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Corinne Galy-Lacaux, Claire Delon. Nitrogen emission and deposition budget in West and Central Africa. Environmental Research Letters, 2014, 9 (12), pp.125002. 10.1088/1748-9326/9/12/125002. hal-02354426

### HAL Id: hal-02354426 https://hal.science/hal-02354426

Submitted on 1 Dec 2022

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To cite this article: C Galy-Lacaux and C Delon 2014 Environ. Res. Lett. 9 125002

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# Nitrogen emission and deposition budget in West and Central Africa

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Received 27 May 2014, revised 15 October 2014 Accepted for publication 27 October 2014 Published 2 December 2014

#### Abstract

Atmospheric nitrogen depends on land surface exchanges of nitrogen compounds. In Sub Saharan Africa, deposition and emission fluxes of nitrogen compounds are poorly quantified, and are likely to increase in the near future due to land use change and anthropogenic pressure. This work proposes an estimate of atmospheric N compounds budget in West and Central Africa, along an ecosystem transect, from dry savanna to wet savanna and forest, for years 2000–2007. The budget may be considered as a one point in time budget, to be included in long term studies as one of the first reference point for Sub Saharan Africa. Gaseous dry deposition fluxes are estimated by considering N compounds concentrations measured in the frame of the IDAF network (IGAC/DEBITS/AFrica) at the monthly scale and modeling of deposition velocities at the IDAF sites, taking into account the bi directional exchange of ammonia. Particulate dry deposition fluxes are calculated using the same inferential method. Wet deposition fluxes are calculated from measurements of ammonium and nitrate chemical content in precipitations at the IDAF sites combined with the annual rainfall amount. In terms of emission, biogenic NO emissions are simulated at each IDAF site with a surface model coupled to an emission module elaborated from an artificial neural network equation. Ammonia emissions from volatilization are calculated from literature data on livestock quantity in each country and N content in manure.  $NO_x$  and  $NH_3$  emission from biomass burning and domestic fires are estimated from satellite data and emission factors. The total budget shows that emission sources of nitrogen compounds are in equilibrium with deposition fluxes in dry and wet savannas, with respectively  $7.40 (\pm 1.90)$ deposited and 9.01 ( $\pm$ 3.44) kgN ha<sup>-1</sup> yr<sup>-1</sup> emitted in dry savanna, 8.38 ( $\pm$ 2.04) kgN ha<sup>-1</sup> yr<sup>-1</sup> deposited and 9.60 ( $\pm$ 0.69) kgN ha<sup>-1</sup> yr<sup>-1</sup> emitted in wet savanna. In forested ecosystems, the total budget is dominated by wet plus dry deposition processes  $(14.75 \pm 2.36 \text{ kgN ha}^{-1} \text{ yr}^{-1})$ , compared to emissions processes  $(8.54 \pm 0.50 \text{ kgN ha}^{-1} \text{ yr}^{-1})$ .

Keywords: atmospheric deposition budget, African ecosystems, nitrogen, wet and dry deposition processes, emission budget

#### Introduction

Reactive nitrogen  $(N_r)$  compounds in the oxidized form  $(NO_x)$ and the reduced form (NH<sub>x</sub>) play an important role in the chemistry of the atmosphere as well as the functioning of aquatic and terrestrial ecosystems (Vitousek et al 1997). Nr inputs to ecosystems have considerably increased since pre industrial times and are still accelerating (Galloway et al 2004, 2008). Increased Nr supports the food and fuel needs of a growing human population, but it affects environmental sustainability, climate, the chemistry of the atmosphere, and the composition and function of terrestrial and aquatic ecosystems (Galloway et al 2008). Considering these multiple impacts of N<sub>r</sub>, evidences are available for temperate ecosystems on critical levels of Nr for atmospheric deposition and pollutant air concentrations above which significant environmental degradation can be expected. Regional Nr assessment are available for the European Union (Sutton et al 2011), North America (Sobota et al 2013) and China (Ti

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*et al* 2012) including sometimes synthesis on the relationships for both water and air  $N_r$  pollution to human health.

However, major uncertainties remain for developing world. Africa remains a region with too little Nr. Insufficient N<sub>r</sub> leads to food insecurity in developing regions of the world, specifically in Sub Saharan Africa (SSA) (Godfray et al 2010). SSA represents at the global scale one of the best case study of the ongoing challenges created by 'too little N', and the on going African Green Revolution that emphasizes overcoming soil nutrient depletion through provision of more nutrients through subsidized fertilizers, combined with improved cropping practices and a better water management (Palm et al 1997, Sanchez et al 2007, 2010, Vitousek et al 2009). If in the 'industrialized world', reducing  $N_r$ creation and its harmful impacts is of critical importance, the initial challenge in SSA remains to increase Nr to produce more food while minimizing nutrient loss and its subsequent environmental damages (Sutton et al 2011).

The work presented in this paper starts in the context of the regional African assessment in the framework of the International Nitrogen Initiative. For SSA, it appears clearly that a better understanding of  $N_r$  inputs and sources is critical to improve the balance between their positive and negative impacts. Except the synthesis of  $N_r$  in Africa published by Robertson and Rosswall in 1986 for West Africa, there is no other global assessment for Africa. This synthesis underlines among other things the need to better estimate the atmospheric nitrogen fluxes according to land use category through direct measurements of N emissions and deposition fluxes.

To contribute to the future global  $N_r$  assessment for Africa, we propose in this work an estimate of the nitrogen atmospheric budget. This budget relies on measurements and/ or calculations of N emissions and deposition fluxes regionally representative of the main African ecosystems defined according to a transect: dry savannas-wet savannas-forests.

Emission inventories of the hemispheric transport of atmospheric pollutants (HTAP) models intercomparison exercise (Vet et al 2014) indicate a global total emissions of NH<sub>3</sub>-N in 2001 to be 58.5 Tg N. Asia accounted for the highest emissions with 39.4% of the global total, with Africa and South America being the next highest with 12.5% and 10.4%, respectively. Global total emissions of  $NO_r$ -N in 2001 were estimated at 46.6 Tg N with Asia accounting for 26.8%, and North America and Africa accounting for 18.7% and 16.7%, respectively. HTAP 2001 simulations predicts that global total deposition of N is around 106.3 Tg N. Total deposition of N to the non-coastal continental areas was highest in Asia (22.2% of global N deposition) followed by Africa (10.0%) and North America (8.0%) (Vet et al 2014). Future projections in Asia and in SSA suggest that these regions can expect substantial increases in reactive N deposition over coming decades (Dentener et al 2006, 2014, Lamarque et al 2013, Sutton et al 2014). Moreover, a recent inventory developed for anthropogenic african combustion (excluding biomass burning) predicts increase higher than 30% for NO<sub>x</sub> emission for the african continent compared to other projections given by RCP projections for the future (Liousse *et al* 2014). In addition to combustion, projected increases of N emissions from agricultural soils in Africa (e.g., from the increasing use of synthetic fertilizers), over tropical forests (e.g., from biomass burning), and in rapidly developing locations around the globe emphasize the need for long-term observations of wet and dry deposition to understand the future impacts.

In this context, the international program Deposition of Biogeochemically Important Trace Species (DEBITS) started in 1990 as part of the IGAC/IGBP (International Global Atmospheric Chemistry/International Geosphere-Biosphere Programme) core project. The aim of the project is to assess the wet- and dry-atmospheric deposition in tropical regions (Pienaar et al 2005. For tropical Africa, the IDAF (IGAC/ DEBITS AFRICA) project started in 1994, in partnership with INSU (Institut National des Sciences de l'Univers, France) and the CNRS (Centre National de la Recherche Scientifique, France) as part of the Environmental Research Observatory (ORE, France) networks. IDAF has the mission of establishing long-term measuring network to study the atmospheric composition and wet- and dry-atmospheric processes. The main objectives of IDAF are to measure wet- and dry-deposition fluxes and to identify the relative contribution of natural and anthropogenic sources of nitrogen. As such, IDAF activity is based on high quality measurements of atmospheric chemical data (gaseous, precipitation and aerosols chemical composition) on the basis of a multi-year monitoring. This project implemented ten monitoring sites distributed in the major African ecosystems over West and Central Africa and South Africa: dry savanna (Niger, Mali, South Africa), wet savanna (Côte d'Ivoire and Benin) and equatorial forest (Cameroon, Congo) (figure 1).

