deposition flux of  $\text{HNO}_3$  was found.

The largest decrease was obtained for monthly ammonia deposition with  $-0.09 \text{ kgN ha}^{-1} \text{month}^{-1}$  representing  $-2.55\% \text{ month}^{-1}$  (table 1). This result leads to an annual trend of  $-2.89\%$ . Monthly

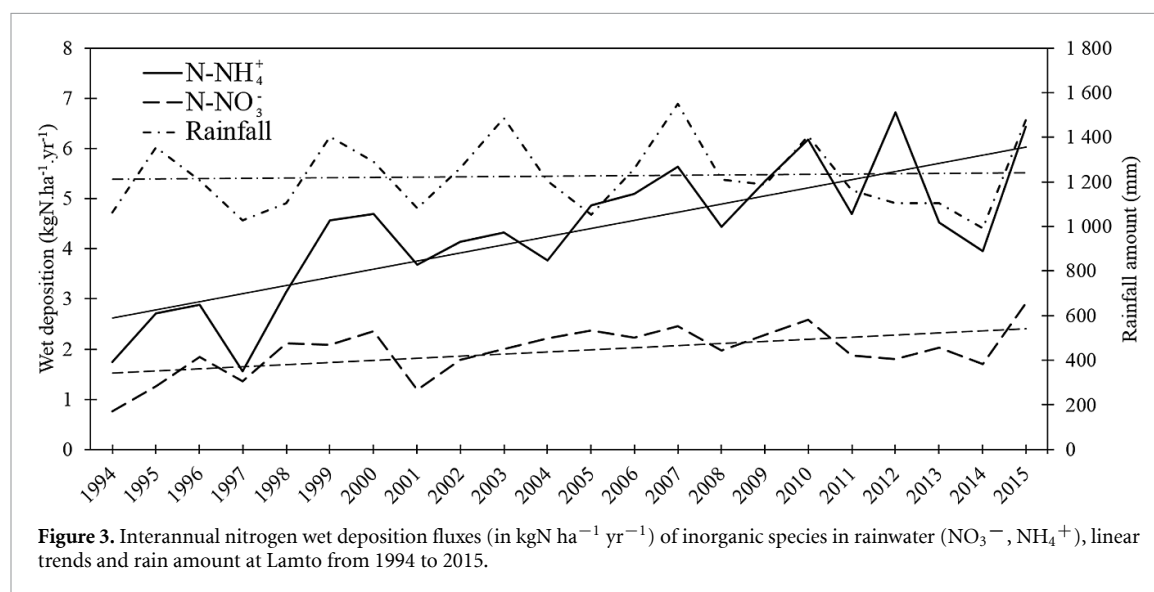
and seasonal  $\text{NO}_2$  decreasing trends are also observed with a wet season decreasing trend 1.5 higher than the dry season trend, mainly due to higher  $\text{NO}_2$  deposition velocities in the wet season. Decreasing trends in dry N- $\text{NO}_2$  could be explained by the annual and dry season decreasing  $\text{NO}_x$  emission trends calculated by Osohou *et al* (2019) to be  $-4.95\% \text{ yr}^{-1}$  and  $-4.90\% \text{ yr}^{-1}$ , respectively. In addition, a Spearman correlation coefficient of 0.6 (99.9% significant) between  $\text{NO}_x$  emissions and dry season N- $\text{NO}_2$  deposition fluxes at Lamto was obtained. Ichoku *et al* (2016) showed that during the burning season over the period 2006–2014, the biomass combustion activity decreased ( $2\text{--}7\% \text{ yr}^{-1}$ ) in different parts of the SSA, in particular in savanna regions. Fire distribution showed appreciable coincidence with land-cover change, cropland increased at an estimated rate of  $0.28\% \text{ yr}^{-1}$  of the total Northern SSA land area over the period 2003–2013, with most of it ( $0.18\% \text{ yr}^{-1}$ ) coming from savanna.

