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**Atmospheric  
nitrogen budget in  
Sahel**

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# Atmospheric nitrogen budget in Sahelian dry savannas

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## Abstract

The atmospheric nitrogen budget depends on emission and deposition fluxes both as reduced and oxidized nitrogen compounds. In this study, a first attempt at estimating the Sahel nitrogen budget for the year 2006 is made, through measurements and simulations at three stations from the IDAF network situated in dry savanna ecosystems. Dry deposition fluxes are estimated from measurements of  $\text{NO}_2$ ,  $\text{HNO}_3$  and  $\text{NH}_3$  gaseous concentrations, and wet deposition fluxes are calculated from  $\text{NH}_4^+$  and  $\text{NO}_3^-$  concentrations in samples of rain. Emission fluxes are estimated including biogenic emission of NO from soils (an Artificial Neural Network module has been inserted into the ISBA-SURFEX surface model), emission of  $\text{NO}_x$  and  $\text{NH}_3$  from domestic fires and biomass burning, and volatilization of  $\text{NH}_3$  from animal excreta.

This study uses original and unique data from remote and hardly-ever-explored regions. The monthly evolution of oxidized N compounds shows that deposition increases at the beginning of the rainy season because of large emissions of biogenic NO (pulse events). Emission of oxidized compounds is dominated by biogenic emission from soils (domestic fires and biomass burning account for 27% at the most, depending on the station), whereas emission of  $\text{NH}_3$  is dominated by the process of volatilization. Deposition fluxes are dominated by gaseous dry deposition processes (58% of the total), for both oxidized and reduced compounds. The average deposition flux in dry savanna ecosystems ranges from 8.6 to 10.9  $\text{kgN ha}^{-1} \text{yr}^{-1}$ , with 30% attributed to oxidized compounds, and the other 70% attributed to  $\text{NH}_x$ . The average emission flux ranges from 7.8 to 9.7  $\text{kgN ha}^{-1} \text{yr}^{-1}$ , dominated by  $\text{NH}_3$  volatilization (67%) and biogenic emission from soils (24%). The annual budget is then balanced, with emission fluxes on the same order of magnitude as deposition fluxes.

When scaled up to the Sahelian region ( $10^\circ \text{N}$ : $20^\circ \text{N}$ ,  $15^\circ \text{W}$ : $10^\circ \text{E}$ ), the estimates of total emission range from 3.6 to 4.5  $\text{TgN yr}^{-1}$  and total deposition ranges from 3.9 to 5  $\text{TgN yr}^{-1}$ . The N budget gives a net deposition flux ranging from 0.2 to 0.6  $\text{TgN yr}^{-1}$ . If scaled up to the global scale (in the tropical band), it is possible to calculate a total

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budget of oxidized and reduced N compounds for dry savannas, with a global nitrogen deposition flux ranging from 11.1 to 14.1 TgN yr<sup>-1</sup>, and a global emission flux ranging from 10.1 to 12.5 TgN yr<sup>-1</sup>. These ecosystems contribute a significant amount (around 12%) to the global nitrogen budget.

## 1 Introduction

Nitrogen is a key compound both as a nutrient for plants and animals and as an atmospheric pollutant. In the atmosphere, several nitrogen trace compounds are present, such as NO, NO<sub>2</sub>, HNO<sub>3</sub>, N<sub>2</sub>O and NH<sub>3</sub>, as well as particulate and aqueous forms such as NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>. At the global scale, the reactive N cycle has been widely impacted by human activities, notably for food production. Indeed, the creation of reactive nitrogen has increased by 120% since 1970 and reached 187 TgN yr<sup>-1</sup> in 2005, which is a consequence of the increase of the world population (Galloway et al., 2008). Anthropogenic emissions of NO<sub>x</sub> are better quantified than natural emissions, and NH<sub>3</sub> emissions from all sources and at all scales remain largely uncertain (Sutton et al., 2007). Nitrogen emissions from the soil are the origin of the above mentioned gaseous products, whereas enrichment of nitrogen in the soil will take place through biological nitrogen fixation, nitrogen wet and dry deposition or nitrogen fertilization (Vlek, 1981). Reactive Nitrogen emissions are influenced by several environmental and physical parameters, such as soil temperature and moisture, soil pH, texture, wind speed, plant cover, floristic composition (e.g. legumes) and N input (fertilization) (Williams et al., 1992; Yienger and Levy, 1995 and references therein; Potter et al., 1996; Bouwman et al., 2002b; Meixner and Yang, 2004; Delon et al., 2007). In semi arid and arid regions, limited water resources will have significant consequences on nitrogen cycling in the soil and the atmosphere. The seasonal rainfall distribution leads to an accumulation of N in soils during the dry season, and to large pulses of N emission at the beginning of the rainy season (Austin et al., 2004; Jaegle et al., 2004). Resulting emissions from these pulse events release high quantities of NO<sub>x</sub> in the atmosphere, contributing to