We present in this paper an atmospheric nitrogen budget including nitrogen emissions and deposition regionally representative of major African ecosystems in west and central Africa. In this study, we propose a compilation of IDAF wet and dry nitrogen deposition fluxes for the period 2000-2007 for different IDAF sites representative of a transect of ecosystem. In addition, atmospheric nitrogen emissions have been estimated to be representative of specific climate and vegetation around each studied ecosystem and will be compared to nitrogen deposition fluxes. The same approach has been previously applied in the Sahelian region for the year 2006 by Delon et al (2010), and for years 2002 -2007 in dry and wet savannas (Delon et al 2012), but never in forested ecosystems. This study in west and central Africa allows giving an original atmospheric nitrogen budget at the regional scale. The approach combines both unique experimental data based on the IDAF deposition program and modeling studies, especially developed for SSA to estimate nitrogen atmospheric exchanges.

This regional N emission deposition budget should give the present status at the scale of the main African ecosystems and should help to quantify the processes that may contribute to the changing levels of N deposition. In addition to changes in  $NO_x$ -N and NH<sub>3</sub>-N emissions, the annual and seasonal rate and distribution of precipitation are also important to understand changes in N deposition over Africa. In dry areas,

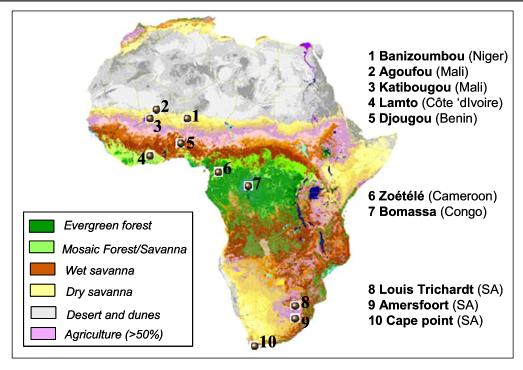


Figure 1. Vegetation and location map showing the distribution of the ten IDAF stations.

for example, N volatilization as  $NH_3$  and NO is strongly dependent on soil moisture and consequently on the precipitation regime (Delon *et al* 2012, 2014, Galy-Lacaux *et al* 2014). Little is known about critical loads in the tropics, which may be less N limited than temperate forests and show different responses to elevated N deposition. Thus, there is an urgent need for a greater understanding of the long-term impacts from low levels of nitrogen deposition in all systems, with particular emphasis on understudied biomes and areas such as the Tropics, Asia, and Africa (Bobbink *et al* 2010)

All studies based on long term observations in industrialized countries have helped to understand the negative effects of increasing N load in the atmosphere, and helped to make decisions on N mitigation. The budget presented in this work may be considered as one point in time budget, to be included in long term studies as one of the first reference point for SSA. Knowing what has been the evolution of nutrient use in industrialized regions, and the subsequent N release to the atmosphere, may help to propose key actions to produce more food and energy with less pollution in SSA. Moreover, it is important to consider to produce more food with a better N use efficiency, in crop and animal production, and increasing the fertilizer equivalence value of animal manure (ONW, Sutton *et al* 2013). For that, the present study gives the current state of N exchanges in SSA.

#### 1. Regional wet and dry nitrogen deposition

Dry and wet deposition of  $N_r$  are the final removal processes of  $N_r$  compounds from the atmosphere. Dry deposition describes the uptake of atmospheric species (gases or aerosol particles) at the surface of the Earth, and provides surface boundary conditions for atmospheric chemistry models. Dry deposition process could be described as the uptake by the surface (i) as an irreversible loss for the species (one-way deposition), or (ii) as a reversible process where the surface provides a reservoir for the species and allows re-emission to the atmosphere (two-way exchange). Many different techniques exist to measure directly dry deposition fluxes but are difficult to organize in monitoring networks because of the requirements for highly sophisticated methods and instrumentation (Wesely and Hicks 2000). The IDAF network in Africa, among few other global networks, has contributed to dry deposition estimates for the WMO assessment (Vet et al 2014) using the so-called 'inferential' technique, which involves measurements of ambient air concentrations of gases and/or particles by multiplying these concentration by dry deposition velocities estimated using models (Wesely and Hicks 2000, Zhang et al 2005, Shen et al 2009). Despite large uncertainties (Wesely and Hicks 2000), inferential dry deposition estimates remain the best available for assessing long term dry deposition at global and regional scale. In this study, we will consider new developments to quantify gaseous nitrogen dry deposition fluxes for African ecosystems performed by Adon et al 2013, including bi directional exchange of ammonia. Uncertainties on dry deposition fluxes are estimated to be larger than 30% (Delon et al 2010, Adon et al 2013, Zhang et al 2003a,b, Zhang et al 2010).

Wet deposition refers to the removal of gases and aerosols by scavenging in clouds and precipitation. Measurements of wet deposition are made by collection and analysis of rain using wet-only samplers, opening and closing the sampler at the beginning and end of rainfall events. Wet deposition is

Site	$(NO_2 + HNO_3 + NH_3)$ Gaseous dry deposition $(kgN ha^{-1} yr^{-1})$	$(p-NO_3^{-\circ}+^{\circ}pNH_4^+)$ Particulate dry deposition (kgN ha <sup>-1</sup> yr <sup>-1</sup> )	$(NH_4^{+\circ}+^{\circ}NO_3^{-})$ wet deposition (kgN ha <sup>-1</sup> yr <sup>-1</sup> )	Total deposition wet + dry $(\text{kgN ha}^{-1} \text{ yr}^{-1})$
Dry savannas	4–5.3	0.07–0.16	2–3,4	6.0-8.9
Wet savannas	3.4-4.6	0.03–0.06	3.5–5.3	6.9–10.0
Forest	11.2–11.8	_	3.6 <sup>a</sup>	14.8–15.4

Table 1. Total nitrogen deposition budget	in kgN ha <sup>-1</sup> yr <sup>-</sup>	<sup>1</sup> including gaseous dry deposition, particulate dry deposition and wet deposition.

<sup>a</sup> Wet deposition in forest: estimation for Zoétélé site.

4

estimated as the product of a species concentration in precipitation by the precipitation depth (WMO/GAW 2004). In the recent WMO global assessment of precipitation chemistry and deposition, all precipitation chemistry and wet deposition data have been carefully screened according international sampling protocols and analytical methods consistent with the WMO/GAW 2004 standard protocols. Data for all the regionally representative african sites (IDAF network) have been approved and included in the final assessment database. Measurements patterns of wet and dry deposition fluxes in Africa have been analyzed for two periods of reference 2000-2007 and 2005-2007 in Vet et al (2014). In this paper, we present the mean N deposition budget using the IDAF database for the whole period 2000-2007. The estimation of the atmospheric nitrogen deposition budget in West and Central Africa, including wet and dry processes, is based on IDAF experimental measurements including gas, particles and rain composition (http://idaf.sedoo.fr). We chose the mean 2000-2007 period because it corresponds to previous analysis given by Sigha et al (2003), Yoboue et al (2005), Galy-Lacaux et al (2009), Laouali et al (2012), Vet et al (2014) and also because of the availability of a combined database composed of rain chemistry, gas concentrations, dry deposition velocity calculations and environmental parameters (Adon et al 2010, 2013) during this period. In this paper, a transect of ecosystem from dry savanna to humid savanna and forest is studied using all the IDAF regionalrepresentative sites located pairs by pairs on each ecosystem. An analysis of the different components of the nitrogen atmospheric deposition in gaseous and particulate forms is performed, associated with measured wet deposition fluxes.