It is important to note that increasing trends in N- $\text{NO}_2$  and N- $\text{NH}_3$  coincide with decreasing trends in  $\text{NO}_3\text{-N}$  and  $\text{NH}_4^+$  in rainfall as mentioned in the following section. On the other hand, we obtained no significant trends for  $\text{NH}_3$  emissions from combustion sources. Monthly decreasing N- $\text{NH}_3$  deposition could be attributed to the decreasing of  $\text{NH}_3$  concentrations, with Pearson correlation coefficient of 0.99 (99.9% significant). Reduction of gaseous dry N deposition by rainfall is confirmed by correlation coefficients of  $-0.19$  and  $-0.15$  (95% significant) between monthly rainfall depths and atmospheric concentrations of  $\text{NO}_2$  (1998–2015) and  $\text{NH}_3$  (2000–2015), respectively. The increasing trend of N- $\text{HNO}_3$  deposition could be explained by a higher rate of  $\text{NO}_2$  to  $\text{HNO}_3$  transformation (Osohou *et al* 2019).



**Table 1.** Monthly, seasonal and annual trends of dry deposition fluxes of N-NO<sub>2</sub> (1998–2015), N-HNO<sub>3</sub> (1998–2015), N-NH<sub>3</sub> (2000–2015) and total dry deposition (2000–2015) in Lamto. Units: kgN ha<sup>-1</sup> month<sup>-1</sup>; % month<sup>-1</sup> (in bracket) for monthly, kgN ha<sup>-1</sup>; % (in bracket) for seasonal, and kgN ha<sup>-1</sup> yr<sup>-1</sup>; % yr<sup>-1</sup> (in bracket) for annual data. \*\*\*, \*\* and \* represent significance levels at 99%, 95% and 90%, respectively. ns = non significant.

	N-NO <sub>2</sub>	N-NH <sub>3</sub>	N-HNO <sub>3</sub>	N-NO <sub>2</sub> + N-NH <sub>3</sub> + N-HNO <sub>3</sub>
Monthly	-0.001*** (-2.33)	-0.090*** (-2.55)	+0.001* (+1.00)	-0.007** (-2.75)
Dry season	-0.004** (-2.05)	ns	ns	ns
Wet season	-0.007* (-3.40)	ns	ns	ns
Annual	-0.010* (-2.54)	-0.058* (-2.89)	ns	ns



**Figure 3.** Interannual nitrogen wet deposition fluxes (in kgN ha<sup>-1</sup> yr<sup>-1</sup>) of inorganic species in rainwater (NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>), linear trends and rain amount at Lamto from 1994 to 2015.

### 3.2.2. Wet nitrogen deposition trends

Monthly, seasonal and annual trends of N-NO<sub>3</sub><sup>-</sup>, N-NH<sub>4</sub><sup>+</sup> WD fluxes (figure 3) and total WD were calculated at Lamto station over the period 1994–2015. In general, we observe an increasing trend at all time scales of wet N deposition with a higher rate for N-NH<sub>4</sub><sup>+</sup> compared to N-NO<sub>3</sub><sup>-</sup> (table 2).