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increased ozone formation in the troposphere and long range transport of ozone. In Sahelian regions, (the part of West Africa defined as (10° N:20° N, 15° W:10° E) nitrogen emissions may come from different sources throughout the year. In this study, we will focus on simulated biogenic soil emissions of NO<sub>x</sub>, calculated biomass burning and domestic fuel emissions of NO<sub>x</sub> and NH<sub>3</sub>, and calculated volatilization of NH<sub>3</sub> from animal manure. Neither industrial N sources nor N emissions from synthetic fertilization are taken into account because of the remote location of the Sahel from big cities or industrial centres, and because synthetic fertilizers are either used sparingly or not at all in this region of the world. Similar to emission, wet and dry deposition play an essential role in determining the concentration of nitrogen compounds in the atmosphere, and the nitrogen input to the soil/plant system.

The IDAF (IGAC/DEBITS/AFrica) programme started in 1995 with the establishment of 10 measurement sites representative of major African ecosystems. The objectives of the programme are to study dry and wet deposition of important trace species and more generally the biogeochemical cycles of key nutrients. In this way, the IDAF activity is based on high quality measurements of atmospheric chemical data (gaseous, precipitation and aerosol chemical composition) on the basis of a multi-year monitoring. Since 2005, the ORE (Environmental Research Observatory) IDAF has been part of the AMMA (African Monsoon Mutidisciplinary Analyses) EOP (Extensive Observation period) and LOP (Long Observation period) program in West Africa, and within the SACCLAP program (South African Climate Change Air Pollution- PICS NRF/CNRS) in South Africa. The IDAF network and the resulting scientific research have been presented in several papers, such as Galy-Lacaux and Modi, 1998; Galy-Lacaux et al., 2001; Yoboué et al., 2005; Galy-Lacaux et al., 2009. The objective of this study is the calculation of the balance between N compounds emission and deposition, in order to quantify the atmospheric nitrogen budget in dry savannas. A focus will be made in 3 study sites which are representative of rural semi-arid savannas: Banizoumbou (Niger, 13.33° N, 2.41° E), Katibougou (Mali, 12.5° N, 7.3° W) and Agoufou (Mali, 15.3° N, 1.5° W), for the year 2006 (see Fig. 1).

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In order to calculate N compound emission and deposition, and to quantify the distribution of different emission sources, methods of calculation of emission and deposition fluxes will be presented. The assumptions made to estimate each contribution will be explained, as well as all sources of uncertainties.

5 In the following, the budget will be calculated first for oxygenated N compounds ( $\text{NO}_x$  for emissions,  $\text{NO}_2$ ,  $\text{HNO}_3$  and  $\text{NO}_3^-$  for deposition) at the monthly and annual scale for the year 2006, and second for  $\text{NH}_x$  products ( $\text{NH}_3$ ,  $\text{NH}_4^+$ ) at the annual time scales. Then, the total nitrogen compound budget will be calculated at the 3 IDAF stations, and scaled up to the Sahelian regional scale ( $10^\circ \text{N}$ : $20^\circ \text{N}$ ,  $15^\circ \text{W}$ : $10^\circ \text{E}$ ). A tentative budget  
10 at the global scale for tropical regions with dry savanna ecosystems will be estimated.