#### 1.1. Wet deposition

An automatic precipitation collector designed for the IDAF network has been installed in all stations. A local operator collects water from each rainfall event in a Greiner tube (50 ml). Rainwater samples were stored in a deep freeze and analyzed for pH, conductivity and major inorganic and organic ions. Analytical procedures are given in Galy-Lacaux and Modi (1998) and on the web site http://idaf.sedoo.fr. International intercomparison studies are carried out each year to ensure the quality of measurements. The Laboratoire d'Aérologie (LA) has been involved in these tests since 1996. All ion chromatography, pH and conductivity analyses carried out at the LA are evaluated through the WMO intercomparison studies and the analytical precision is estimated to be: 5% or better for mineral ions. Combining all the uncertainties of measurements and calculations, we estimate the global uncertainty of the wet deposition fluxes to be about 10%. To calculate wet nitrogen deposition in African ecosystems, we have compiled mean annual nitrate and ammonium content from the precipitation collected and a mean annual rainfall for the studied period from five IDAF sites. We propose here to quantify a range of wet deposition fluxes representative of the period 2000-2007 for the three main types of ecosystems: dry savannas, wet savannas and forests. The mean rainfall depth registered from 2000 to 2007 in

5

Banizoumbou (Niger) and Katibougou (Mali) representative of dry savannas was 449 and 744 mm, respectively; in Lamto (Côte d'Ivoire) and Djougou for wet savanna 1274 and 1261 mm, respectively; and in Zoétélé Cameroon for equatorial forests, 1557 mm. Mean nitrate and ammonium concentrations measured from 2000 to 2007 in the dry savannas sites in Niger and in Mali were  $11 \mu \text{eq } \text{L}^{-1}$  of NO<sub>3</sub> and 19  $\mu$ eq L<sup>-1</sup> of NH<sub>4</sub><sup>+</sup> (Galy-Lacaux *et al* 2009, Laouali *et al* 2012). In the wet savanna, measurements were  $8 \mu \text{eq } \text{L}^{-1}$  of  $NO_3^-$  and 18  $\mu$ eq L<sup>-1</sup> of NH<sub>4</sub><sup>+</sup> (Yoboue *et al* 2005). In forested ecosystem, we measure from 2000 to 2007 around 7  $\mu$ eq L<sup>-1</sup> of NO<sub>3</sub> and 10  $\mu$ eq L<sup>-1</sup> of NH<sub>4</sub><sup>+</sup>. These contents are comparable to those mentioned by Sigha et al (2003) from 1996 to 2000 and to previous measurements performed by Lacaux et al 1988 in the African equatorial forest in Congo, Dimonika site, for one year in 1987 (6.4  $\mu$ eq L<sup>-1</sup> of NH<sub>4</sub><sup>+</sup> and 8.6  $\mu$ eq L<sup>-1</sup> of NO<sub>3</sub>). However, nitrate and ammonium content at the congolese forested site of Boyélé in 1987 are higher, i.e.,  $15.4 \mu \text{eq } \text{L}^{-1}$  of NH<sup>+</sup><sub>4</sub> and  $19.4 \mu \text{eq } \text{L}^{-1}$  of NO<sup>-</sup><sub>3</sub>. The chemical composition of rain shows a decreasing gradient of nitrate and ammonium content from the dry savannas to the forest. In the dry savannas, biogenic emission of  $NO_x$  in soils have been identified as the major contributor to nitrate in rain water while ammonia emissions from animals waste are believed to be responsible for the high ammonium content (Galy and Modi 1998, Serça et al 1998, Galy-Lacaux et al 2009, Delon et al 2010). Future growth of population in agro-pastoral areas in the Sahel, where changes in land cover and soil degradation due to changes in land use are problematic, may increase the contribution of ammonia emission (i.e. use of synthetic fertilizers), whereas purely pastoral areas of the Sahel suffer less from man induced effects due to a lower population density (Mougin et al 2009).

Nitrogen wet deposition in the forms of nitrate and ammonium were calculated to be around 2-3.4 kgN ha<sup>-1</sup> yr<sup>-1</sup> in dry savannas, 3.5-5.3 kgN ha<sup>-1</sup> yr<sup>-1</sup> in wet savannas and about 3.6 kgN ha<sup>-1</sup> yr<sup>-1</sup> in forests (based only on Zoétélé site precipitation chemistry measurements) (table 1). According to the scarcity of the data for long term precipitation chemistry in African equatorial forest, we think that N wet deposition calculated for Zoétélé (Cameroon) using seven years of data represent the low range of N wet deposition fluxes. Despite the higher measured ammonium and nitrate rain content in dry savannas, the atmospheric nitrate and ammonium wet deposition fluxes present a positive gradient from the dry savanna to the wet savannas and the forest. Wet deposition fluxes are directly linked to the rainfall gradient with a range from about 450 to  $1550 \text{ mm yr}^{-1}$  along the studied ecosystems transect. According to the synthesis papers for the different IDAF sites that have studied the interannual variability of wet deposition fluxes, we assume that the total N wet deposition presents a maximum variation of ±25% around the mean (Sigha et al 2003, Yoboue et al 2005, Galy-Lacaux et al 2009, Laouali et al 2012). Even if the theoretical uncertainty on wet deposition fluxes measurement is about 10%, it seems reasonable to consider for N wet deposition fluxes an uncertainty of about 25% related to interannual variability combining the natural variability of rainfall depths

and of the different sources of gases and particles influencing nitrogen rain content.

#### 1.2. Dry deposition

*Gaseous dry deposition flux of nitrogen* has been calculated as the sum of dry deposition fluxes of ammonia (NH<sub>3</sub>), nitric acid (HNO<sub>3</sub>) and nitrogen dioxide (NO<sub>2</sub>) (table 1). Gaseous measurement (NH<sub>3</sub>, HNO<sub>3</sub>, NO<sub>2</sub>) are monthly integrated samples using passive sampling techniques following the work of Ferm (1994). This technique has been tested in different tropical and subtropical region (Ferm and Rodhe 1997, Carmichael *et al* 2003, Martins *et al* 2007). The uncertainties on HNO<sub>3</sub>, NO<sub>2</sub> and NH<sub>3</sub> concentration measurements, determined from the covariance of duplicates, are equal to 20%, 9.8% and 14.3%, respectively.

Mean annual gases concentrations for the different IDAF sites measured over the period 1998-2007 have been reported in Adon et al 2010. Mean concentrations are calculated for each type of ecosystems (same type of vegetation, same amount of rain) by grouping two sites in dry savannas (Banizoumbou (Niger) and Katibougou (Mali), two sites in wet savannas (Djougou (Benin and Lamto (Côte d'Ivoire) and two sites in forests (Zoétélé (Cameroon) and Bomassa (Congo). Concentrations are ranged from 1 in wet savannas and forest to 2.4 ppb in dry savannas for NO<sub>2</sub>, from 0.3 in forest to 0.5 ppb in dry savanna for HNO<sub>3</sub> and from 3.9 in wet savanna to 6.7 ppb in dry savnnas for NH<sub>3</sub>, respectively. As mentioned in the introduction of the regional N deposition budget, dry deposition flux is computed using the inferential method. It represents the product of measured gaseous concentrations (NO<sub>2</sub>, NH<sub>3</sub>, HNO<sub>3</sub>), and realistic dry deposition velocity according to the recent work of Adon et al 2013. Annual deposition velocities calculated from 2002 to 2007 range from 0.15 to  $0.32 \text{ cm s}^{-1}$  for NO<sub>2</sub>, from 0.68 to  $2.20 \text{ cm s}^{-1}$  for HNO<sub>3</sub> and from 0.22 to 0.84 cm s<sup>-1</sup> for NH<sub>3</sub> (Adon et al 2013). We assume that the uncertainties on dry nitrogen deposition fluxes are larger than 30% and mainly due to the concentration measurements, and to the estimate of the deposition velocity (Adon et al 2013, Delon et al 2010).

Detailed results, gas by gas, indicate that NO<sub>2</sub> dry deposition fluxes in annual mean presents a small variability on the period 2000-2007 according to the type of ecosystem with fluxes varying from 0.4 ( $\pm 0.$ ) kgN ha<sup>-1</sup> yr<sup>-1</sup> in wet savanna, 0.6  $(\pm 0.1)$  kgN ha<sup>-1</sup> yr<sup>-1</sup> in dry savanna to 0.7  $(\pm 0.2)$  kgN ha<sup>-1</sup> yr<sup>-1</sup> in forested ecosystem. NH<sub>3</sub> dry deposition fluxes present higher annual mean values with  $3.2 (\pm 0.6)$ and 2.9 ( $\pm 0.8$ ) kgN ha<sup>-1</sup> yr<sup>-1</sup> in dry savanna and wet savanna, respectively and 9.9 ( $\pm 0.2$ ) kgN ha<sup>-1</sup> yr<sup>-1</sup> over the forest. Major sources of NH<sub>3</sub> include bacterial decomposition from urea in animal excreta and emissions by natural or fertilized soils (Schlesinger and Hartley 1992). In Africa, another significant source of ammonia is produced by savanna fires and domestic fuelwood burning (Delmas et al 1995). Dry deposition fluxes of HNO3 are very low compared to NO2 and  $NH_3$  with values ranging between 0.7 (±0.01) and 1.0  $(\pm 0.01)$  kgN ha<sup>-1</sup> yr<sup>-1</sup> for all the ecosystems. This result is correlated to very low HNO<sub>3</sub> concentrations measured in all the stations (Adon *et al* 2010). Total dry deposition fluxes of gases (sum of the three dry deposition gases fluxes) are presented in table 1 and indicates a total of  $4-5.3 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  in dry savannas,  $3.4-4.6 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  in wet savannas to  $11.2-11.8 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  in forested ecosystems. The range is based on the annual mean from 2000 to 2007 for the two different sites regionally-representative of each ecosystem.