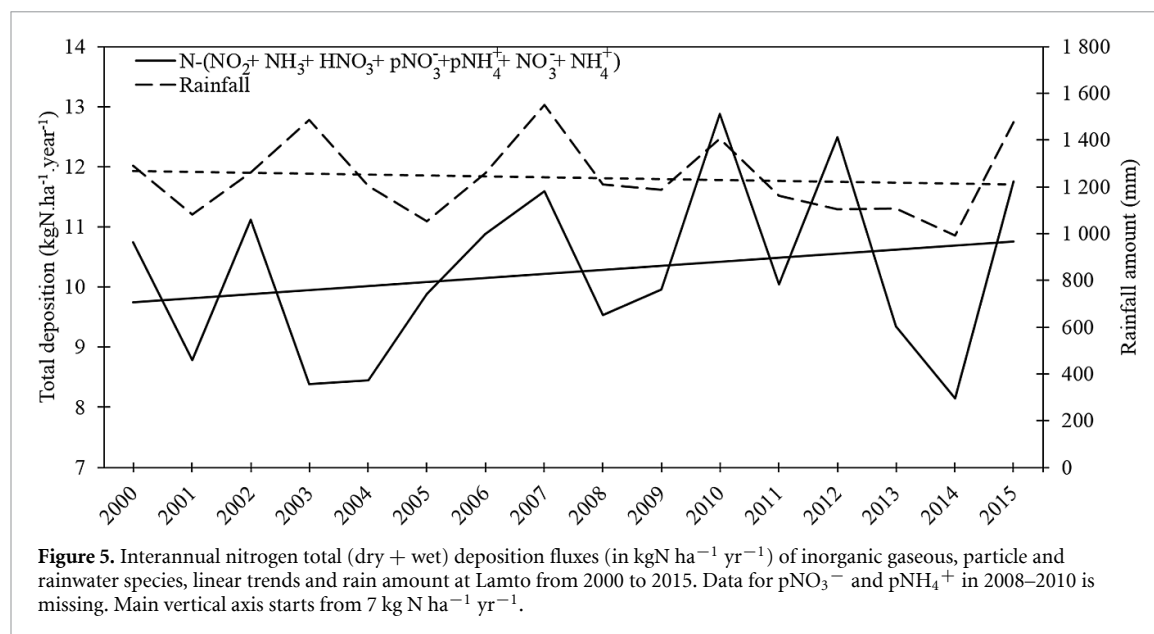
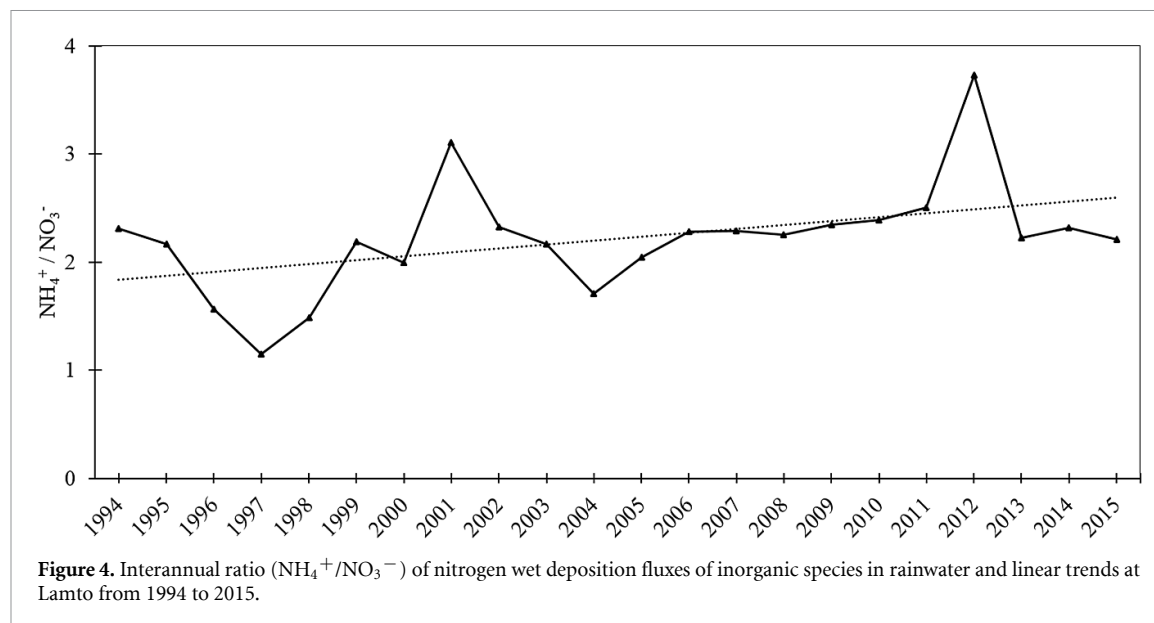
Statistical trend analysis on rainfall depths at Lamto from 1994 to 2015 showed increasing trends in monthly, dry season and annual rainfall (table 2). We assume that NO<sub>2</sub> and NH<sub>3</sub> in the gas phase are increasingly scavenged by rainfall, involving a decrease in dry N-NO<sub>2</sub> and N-NH<sub>3</sub> deposition and consequently an increase in wet N-NO<sub>3</sub><sup>-</sup>, N-NH<sub>4</sub><sup>+</sup> deposition. Increasing trends in wet N deposition in Lamto are due to increasing precipitation amounts, with correlation coefficients of 0.63 and 0.57 (99.9% significant) between rainfall amounts and monthly wet N-NO<sub>3</sub><sup>-</sup> and N-NH<sub>4</sub><sup>+</sup> deposition fluxes, respectively. Indeed, large amounts of ammonium sulfate and ammonium nitrate aerosols are first formed in the dry season and then effectively washed out by rains during the wet season.