## 2 Material and methods

### 2.1 Deposition

The IDAF network is the French contribution to the international IGAC (International Global Atmospheric Chemistry)/DEBITS (Deposition of Biogeochemically Important Trace Species) program. The DEBITS committee has defined a set of experimental and analytical protocols to have comparable measurements from all the DEBITS stations. The IDAF network has adopted these protocols as well. Further information on  
15 the IDAF network may be found at <http://medias.obs-mip.fr/idaf/>.

Continuous wet and dry deposition measurements have been performed in Bani-zoumbou ( $13.3^\circ \text{N}$ ,  $2.41^\circ \text{E}$ , Niger) from 1994 until now, in Agoufou ( $15.3^\circ \text{N}$ ,  $1.5^\circ \text{W}$ , Mali) from 2004 to 2007, and in Katibougou ( $12.5^\circ \text{N}$ ,  $7.3^\circ \text{W}$ , Mali) from 1997 up to now. Comprehensive descriptions of the stations can be found in Mougou et al. (2009) for Agoufou, in Galy-Lacaux and Modi (1998) for Banizoumbou, and in Adon et al. (2009) for Katibougou. Agoufou and Banizoumbou are part of the AMMA CATCH  
20 (Couplage de l'Atmosphère Tropicale et du Cycle Hydrologique) observatory. Bani-zoumbou is located in a rural and agro pastoral area of the Sahelian region of Niger,

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approximately at 60 km from Niamey. Katibougou is located at 60 km from Bamako. No major source of anthropogenic pollution influences the station. Agoufou is situated towards the northern limit of the area reached by the West African monsoon.

The atmospheric nitrogen deposition budget estimated in this paper is based on experimental measurements for the year 2006. Because of missing data for  $\text{NH}_3$  concentrations in Agoufou during several months in 2006, a mean value (integrating measurements performed during the years 2005, 2006 and 2007) was used for  $\text{NH}_3$  dry deposition fluxes in Agoufou.

To estimate atmospheric nitrogen deposition, including both wet and dry processes, we compiled the IDAF nitrogen data from gas and rain chemistry measurements. This study will give the relative contribution of dry and wet deposition processes to the total nitrogen deposition in 2006.

### 2.1.1 Dry deposition

Gaseous measurements ( $\text{NH}_3$ ,  $\text{HNO}_3$ ,  $\text{NO}_2$ ) are monthly integrated samples using passive sampling techniques following the work of Ferm et al. (1994). This technique has been tested in different tropical and subtropical regions (Ferm and Rodhe, 1997; Carmichael et al., 2003; Martins et al., 2007). All samples of the IDAF west-central African stations are brought to the Laboratory of Aerology (LA) in Toulouse, France, for ionic chromatography analysis. All details and performances of IDAF passive samplers are given in Martins et al. (2007).

At each IDAF station, passive samplers have been exposed monthly in duplicate. To give an indication of the precision of this sampling technique, the covariance of all duplicate samples were calculated and found to be 20%, 9.8%, 14.3% for  $\text{HNO}_3$ ,  $\text{NO}_2$  and  $\text{NH}_3$ , respectively. The covariances compare well with those reported by Martins et al. (2007), at the Southern African sites, for the same pollutants (20%, 8.3%, 15.3%, for  $\text{HNO}_3$ ,  $\text{NO}_2$  and  $\text{NH}_3$ , respectively. The average of the duplicate samplers was used in all cases except when contamination of one of the samplers was suspected (this happened in less than 5% of all data). The detection limit of the passive samplers

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was evaluated with the variations in the blank amounts of the impregnated filters and found to be  $0.05(\pm 0.03)$  ppb for  $\text{HNO}_3$ ,  $0.2(\pm 0.1)$  ppb for  $\text{NO}_2$  and  $1(\pm 0.5)$  ppb for  $\text{NH}_3$ , respectively. For the  $\text{HNO}_3$  and  $\text{NO}_2$  measurements, 11.5% and 3.8% were below the detection limit, respectively.