Dry deposition of nitrogen from particles was calculated according to particulate ammonium (pNH<sub>4</sub><sup>+</sup>) and nitrate (pNO<sub>3</sub>) concentrations determined in the water soluble content of the bulk aerosols sampling. The annual mean concentrations values were calculated for the period 1998-2000 (available data). To estimate nitrogenous dry deposition fluxes of particles, a dry deposition velocity value of  $0.1-0.2 \text{ cm s}^{-1}$  was used according to Zhang *et al* (2009) and Whelpdale et al (1996). The results of total dry deposition of  $(pNH_4^+)$  and  $(pNO_3^-)$  were of the same order in all types of ecosystems with relatively low values. For the mean particle concentrations measured in Banizoumbou and Katibougou, were  $pNH_4^+ = 0.31 \pm 0.02 \text{ ppb}$  and  $pNO_3^- = 0.16 \pm 0.02 \text{ ppb}$ 0.03 ppb in Banizoumbou,  $pNH_4^+ = 0.17 \pm 0.06$  ppb and  $pNO_3 = 0.23 \pm 0.06 ppb$  in Katibougou (Galy-Lacaux *et al* 2014). However the concentrations of aerosol with gaseous NH<sub>3</sub> observed in the two dry savannas sites ranged from 2.9 to 10.4 ppb in Banizoumbou and 1.8 to 6.9 ppb in Katibougou respectively which implied that particulate deposition were negligible compared to gaseous dry deposition. The mean annual particulate deposition fluxes  $(pNH_4^+ + pNO_3^-)$ , in the semi-arid, wet savanna ecosystems are around 0.07-0.16, 0.03-0.06 kgN ha<sup>-1</sup> yr<sup>-1</sup> respectively (table 1).

#### 2. Emissions of nitrogenous compounds

Each potential source of N compound emission to the west and central African atmosphere will be detailed in this paragraph. Methods of calculation will be presented briefly, and the reader is invited to refer to Delon *et al* (2010, 2012) for more information. The assumptions made to estimate each contribution will be explained, as well as all the uncertainties.

The N emission sources take into account simulated NO biogenic emission from soils,  $NH_3$  emission by volatilization estimated from the literature (merging measurements of N content in animal excreta and statistic data on livestock population) and  $NO_x$  and  $NH_3$  emission from biomass burning and domestic fires, both estimated from satellite data of burnt areas and existing data on emission factors.

#### 2.1. NO biogenic emission from soils

Biogenic emissions from soils are derived from an artificial neural network (ANN) approach. The resulting algorithm provides on line biogenic NO emissions and has been developed in Delon *et al* (2007). Fluxes depend on surface water filled pore space (WFPS) and temperature, soil deep temperature (20–30 cm), pH, sand percentage, fertilization rate and wind speed. It has been fully coupled to the SVAT model ISBA (Noilhan and Mahfout 1996) to reproduce NO

pulses in the Sahel region (Delon et al 2008), at the beginning of the wet season. Indeed, Pulsed NO emissions occur when very dry soil is wetted resulting in a reactivation of waterstressed bacteria (Hudman et al 2012). This algorithm has been adapted to reproduce NO biogenic emissions from soils in the whole West African region. NO emissions from soils in ISBA are obtained in a domain from -20 to 30°E, and from -5 to 20°N, at a spatial resolution of 0.5° and a time resolution of 3 h. In the following sections, NO fluxes are averaged on a 3°/3°, window around each specific IDAF station. The meteorological forcing used in the model has been developed within ALMIP (AMMA Land surface Model Intercomparison Project), from a data set based on the merging of ECMWF (European Centre of Medium Range-Weather Forecast) atmospheric state variables, and TRMM-3B42 3-hourly data for the precipitation. A more comprehensive description of ALMIP may be found in Boone et al (2009).

We assume that the most important driving parameters for our study are surface soil temperature and WFPS (calculated from soil moisture). Therefore, the uncertainty of the biogenic NO flux is calculated from the fluctuation of these two parameters and ranges from 3% to 45%.

The fertilization rate provided to the model is based on the calculation of N released by organic fertilization (i.e. cattle dung and manure), for each country. Indeed, data bases of land use do not provide enough information concerning synthetic fertilization. Galloway et al (2004) give an estimate of 2.5 TgN yr<sup>-1</sup> for the N input by fertilization production for the whole African continent. Therefore, significant work was done to develop our own data base for organic fertilization for 23 countries of West and Central Africa which are contained in our simulation domain. The N quantity released by livestock is calculated from Schlecht et al (1998), in gN head<sup>-1</sup> day<sup>-1</sup>, for cows, sheep and goats. This estimate is multiplied by the number of animals per km<sup>2</sup> in each country. The animal population is obtained in each region of each country from the FAO (Food and Agriculture Organization) Global Livestock Production and Health Atlas GLiPHA (2009, http://kids.fao.org/glipha/). 70% of the resulting N input is used to calculate NO emissions from soils (this value constitutes one of the seven factors used to calculate biogenic NO emission with a neural network approach), the rest (30%) will be used for the calculation of NH<sub>3</sub> volatilization. The overall uncertainty applied on NH<sub>3</sub> volatilization is 50% (estimated from Bouwman et al 1997, taking into account N release by livestock and animal population).

#### 2.2. NH<sub>3</sub> emission by volatilization

Ammonia is formed in soils from biological degradation of organic compounds and ammonium and depends on soil cation exchange reactions, soil moisture content and net mineralization. The current annual  $NH_3$  emission in developing countries of 15 TgN accounts for 2/3 of the global emissions from animal excreta. The fraction of the N excretion that is lost as  $NH_3$  ranges from 10% to 36%, depending on animal-waste management and animal category

(Bouwman and Van Der Hoek 1997, Bouwman *et al* 1997). More recently, Bouwman *et al* (2002a) stated that 25%, 28% and 33% of N use in Western Africa is released as  $NH_3$  in intensive grasslands, upland crops and wetland rice, respectively. The difficulty of obtaining reliable data concerning the use of animal manure, and management practices in tropical countries leads to uncertainties in estimating the  $NH_3$  loss.

As explained above, NH<sub>3</sub> volatilization rate is 30% of the N input by organic fertilization (N quantity contained in animal excreta multiplied by the number of heads of livestock in a given region). NH<sub>3</sub> volatilization estimate is therefore 5.5  $(\pm 3)$  kgN ha<sup>-1</sup> yr<sup>-1</sup> in dry savanna, 2.4  $(\pm 0.1)$  kgN ha<sup>-1</sup> yr<sup>-1</sup> in wet savanna and 0.6  $(\pm 0.5)$  kgN ha<sup>-1</sup> yr<sup>-1</sup> in forest.

Projected loss rates for 2025 (compared to 1990) for developing region calculated in Bouwman *et al* (1997) show an increase from 0.1 to 0.2 MtonsN yr<sup>-1</sup>. According to this study, the scenarios of food production and associated NH<sub>3</sub> emissions in developing countries show that the NH<sub>3</sub> emissions may increase significantly in the future, but less rapid than food production, due to an increase in efficiency of livestock and crop production.