Furthermore, we observed annual N-NH<sub>4</sub><sup>+</sup>/N-NO<sub>3</sub><sup>-</sup> ratios greater than 1 over the 22 years of study, ranging from 1.1 (in 1997) to 3.7 (in 2012) with an average of 2.2 ± 0.3 (figure 4). This result indicates

that N content in WD at Lamto is highly related to agricultural activities (Xu *et al* 2015). MK test applied to the annual N-NH<sub>4</sub><sup>+</sup>/N-NO<sub>3</sub><sup>-</sup> ratio showed significant increasing trends of +2.17% (*p*-value < 0.05) in the dry season and +1.09% (*p*-value < 0.1) for annual means. These trends clearly show that agricultural activities have been increasing in the Lamto area from 1994 to 2015, as shown by the increase of agricultural land percentage in Côte d'Ivoire from 62% to 65% between 1994 and 2015 (source: <https://donnees.banquemondiale.org/indicateur/AG.LND.AGRI.ZS>) and cropland increased in SSA savannas (Ichoku *et al* 2016). The extension of arable land, charcoal production and various agricultural practices lead to an increased use of pesticides and nitrogenous plant protection products such as imidacloprid used by rubber tree growers at the regional scale around the Lamto station (Dosso and Kone 2016). According to the International Fertilizer Association, fertilizer consumption in Africa has increased by 70% between 2009 and 2019. However, annual Africa's fertilizer use averages only 8 kg ha<sup>-1</sup>, i.e. only 10% of the world's average. The trend of fertilizers use showed an increase in SSA (excluding South Africa) from 6 kg ha<sup>-1</sup> in 2000 to 16 kg ha<sup>-1</sup> in 2017 towards 20 kg ha<sup>-1</sup> in 2020 (<http://AfricaFertilizer.org>).

**Table 2.** Monthly, seasonal and annual trends of rainfall (in mm), wet deposition fluxes of  $\text{N-NO}_3^-$ ,  $\text{N-NH}_4^+$ , and total deposition at Lamto over the period 1994–2015. Units:  $\text{kgN ha}^{-1} \text{ month}^{-1}$ ; %  $\text{month}^{-1}$  (in bracket) for monthly,  $\text{kgN ha}^{-1}$ ; % (in bracket) for seasonal, and  $\text{kgN ha}^{-1} \text{ yr}^{-1}$ ; %  $\text{yr}^{-1}$  (in bracket) for annual data. \*\*\*, \*\* and \* represent significance levels at 99%, 95% and 90%, respectively. ns : non significant

	Rainfall	$\text{N-NO}_3^-$	$\text{N-NH}_4^+$	$\text{N-NO}_3^- + \text{N-NH}_4^+$
Monthly	+1.487** (+1.53)	+0.002*** (+1.67)	+0.007*** (+2.33)	+0.01*** (+2.38)
Dry season	+2.084** (+3.31)	+0.014** (+2.43)	+0.060*** (+4.22)	+0.076*** (+3.80)
Wet season	ns	+0.016* (+1.89)	+0.052** (+2.65)	+0.063** (+2.27)
Annually	+0.918** (+0.94)	+0.030** (+2.13)	+0.112*** (+3.36)	+0.153*** (+3.25)



### 3.2.3. Total nitrogen deposition trends

MK and SMK statistical tests were applied to the reduced ( $\text{NH}_3 + \text{NH}_4^+$ ), oxidized ( $\text{NO}_2 + \text{HNO}_3 + \text{NO}_3^-$ ) and total (reduced + oxidized) nitrogen deposition flux times series over the period 2000–2015. Results showed a low interannual variability with a variation coefficient of 13%