5 The dry deposition flux is the product of the measured concentration in the air (ppb) with the corresponding deposition velocity, which is estimated according to the site and the species. Gaseous dry deposition of nitrogen has been calculated as the sum of dry deposition fluxes of ammonia ( $\text{NH}_3$ ), nitric acid ( $\text{HNO}_3$ ) and nitrogen oxide ( $\text{NO}_2$ ).

10 Zhang et al. (2003) has calculated maximum deposition velocities for 26 different land use categories. The most appropriate land use category for our study could be the desert, which has surface properties that are close to dry savanna ecosystems, and present deposition velocity of 0.2–0.2 cm/s for  $\text{NO}_2$  2–3.3 cm/s for  $\text{HNO}_3$  and 0.1–1.5 cm/s for  $\text{NH}_3$  (the first value is for dry ecosystems, and the second value is for wet ecosystems). As a comparison, Zhang et al. (2009) have calculated several dry  
15 deposition velocities for nitrogen compounds at rural sites in Canada which are small values even above forests or grasslands (0.17 cm/s for  $\text{NO}_2$ , 1.2 cm/s for  $\text{HNO}_3$  and 0.4 cm/s for  $\text{NH}_3$  as mean values for all referenced sites). If low values are found above vegetated areas in Canada, we theorize that Sahelian ecosystems will not have high deposition velocities. Considering that no deposition velocities have been measured  
20 in dry savanna areas, the following values have been chosen (according to the above cited studies) in this work to calculate deposition fluxes: 0.2 cm/s for  $\text{NO}_2$ , 1 cm/s for  $\text{HNO}_3$ , and 0.35 cm/s for  $\text{NH}_3$  as mean deposition velocities. It is obvious that the choice of constant deposition velocities all year long will introduce supplementary uncertainties in the deposition flux calculation, but this assumption is necessary to  
25 achieve the calculation of the N budget with the available data and is consistent with temporal scale of gas measurements. Particulate N dry deposition ( $\rho\text{NH}_4^+$  and  $\rho\text{NO}_3^-$ ) is not taken into account in this budget. Indeed, Galy-Lacaux et al. (2003) have shown that N dry deposition from particles have relatively low values ( $0.12\pm 0.01$ ,  $0.08\pm 0.01$  and  $0.06\pm 0.01$  kg N ha<sup>-1</sup> yr<sup>-1</sup>, respectively, in semi-arid, humid savanna and forested

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ecosystems). One should note that N particulate dry deposition is smaller than that of wet deposition by an order of magnitude. As far as the authors know, no other particulate dry deposition measurements have been made in the remote regions of Sahel.

## 2.1.2 Wet deposition

An automatic precipitation collector specially designed for the IDAF network has been installed at all stations. A local operator collects water from each rainfall event in a Greiner tube (50 ml). Preserving the rainwater samples from contamination is an important issue since microbial input could modify its chemical composition. Samples are refrigerated at 4°C and preserved with 15 mg of thymol biocide or stored in a deep freeze environment. Ion Chromatography (IC) procedures are given in Galy-Lacaux and Modi (1998). To calculate wet nitrogen deposition in African dry savannas for the year 2006, we have compiled the annual Volume Weighed Mean (VWM) concentrations of nitrate and ammonium from the precipitation collected at the 3 IDAF stations. The computation of nitrate and ammonium wet deposition has been done according to the 2006 annual rainfall for the studied sites. The rainfall depths registered at Banizoumbou (Niger), Katibougou and Agoufou (Mali) are 505.2 mm, 588.2 mm and 374 mm, respectively. Nitrate and ammonium concentrations at the dry savannas sites are in the upper range of all the African ecosystems, with values from 37 (in Banizoumbou) to 31 (in Katibougou)  $\mu\text{eq L}^{-1}$  of  $\text{NH}_4^+$  and a range of 7 (Banizoumbou) to 16 (Katibougou)  $\mu\text{eq L}^{-1}$  of  $\text{NO}_3^-$  (Sigha et al., 2003; Yoboué et al., 2005; Dentener et al., 2006).