#### 2.3. $NO_x$ and $NH_3$ emission from biomass burning

Global biomass burning inventories for gases and particles are available from January 2005 to December 2006 on the Laboratoire d'Aerologie website (http://www.aero.obs-mip. fr:8001/). The available spatial resolution is 1 km. and the time scale is daily. These global inventories use the L3JRC burnt area product (Liousse et al 2010) based on the SPOT -VGT vegetation satellite and Global Land Cover (GLC) vegetation map, together with data on biomass densities and burning efficiencies. Emission factors for gaseous species (NO, NO<sub>2</sub> and NH<sub>3</sub>) were chosen following Andreae and Merlet (2001). In dry and wet savannas, monthly means and averages of emissions from fires in a 5°/5° window around each specific station (to ensure a sufficient sampling of fire events) are used to evaluate the potential impact of biomass fires at the local/regional scale. In forest, a larger window has been used  $(6^{\circ}/6^{\circ})$ , to ensure a greater impact of fires in the region of Africa where biomass burning is one of the most active source of emission. The total uncertainty for both NH<sub>3</sub> and  $NO_x$  fluxes from biomass burning is 54%, calculated from burned areas, biomass density, burning efficiency and emission factors. Emission of  $NO_x$  and  $NH_3$  from biomass burning fires is 1.51 (±0.27) kgN  $ha^{-1}$  yr<sup>-1</sup> in dry savanna, 4.11  $(\pm 0.96)$  kgN ha<sup>-1</sup> yr<sup>-1</sup> in savanna wet and 6.13  $(\pm 1.61)$  kgN ha<sup>-1</sup> yr<sup>-1</sup> in forest.

#### 2.4. NO<sub>x</sub> and NH<sub>3</sub> emission from domestic fires

Combustion of biofuel is mainly used for cooking in West and Central Africa. Biofuel use provides a constant emission all year long and is a potential source of trace gases. In the present study,  $NO_x$  and  $NH_3$  emission from domestic fires uses the methodology from Junker and Liousse (2008), for the most recent existing year (2003). Reference  $NO_x$  emission factors values were given by Andreae and Merlet (2001), and on the two following websites: http://www.naei.org.uk/ reports.php, and http://www.transport.govt.nz/. NH<sub>3</sub> emissions factors are also given in Andreae and Merlet (2001). Consumption data are given by the United Nations database. Annual emissions are calculated country by country, and then gridded at 25 km/25 km resolution (Assamoi and Liousse 2010). The monthly input of nitrogen compounds is therefore constant all year long, and is averaged over a  $5^{\circ}/5^{\circ}$ window around each specific station to consider the local/ regional impact of these emissions.

The uncertainty applied to domestic fires (mainly linked to wood and charcoal consumption estimates from United Nation database and the lack of knowledge of emission factors as a function of countries, fuels, activity, technology and norm combinations) is 60% for both compounds. Emission of NO<sub>x</sub> and NH<sub>3</sub> from domestic fires are 0.39 ( $\pm 0.11$ ) kgN ha<sup>-1</sup> yr<sup>-1</sup> in dry savanna, 0.97 ( $\pm 0.02$ ) kgN ha<sup>-1</sup> yr<sup>-1</sup> in wet savanna and 0.22 ( $\pm 0.27$ ) kgN ha<sup>-1</sup> yr<sup>-1</sup> in forest, showing a lower impact than every other source of N compounds emission.

Anthropogenic emissions are not taken into account in this study. Indeed, IDAF sites where the different estimates are made remain far from large cities and from anthropogenic emission influence. Emissions of gases and particles from the combustion of fossil fuels and biofuels in Africa are expected to increase significantly in the near future due to the rapid growth of African cities and megacities, and the magnitude of African anthropogenic emissions could be similar to African biomass burning emissions around 2030 (Liousse *et al* 2010, 2014).

#### 3. Nitrogen deposition and emission budget

Dry deposition fluxes, estimated for the three African ecosystems were combined with those associated with wet deposition to provide an estimate of the annual nitrogen atmospheric deposition (table 1). The total nitrogen deposition is estimated to be around  $6.0-8.9 \text{ kgN ha}^{-1} \text{ yr}^{-1}$ ,  $6.9 -10.0 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  and  $14.8-15.4 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  respectively over dry savanna, humid savanna and over the forests (table 1). These values should be taken with caution and we estimated the uncertainties on the budget to be around 30%.

Stoorvogel *et al* (1993) assumes that annual nitrogen depletion rates averaged 22 kgN ha<sup>-1</sup> over 30 years in 37 sub-Saharan countries. Nitrogen deficiency is prevalent for smallholder agriculture in SSA, with only an average application of 8 kg of nutrients (not just nitrogen) per hectare in comparison with 100 in the US and India and 220 in China (UN Millennium Project Task Force on Hunger 2005). Our results show that in the current situation, atmospheric N deposition fluxes represent a significant nutrients input to the African ecosystems. These results are important to be taken in consideration for the agrosystems management, especially in dry savannas where N deposition, estimated to be in the range from 6 to  $8.9 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  is high compared to other ecosystems. Moreover, it is important to note that N deposition fluxes in the dry savannas are maximum and concentrated

during the wet season that lasts only few months at the yearly scale (three to four months in the sahelian ecosystems studied).

It is also important to mention that our budget does not take into account all nitrogenous species, especially organic nitrogen species. Africa is currently entirely missing from a database for organic nitrogen, although substantial research investment is needed. Some results suggest that organic nitrogen accounts for a significant fraction of total atmospheric nitrogen deposition (wet+dry deposition). (Cornell *et al* 2003, and references therein). In China, Cui *et al* (2014) indicates that manure application led to significant DON wet deposition and further to the increase of total N deposition.

The estimation of mean annual emissions including biogenic NO from soils, NO<sub>x</sub> and NH<sub>3</sub> from biomass burning and domestic fires, and NH<sub>3</sub> from volatilization are presented in table 2 according to the three main types of ecosystems. Dry savannas ecosystems are dominated by natural emissions of NO<sub>x</sub> from soils and NH<sub>3</sub> volatilization from animal excreta, accounting for 17% and 62% respectively of the total emission. In wet savannas, contributions of natural and biomass burning nitrogen compounds sources are in the same order of magnitude (25% of the total for the volatilization of NH<sub>3</sub>, 22% for biogenic NO emission and 43% for biomass burning emission of both  $NO_x$  and  $NH_3$ ). In forested ecosystems, biomass burning sources become dominant (72% of the total) and NH<sub>3</sub> from volatilization remains low (7% of the total emission budget) (figure 2). Net NO emissions from soils are calculated above canopy, a portion of NO<sub>2</sub> (oxidation product of NO) being deposited within the canopy. Biomass burning emissions of NH<sub>3</sub> and NO<sub>x</sub> increase along the latitudinal transect dry savanna/wet savanna/forest.

Figure 2 presents the results of the annual emission/ deposition budget at the regional scale of the African ecosystems including all the different estimated contributions. An important result highlighted by this budget is the importance of dry deposition processes in west and central Africa for nitrogenous gaseous compounds. In dry savanna and forest, the relative contribution of dry deposition is between 63% and 76%. In wet savanna the contribution is around 50%. In the deposition part of the budget, the dry deposition of ammonia is dominant for all the ecosystems representing 34-53% of the total. In addition, we note that wet deposition of ammonium represents the second predominant contribution for all the ecosystems accounting for 14-36% of the total deposition. This deposition of reduced N compounds should be mainly related to the emission through the volatilization of NH<sub>3</sub> that dominates in dry savannas with 62%, and represents 25% and 24% in wet savannas and forests, respectively. In dry and wet savanna, the estimated contributions of oxidized N compounds in the emission and deposition part of the budget are in the same order of magnitude. The sum of  $NO_2 + HNO_3$  dry deposition and nitrate wet deposition represents 32% in dry savanna and 30% in wet savanna whereas total emissions of NO<sub>x</sub> from biomass burning and NO from soils are 27% in dry savanna and 45% in wet savanna. In the forest, the N oxidized deposition representing

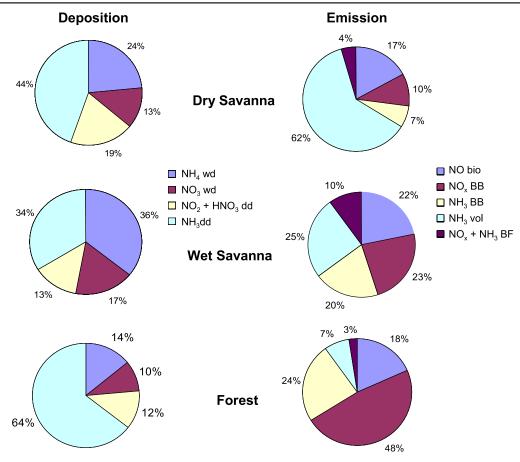


Figure 2. Nitrogen Emission deposition budget for the three main type of ecosystems.