(figure 5). Observed decreasing trends in gases  $\text{N-NO}_2$  and  $\text{N-NH}_3$  dry deposition seem to compensate the observed increasing trend of wet  $\text{N-NO}_3^-$  and  $\text{N-NH}_4^+$  and dry  $\text{N-HNO}_3$  deposition. As a result, there is no significant trend in total N deposition over the 16 year study period at the Lamto site representative of African wet savanna. However, oxidized

N ( $\text{NO}_2 + \text{HNO}_3 + \text{NO}_3^-$ ) deposition showed an increasing trend in dry season by  $0.03 \text{ kgN ha}^{-1}$  or 1.81% significant at 95%.

#### 4. Conclusion

This study provides for the first time a robust assessment of long-term N deposition trends on a regionally representative West African wet savanna ecosystem over a 16 year period. The work relies on a unique long-term dataset on precipitation, particulate and gaseous N contents at Lamto station (Côte d'Ivoire) using reliable and validated approaches.

$\text{NH}_3$  is the most important contributor to dry deposition, followed by  $\text{HNO}_3$  and  $\text{NO}_2$ , respectively. Mean monthly N deposition from particles shows a marked seasonal cycle characterized by the highest values during the dry season and the lowest values in the wet season with the seasonality of N-p $\text{NO}_3^-$  more obvious than that of N-p $\text{NH}_4^+$ . WD is dominated by N- $\text{NH}_4^+$ , with the maximum WD obtained at the end of the dry season-beginning of the wet season.

The total inorganic N deposition budget at the wet savanna site of Lamto station over the period 2000–2015 is  $10.3 \pm 1.2 \text{ kgN ha}^{-1} \text{ yr}^{-1}$ . WD remains the largest contributor (68%) compared to dry particulate and gas N deposition (32%) to the total N deposition fluxes. This study gives unprecedented insights in the seasonality of N WD inputs. We emphasize that nitrogen deposition in Lamto could be seasonally and annually influenced by the variation in emission source strength, mainly  $\text{NO}_x$  and  $\text{NH}_3$  emissions from biomass burning and agricultural sources, and rainfall amount. Total N deposition based on this study (2000–2015) has increased by 8% compared to a previous estimate proposed by Galy-Lacaux and Delon (2014) based on a 2000–2007 period of analysis.

MK and SMK statistical tests indicated decreasing trends for dry N deposition (N- $\text{NO}_2$  and N- $\text{NH}_3$ ), increasing trends for N- $\text{HNO}_3$  dry deposition flux and WD of N- $\text{NO}_3^-$  and N- $\text{NH}_4^+$ . Annually, no significant trend in total dry gaseous N deposition appeared. The use of gaseous and precipitation N contents allowed to conclude that there is no significant trend in total N deposition over the 16 year study period. This evaluation should be taken with caution as dissolved organic nitrogen fraction is missing. Nevertheless, increasing trends in wet DIN compounds demonstrated that SSA is already under the influence of demographic pressure and economic development leading to an increasing loss of reactive N into the environment.

Indeed, until now, agro-systems in West and Central Africa have been managed using very few quantities of synthetic N fertilizers ( $8 \text{ kgN ha}^{-1} \text{ yr}^{-1}$ ) and our study highlights that current N atmospheric deposition is equivalent to this fertilization rate. The wet savanna zone covers  $0.5 \times 10^6 \text{ km}^2$  in

West Africa and is characterized by very low soil N content limiting primary production. Africa, with insufficient access to nutrients, should be a priority for future global assessment on the nitrogen cycle and the improvement of nutrient use efficiency. Our work on N deposition at Lamto site, used as a reference for a wet savanna ecosystem in the INDAAF network, represents an important contribution for 'low N input regions' that should be taken into consideration in future integrated nitrogen management systems for Africa.

#### Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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#### ORCID iD

Money Ossohou  <https://orcid.org/0000-0001-5891-2182>

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