The major source of precipitation nitrate content comes from natural  $\text{NO}_x$  emissions from soils, whereas  $\text{NO}_x$  production from lightning plays a minor role for wet deposition (Tost et al., 2007), and it is not taken into account in this budget. Nitrate concentration in precipitation represents the final result of homogeneous and heterogeneous processes of nitrogenous gases and particles in the atmosphere and cloud water. Nitric oxide (NO) is the major nitrogen compound released from savanna soils in the non-burning season (Serça et al., 1998), and a large fraction of NO produced is oxidized in the

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atmosphere through photochemical reactions, into  $\text{HNO}_3$  or organic nitrates.  $\text{HNO}_3$ , which is extremely soluble in water, is thus easily scavenged by clouds. Galy-Lacaux et al. (2001) have demonstrated that heterogenous processes between nitric acid and mineral particles are present all over the African continent.

Ammonium content in precipitation results from inclusion of gaseous ammonia and particles containing  $\text{NH}_4^+$  in precipitating clouds. The highest values of ammonium compounds in precipitation registered in the semi-arid regions have been attributed to strong sources of ammonia from domestic and pastoral animals during the wet season (Galy-Lacaux and Modi, 1998).

## 2.2 Emission

### 2.2.1 NO biogenic emission from soils: model characteristics

Biogenic emissions from soils are derived from an Artificial Neural Network (ANN) approach. The resulting algorithm provides on line biogenic NO emissions and is developed in Delon et al. (2007). It is fully coupled to the Soil Vegetation ATmosphere (SVAT) model ISBA (Interactions between Soil Biosphere and Atmosphere, Noilhan and Mahfouf, 1996), and has been previously tested in the 3-D coupled chemistry-dynamics model MesoNH-C (which uses surface scheme ISBA) to reproduce NO pulses after a rain event in Niger (Delon et al., 2008). NO emissions from soils in ISBA are obtained for the year 2006 at a spatial resolution of  $0.5^\circ$  and a time resolution of 3 h. In the following sections, NO fluxes are averaged on a  $3^\circ/3^\circ$  window around each specific station. The simulated domain extends from  $5^\circ$  S to  $20^\circ$  N in latitude, and from  $20^\circ$  W to  $30^\circ$  E in longitude, but only the Sahel region ( $15^\circ$  W: $10^\circ$  E,  $10^\circ$  N: $20^\circ$  N) will be explored in this study. The meteorological forcing, developed within the AMMA Land surface Model Intercomparison Project (ALMIP), is obtained 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. LAND-SAF data is used for downwelling longwave and shortwave radiative fluxes (Geiger et al., 2008; Huffman

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et al., 2007). A more comprehensive description of ALMIP may be found in Boone et al. (2009).

The NO flux from soil is a non linear function of seven soil surface parameters: surface Water Field Pore Space (WFPS), surface and deep soil temperatures, pH, sand percentage, fertilisation rate and wind speed. Surface WFPS is deduced from the simulated soil surface moisture. Surface and deep soil temperatures are also calculated by the model. Wind speed is provided by the atmospheric forcing. The pH map is obtained from IGBP-DIS (1998, <http://www.sage.wisc.edu/atlas>) soil data base, at a spatial resolution of  $0.5^\circ/0.5^\circ$ . Sand percentage is obtained from the ECOCLIMAP data base (Masson et al., 2003), at a resolution of 1 km/1km.

The fertilization rate provided to the model is based on the calculation of N released by organic fertilization (i.e. cattle dung), for each country. Indeed, data bases of land use do not provide enough information in the Sahel concerning synthetic fertilization. Galloway et al. (2004) give an estimate of  $2.5 \text{ Tg N yr}^{-1}$  for the N input by fertilization production for the whole African continent, and Bouwman et al. (1997) show no emission from synthetic fertilizer use in the Sahelian band ( $10\text{--}20^\circ \text{ N}$ ). Schlecht and Hiernaux (2004) state that manure from livestock is an important source of organic matter and nutrients due to limited access to mineral fertilizers. These low assessments corroborate the assumption that the use of synthetic fertilizers in the Sahel is not a common feature, and can be neglected in our inventory.