Table 2. Nitrogen	emission	budget	in	kgN ha	-1 yr-	<sup>1</sup> .
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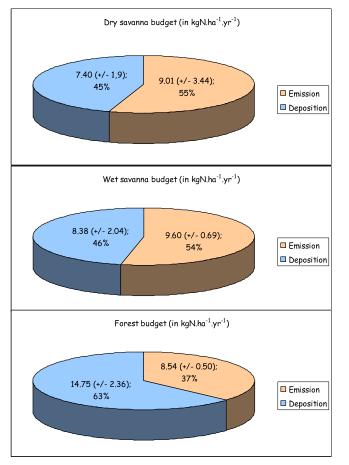
Site	Biogenic NO (kgN ha <sup>-1</sup> yr <sup>-1</sup> )	$NO_x BB (kgN ha^{-1} yr^{-1})$	$\frac{\rm NH_3 BB}{\rm (kgN ha^{-1} yr^{-1})}$	NH <sub>3</sub> volatiliza- tion (kgN ha <sup>-1</sup> yr <sup>-1</sup> )	$NH_3^{\circ} + {}^{\circ}NO_x$ domestic fires (kgN ha <sup>-1</sup> yr <sup>-1</sup> )	Total emission $(kgN ha^{-1} yr^{-1})$
Dry savannas	1.50-1.59	0.80-1.02	0.52-0.68	3.43-7.68	0.32-0.47	6.98–11.44
Wet savannas	1.98-2.26	1.91-2.48	1.51-2.30	2.34-2.47	0.96-0.99	9.12-10.09
Forest	1.37–1.76	3.48-4.70	1.51-2.57	0.22-1.02	0.03-0.41	7.80–9.27

Note: BB-biomass burning.

22% of total deposition is over balanced by emission of oxidised N compounds which reaches 66% of the total emission. Local sources of nitrogen oxidized emissions taken into account in dry and wet savanna may balance the deposition budget, which is not the case in the forest. We suppose that long range transport of biomass burning emissions from both hemispheres influence the forested studied sites.

To illustrate the nitrogen emission/deposition budget, figure 3 compares the total annual emission and deposition fluxes. Figure 3 shows that emission sources of nitrogen compounds are in equilibrium with deposition fluxes in dry and wet savannas, with respectively 7.4 ( $\pm$ 1.9) deposited and 9.01 ( $\pm$ 3.44) kgN ha<sup>-1</sup> yr<sup>-1</sup> emitted in dry savanna, 8.38 ( $\pm$ 2.04) kgN ha<sup>-1</sup> yr<sup>-1</sup> deposited and 9.60 ( $\pm$ 0.69) kgN ha<sup>-1</sup> yr<sup>-1</sup> emitted in wet savanna. In forested ecosystems, the nitrogen budget is dominated by wet+dry

deposition processes  $(14.75 \pm 2.36 \text{ kgN ha}^{-1} \text{ yr}^{-1})$ , compared to emission processes  $(8.54 \pm 0.50 \text{ kgN ha}^{-1} \text{ yr}^{-1})$ . Equatorial forests could be influenced by emissions coming from the both hemispheres, especially by biomass burning emissions. Indeed, Chen *et al* (2010) showed that the highest levels of reactive N and N<sub>2</sub> fire emissions occurred at about 7°N in the northern hemisphere and about 10°S in the southern hemisphere. Therefore, the N deposition over forests could be affected by savanna fires emissions transported further equator ward and reaching tropical forests. This equatorward transport pattern was not unique to Africa and has been retrieved in Southeast Asia and South America. This may be explained more generally by a covariance between fire emissions and relatively strong surface winds (toward the ITCZ) associated with the winter hemisphere of a Hadley cell (Plumb and Mahlman 1987). In addition, equatorial forests are well known to be a strong sink of compounds in term of



**Figure 3.** N emission and deposition budget for the three major ecosystems in west and Central Africa, wd: wet deposition, dd: dry deposition, bio: biogenic emissions, BB: biomass burning emissions, vol: volatilization.

dry deposition processes (Saunois *et al* (2009), Mari *et al* (2011)). Wet deposition fluxes are also assumed to be large considering the high amount of rain above this ecosystem.

#### Conclusion

In this study, we have taken into account the contribution of major N compounds to establish and document the annual N atmospheric budget at the regional scale for three main types of ecosystems in West and Central Africa (dry savanna, wet savanna and forest). This original budget relies on measurements and/or calculations of N emissions and deposition fluxes regionally representative of the main African ecosystems. The approach combines both unique experimental data based on the IDAF deposition program and modeling studies, especially developed for SSA to estimate nitrogen atmospheric exchanges.

NH<sub>3</sub> remains the largest contributor to the total budget, both for emission and deposition processes, in the three types of ecosystem. Big differences however are to be noted in the source of emission, largely dominated by NH<sub>3</sub> volatilization from animal excreta in dry savanna, and by biomass burning in wet savanna and forests. The consequence on deposition is that NH<sub>3</sub> is the most important compound involved in deposition processes. Models usually underestimate total deposition of N in forests and savanna ecosystems, with an underestimation of a factor 2 in dry savannas (Vet *et al* 2014). Simulated wet deposition fluxes are quite in agreement with measurements on the African continent; however modelled dry deposition fluxes are more uncertain. Our results highlight the significance of dry deposition processes in the total deposition budget in dry savanna, and for 40–60% and 70% in wet savanna and forests respectively.

Emission sources of nitrogen compounds are in equilibrium with deposition fluxes in dry and wet savannas, whereas in forested ecosystems, the nitrogen budget is dominated by wet+dry deposition processes, influenced by biomass burning emissions coming from savannas situated northward and southward and transported over the Central African forest.

This work brings a new insight in deposition and emission regional budgets in remote areas in Africa. Our results show that in the current situation, atmospheric N deposition fluxes represent a significant nutrients input to the African ecosystems (from 8 to  $14 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ). However, a lot of uncertainties remain, in particular concerning the importance of organic compounds in wet deposition. As far as the authors know, these compounds have been neither measured nor calculated on African ecosystems,.

In the future, N emissions and deposition are likely to decrease for certain compounds like  $NO_x$  in temperate regions, particularly in developed countries (Vet *et al* 2014), but may increase more than predicted in the models for other compounds like  $NH_3$  at the global scale (Lieven *et al* 2009). In contrast, N emissions and deposition in tropical and subtropical regions are likely to increase for all compounds. (Dentener *et al* 2006, Galloway *et al* 2008, Lamarque *et al* 2013, Liousse *et al* 2014). Therefore, it is important to study present N levels and underlying processes, to be able in the future to detect potential trends on N compounds exchanges in SSA.

Dentener et al (2006) used the IDAF measurements to validate deposition fluxes over the African continent in a global modelling study showing that only 9.3% of the tropical forests have N deposition rate greater than  $10 \text{ kgN ha}^{-1} \text{ yr}^{-1}$ , a critical load over which the negative effect is evident, such as an increase in ecosystem productivity with possible consequences for the global carbon cycle, or possible saturation in soil and biomass storage leading to losses to the atmosphere and aquatic systems (Dentener et al 2006, Phoenix et al 2006). Forests in 'Too little N' regions may be poorly buffered against additional acidifying inputs, and may be more prone to release of N species through gaseous losses and leaching (Matson et al 1999). In 'Too much N' regions, N accumulation is the main driver of changes to species composition and is a recognized threat to plant diversity (Bobbink et al 2009)

Until now, agro-systems in West and Central Africa have been managed using very few quantities of synthetic N fertilizers (Bouwman *et al* 1997, Potter *et al* 2010). In the future, at least in wet savanna areas, land use change, through intensive deforestation, will imply the use of larger quantities of synthetic fertilizer, leading to high quantities of N released to the atmosphere in the form of NO and NH<sub>3</sub>. There is a need for a greater understanding of the long-term impacts from low levels of nitrogen deposition in all systems, with particular emphasis on understudied biomes and geographic areas such as the Tropics, Asia, and Africa (Bobbink et al 2009, Clark et al 2013). An important purpose would be to provide future assessments and policies for Africa focused on a multi criteria approach to meeting anthropogenic needs and rendering ecosystems sustainable. Regional modelling of N emission compounds with new fertilization rates needs to be done to quantify the N release from soils in the near future. In addition, more detailed N emission measurements are needed to better document and validate modelling studies of biogenic emissions from soils in regions where data are seldom available. Improvement of our understanding of atmospheric N fluxes in tropical regions, as in Africa require long-term measurements program to collect N deposition measurement including wet and dry processes. Africa, with insufficient access to nutrients, should be a priority for future global assessment on the nitrogen cycle and the improvement in nutrient use efficiency at the global scale. Our work on atmospheric nitrogen budget, regionally representative of the main African ecosystems, represents an important contribution for 'low N input regions', which could be taken (1) as a baseline for quantities of N exchanged between the soil and the atmosphere and (2) as a way to discriminate the importance of different sources of N depending on the type of ecosystem (dry savanna, wet savanna or forest) in non perturbed ecosystems of Western Africa. This contribution should be taken into consideration in future integrated nitrogen management system for Africa as a reference point before possible anthropogenic perturbations such as substantial N losses to the atmosphere and aquatic environments.

#### Acknowledgments

This work is part of the **IDAF** (IGAC/DEBITS/AFRICA) project; and was funded by INSU/CNRS 'Institut National des Sciences de l'Univers/ Centre National de Recherche Scientifique'. The authors are grateful to all the principal investigators of the IDAF network, to the field technician at the African station and IRD (Institut de Recherche pour le Developpement) for logistical support.

#### References

- Adon M, Galy-Lacaux C, Delon C, Yoboue V, Solmon F and Kaptue Tchuente A T 2013 Dry deposition of nitrogen compounds (NO<sub>2</sub>, HNO<sub>3</sub>, NH<sub>3</sub>), sulfur dioxide and ozone in west and central African ecosystems using the inferential method *Atmos. Chem. Phys.* **13** 11351–74
- Adon M et al 2010 Long term measurements of sulfur dioxide, nitrogen dioxide, ammonia, nitric acid and ozone in Africa

using passive samplers *Atmos. Chem. Phys. Discuss.* **10** 4407–61

- Andreae M O and Merlet P 2001 Emission of trace gases and aerosols from biomass burning *Glob. Biogeochem. Cycles* **15** 955–66
- Boone A 2009 The AMMA Land Surface Model Intercomparison Project (ALMIP) *Bull. Amer. Meteor. Soc.* 1865–80
- Bouwman A F, Boumans L J and Batjes N H 2002a Estimation of global NH<sub>3</sub> volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands *Glob. Biogeochem. Cycles* **16** 1024
- Bouwman A F, Lee D S, Asman W A H, Dentener F J, Van Der Hoek K W and Olivier J G J 1997 A global highresolution emission inventory for ammonia *Glob. Biogeochem. Cycles* **11** 561–87
- Bouwman A F and Van Der Hoek K W 1997 Scenarios of animal waste production and fertilizer use and associated ammonia emission for the developping countries *Atmos. Environ.* **31** 4095–102
- Bobbink R *et al* 2010 Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis *Ecological Applications* **20** 30–59
- Carmichael G R *et al* 2003 Measurements of sulfur dioxide, ozone and ammonia concentrations in Asia, Africa, and South America using passive samplers *Atmos. Environ.* **37** 1293–308
- Chen Y, Randerson J T, Van Der Werf G R, Morton D C, Mu M and Kasibhatla P S 2010 Nitrogen deposition in tropical forests from savanna and deforestation fires *Glob. Change Biol.* 16 2024–38
- Clark C M *et al* 2013 Nitrogen deposition and terrestrial biodiversity *Encyclopedia of Biodiversity* vol 5 2nd edn ed S A Levin (Waltham, MA: Academic) pp 519–36
- Cornell S E, Jickells T, Cape J N and Duce R A 2003 Organic nitrogen deposition on land and coastal environments: a review of methods and data *Atmos. Environ.* **37** 2173–91
- Cui J, Zhou J, Peng Y, He Y, Yang H and Mao J 2014 Atmospheric wet deposition of nitrogen and sulfur to a typical red soil agroecosystem in Southeast China during the ten-year monsoon seasons (2003–2012) Atmos. Environ. 82 121–9
- Delmas R A, Lacaux J P, Menaut J C, Abbadie L, Le Roux X, Helas G and Lobert G 1995 Nitrogen compound emission from biomass burning in tropical African savanna, FOS/DECAFE 91 experiment (Lamto, Ivory Coast) J. Atmos. Chem. 22 175–94
- Delon C, Galy-Lacaux C, Adon M, Liousse C, Boone A, Serça D, Diop B, Akpo A and Mougin E 2014 Interannual Variability of the Atmospheric Nitrogen Budget in West African Dry Savannas. Chapter 11. Nitrogen Deposition, Critical Loads and Biodiversity. Sutton M A, Mason K E, Sheppard L J, Sverdrup H, Haeuber R, Hicks W K (Eds.) (Berlin: Springer) XXIX
- Delon C, Galy-Lacaux C, Adon M, Liousse C, Serça D, Diop B and Akpo A 2012 Nitrogen compounds emission and deposition in West African ecosystems: comparison between wet and dry savanna *Biogeosciences* **9** 385–402
- Delon C, Serca D, Boissard C, Dupont R, Dutot A, Laville P, de Rosnay P and Delmas R 2007 Soil NO emissions modelling using artificial neural network *Tellus* B **59** 502–13
- Delon C et al 2010 Atmospheric nitrogen budget in Sahelian dry savannas Atmos. Chem. Phys. 10 2691–708
- Delon C, Reeves C E, Stewart D J, Serca D, Dupont R, Mari C, Chaboureau J-P and Tulet P 2008 Biogenic nitrogen oxide emissions from soils-impact on NOx and ozone over West Africa during AMMA (African Monsoon Multidisciplinary Analysis): modelling study Atmos. Chem. Phys. 8 2351–63
- Dentener F, Vet R, Dennis R L, Du E, Kulshrestha U C and Galy-Lacaux C 2014 Progress in monitoring and modelling estimates of nitrogen deposition at local, regional and global

scales *Nitrogen Deposition, Critical Loads and Biodiversity* ed M A Sutton *et al* (Berlin: Springer) chapter 2

- Dentener F *et al* 2006 Nitrogen and sulfur deposition on regional and global scales: a multimodel evaluation *Glob. Biogeochem. Cycles* **20** GB4003
- Ferm M, Lindskog A, Svanberg P-A and Boström C-A 1994 New measurement technique for air pollutants (in Swedish) *Kemisk Tidskrift* **1** 30–2
- Ferm M and Rodhe H 1997 Measurements of air concentrations of SO<sub>2</sub>, NO<sub>2</sub> and NH<sub>3</sub> at rural and remote sites in Asia *J. Atmos. Chem.* 27 17–29
- Galloway J N, Townsend A R, Erisman J W, Bekunda M, Cai Z, Freney J R, Martinelli L A, Seitzinger S P and Sutton M A 2008 Transformation of the nitrogen cycle: recent trends, questions, and potential solutions *Science* **320** 889–92
- Galloway J N *et al* 2004 Nitrogen cycles: past, present, and future *Biogeochemistry* **70** 153–226
- Galy-Lacaux C, Laouali D, Descroix L, Gobron N and Liousse C 2009 Long term precipitation chemistry and wet deposition in a remote dry savanna site in Africa (Niger) *Atmos. Chem. Phys.* 9 1579–95
- Galy-Lacaux C and Modi A I 1998 Precipitation chemistry in the Sahelian savanna of Niger, Africa J. Atmos. Chem. **30** 319–43
- Galy-Lacaux C *et al* 2014 Dry and wet atmospheric nitrogen deposition in West Central Africa *Nitrogen Deposition, Critical Loads and Biodiversity* ed M A Sutton *et al* XXIX (Berlin: Springer) chapter 10
- Godfray H C J, Beddington J R, Crute I R, Haddad L, Lawrence D, Muir J F, Pretty J, Robinson S and Thomas S M 2010 Camilla toulmin. food security: the challenge of feeding 9 billion people Science 327 812–8
- Hudman R C, Moore N E, Mebust A K, Martin R V, Russell A R, Valin L C and Cohen R C 2012 Steps towards a mechanistic model of global soil nitric oxide emissions: implementation and space based-constraints *Atmos. Chem. Phys.* **12** 7779–95
- Junker C and Liousse C 2008 A global emission inventory of carbonaceous aerosol including fossil fuel and biofuel sources for the period 1860–1997 Atmos. Chem. Phys. 8 1–13
- Lacaux J P, Servant J, Huertas M L, Cros B and R Delmas 1988 Precipitation chemistry from remote sites in African equatorial forest *Eos Trans. AGU* **69** 1069
- Lamarque J-F *et al* 2013 Atmospheric chemistry and climate model intercomparison project (ACCMIP): evaluation of historical and projected future changes *Atmos. Chem. Phys.* **13** 7997–8018
- Laouali D, Galy-Lacaux C, Diop B, Delon C, Orange D, Lacaux J P, Akpo A, Lavenu F, Gardrat E and Castera P 2012 Long term monitoring of precipitation chemical composition and wet deposition over three Sahelian Savannas Atmos. Environ. 50 314–27
- Liousse C, Assamoi E, Criqui P, Granier C and Rosset R 2014 Explosive growth in African combustion emissions from 2005–2030 *Environ. Res. Lett.* **9** 35003–12
- Liousse C *et al* 2010 Updated African biomass burning emission inventories in the framework of the AMMA-IDAF program, with an evaluation of combustion aerosols *Atmos. Chem. Phys.* **10** 9631–46
- Mari C *et al* 2010 Atmospheric composition of West Africa: highlights from the AMMA program *Atmos. Sci. Lett.* **12** 13–8
- Martins J J, Dhammapala R S, Lachmann G, Galy-Lacaux C and Pienaar J J 2007 Long-term measurements of sulphur dioxide, nitrogen dioxide, ammonia, nitric acid and ozone in southern Africa using passive samplers *South Afr. J. Sci.* **103** 336–42
- Matson P A *et al* 1999 The globalization of N deposition: ecosystem consequences in tropical environments *Biogeochemistry* **46** 67–83
- Mougin E *et al* 2009 The AMMA-CATCH Gourma observatory site in mali: relating climatic variations to changes in vegetation,

surface hydrology, fluxes and natural resources *J. Hydrol.* **375** 14–33

- Noilhan J and Mahfouf J-F 1996 The ISBA land surface parameterization scheme *Glob. Planet. Change* **13** 145–59
- Palm C A, Myers R J K and Nandwa S M 1997 ReplenishingSoil Fertility in Africa ed R J Buresh et al (Madison, WI: Soil Science Society of America) pp 193–217 special publ. 5
- Phoenix G K *et al* 2006 Atmospheric nitrogen deposition in world biodiversity hotspots: the need for a greater global perspective in assessing N deposition impacts *Glob. Change Biol.* **12** 470–6
- Pienaar J J 2005 Proposal of a new IGAC II task: DEBITS II (Deposition of Biogeochemically Important Trace Species), available at www.igac.noaa.gov/DEBITS.php
- Plumb R A and Mahlman J D 1987 The zonally averaged transport characteristics of the GFDL general circulation model *J. Atmos. Sci.* 44 298–327

Potter P, Ramankutty N, Bennett E N and Donner S D 2010 Characterizing the spatial patterns of global fertilizer

- application and manure production *Earth Interact.* **14** 1–22 Sanchez P 2010 Soil fertility in tropical Africa *Nat. Geosci.* **3** 299–300
- Sanchez P et al 2007 Poverty and hunger special feature—research articles—social sciences—sustainability science. the African Millennium Villages Proc. Natl Acad. Sci. **104** 16775–80
- Saunois M, Reeves C E, Mari C, Murphy J G, Stewart D J, Mills G P, Oram D E and Purvis R M 2009 Ozone budget in the West African lower troposphere during the AMMA (African monsoon multidisciplinary analysis) campaign Atmos. Chem. Phys. 9 6979–7032
- Schlesinger W H and Hartley A E 1992 A global budget for NH<sub>3</sub> Biogeochemistry 15 191–211
- Serça D, Delmas R, Le Roux X, Parsons D A B, Scholes M C, Abbadie L, Lensi R, Ronce O and Labroue L 1998 Comparison of nitrogen monoxide emissions from several African tropical ecosystems and influence of season and fire *Glob. Biogeochem. Cycles* 12 637–51
- Shen J L, Tang A H, Liu X J, Fangmeier A, Goulding K T W and Zhang F S 2009 High concentrations and dry deposition of reactive nitrogen species at two sites in the North China Plain *Environ. Pollut.* **157** 3106–13
- Sigha-Nkamdjou L, Galy-Lacaux C, Pont V, Richard S, Sighoumnou D and Lacaux J P 2003 Rainwater chemistry and wet deposition over the equatorial forested ecosystem of Zoétélé (Cameroon) J. Atmos. Chem. 46 173–98
- Sobota D J, Compton J E and Harrison J A 2013 Reactive nitrogen inputs to US lands and waterways: how certain are we about sources and fluxes *Frontiers Ecol. Environ.* **11** 82–90
- Stoorvogel I J, Smaling E M A and Janssen B J 1993 Calculating soil nutrient balances in Africa at different scales *Fertilizer Res.* 35 227–35
- Sutton M A, Howard C M, Erisman J W, Billen G, Bleeker A, Grennfelt P, van Grinsven H and Grizzetti B 2011 *The European Nitrogen Assessment: Sources, Effects, and Policy Perspectives 466* (Cambridge: Cambridge University Press)
- Sutton M A, Mason K E, Sheppard L J, Sverdrup H, Haeuber R and Hicks W K (ed) 2014 *Nitrogen Deposition, Critical Loads and Biodiversity* XXIX (Berlin: Springer) p 535
- Sutton M A *et al* 2013 Our Nutrient World: The challenge to produce more food and energy with less pollution. Global Overview of Nutrient Management. Centre for Ecology and Hydrology, Edinburgh on behalf of the Global Partnership on Nutrient Management and the International Nitrogen Initiative
- Ti C P, Pan J J, Xia Y Q and Yan X Y 2012 A nitrogen budget of mainland China with spatial and temporal variation *Biogeochemistry* 108 381–94
- Vet R *et al* 2014 A global assessment of precipitation chemistry and sulfiur nitrogen, sea salt, base cations, organic acids a,d pH, and phosphorus *Atmos. Environ.* **93** 3–100

- Vitousek P M, Aber J D, Howarth R W, Likens G E, Matson P A, Schindler D W, Schlesinger W H and Tilman D G 1997 Human alteration of the global nitrogen cycle: sources and consequences *Ecol. Appl.* **7** 737–50
- Vitousek P M *et al* 2009 Nutrient imbalances in agricultural development *Science* **324** 1519–20
- Wesely M L 1989 Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models *Atmos. Environ.* 23 1293–304
- Wesely M L and Hicks B B 2000 A review of the current status of knowledge in dry deposition Atmos. Environ. 34 2261–82
- Whelpdale D M et al 1996 Global acid deposition assessment ed D M Whelpdale and M S Kaiser (World Meteorological Organisation, Global Atmosphere Watch) WMO Report No 106
- Yoboue V, Galy-Lacaux C, Lacaux J P and Silue S 2005 Rainwater chemistry and wet deposition over the wet savanna ecosystem of lamto (Côte d'Ivoire) *J. Atmos. Chem.* **52** 117–41

- Zhang L, Brook J R and Vet R 2003a Evaluation of a non-stomatal resistance parameterization for SO2 dry deposition *Atmos. Environ.* **37** 2941–7
- Zhang L, Brook J R and Vet R 2003b A revised parameterization for gaseous dry deposition in air-quality models *Atmos. Chem. Phys.* **3** 2067–82
- Zhang L, Brook J R, Vet R, Wiebe A, Mihele C, Shaw M, OBrien J M and Iqbal S 2005 Estimation of contributions of NO2 and PAN to total atmospheric deposition of oxidized nitrogen across Eastern Canada Atmos. Environ. 39 7030–43
- Zhang L, Vet R, OBrien J M, Mihele C, Liang Z and Wiebe A 2009 Dry deposition of individual nitrogen species at eight Canadian rural sites J. Geophys. Res. 114 D02301
- Zhang L, Wright P L and Asman W A H 2010 Bi-directional air surface exchange of atmospheric ammonia—a review of measurements and a development of a big-leaf model for applications in regional-scale air-quality models *J. Geophys. Res.* **115** D20310