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. These countries are, in alphabetical order: Benin, Burkina Faso, Cameroon, Chad, Congo, Democratic Republic of Congo, Equatorial Guinea, Gabon, Gambia, Ghana, Guinea Bissau, Guinea, Ivory Coast, Liberia, Mali, Mauritania, Niger, Nigeria, Republic of Centre Africa, Senegal, Sierra Leone, Sudan, and Togo.

The N quantity released by livestock is calculated from Schlecht et al. (1998), in  $\text{gN head}^{-1} \text{ day}^{-1}$ , for cows, sheep and goats. This estimate is multiplied by the number of animals per  $\text{km}^2$  in each country. The animal population is obtained from a

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FAO (Food and Agriculture Organization) report for the years 1985–1986 (Hoste et al., 1988), and it was re estimated for the year 2006 as advocated in the report. The resulting inventory of N is introduced in the ANN algorithm as N input in  $\text{kgN ha}^{-1} \text{hour}^{-1}$ . The data needed to build this inventory are, however, a source of uncertainty. Even if the number of livestock is readjusted for the year 2006, the N input calculated from Schlecht et al. (1998), which gives  $45 \text{ kgN animal}^{-1} \text{yr}^{-1}$  in a small region of Niger, remains low when generalized to the West African region. Indeed, Mosier et al. (1998) gives an estimate of  $170 \text{ kgN animal}^{-1} \text{yr}^{-1}$  for the whole African continent, and Bouwman and Van Der Hoek (1997) gives an estimate of  $130 \text{ kgN animal}^{-1} \text{yr}^{-1}$  in developing countries. These differences may be explained first because poultry, camels and donkey excreta are not taken into account in Schlecht et al. (1998) study, and second because this study is limited to a small region in Niger where the acidic sandy soils, which are predominant in the region, are inherently poor in N (Schlecht et al., 2004). Compared to the West African region, they do not provide a representative uptake of N in food for grazing. Therefore, we have taken an intermediate value of  $70 \text{ kgN animal}^{-1} \text{yr}^{-1}$  for cows and  $20 \text{ kgN animal}^{-1} \text{yr}^{-1}$  for sheep and goats for our study domain, which yields a total N input of  $9 \text{ kgN ha}^{-1} \text{yr}^{-1}$  in Mali and  $7 \text{ kgN ha}^{-1} \text{yr}^{-1}$  in Niger. (These two countries are of particular interest because of the location of the IDAF stations).

However, this estimate remains low when compared to other information sources (where available). The Ministry of Agriculture in Mali (Direction Nationale de la Production et des Industries Animales, DNPIA) indicate a number of 29 340 000 for cows, sheep and goats (unpublished data) for the whole country in 2006 (F. Gangneron, personal communication, 2009). The estimate given by the FAO (and re-adjusted for the year 2006) is only 17 770 000, which is approximately 40% less. We can therefore reasonably suppose that the estimate from the FAO is underestimated for some other countries.

Furthermore, the majority of the livestock population is concentrated in regions where people live, i.e. the human and livestock population is more significant in the southern part of Mali and Niger than in the northern part. As a consequence, and con-

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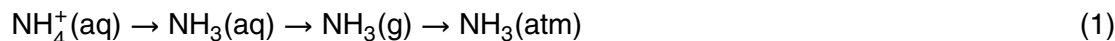
sidering that the livestock population is underestimated by the FAO inventory, we have chosen to increase the N input by 40% in a band extending from 12° N to 16° N, without decreasing the input northward in order to be more in accordance with the above cited studies. This assumption leads to a total N input of 12 kgN ha<sup>-1</sup> yr<sup>-1</sup> in southern Mali, and 10 kgN ha<sup>-1</sup> yr<sup>-1</sup> in southern Niger from animal manure.

Half of this N will be used as input for the calculation of NO biogenic emissions by the ANN algorithm in the ISBA-SURFEX surface model, the other half will be used for the calculation of NH<sub>3</sub> volatilization (the justification for this distribution is explained below, Sect. 2.2.2).

Figure 1 shows the NO biogenic flux from soils in the simulation domain in terms of the annual mean in figure 1a and JJAS (June July August September) in Fig. 1b. Indeed, JJAS corresponds to the wet season in West Africa, when the monsoon flux (cool and wet air coming from the ocean) reaches its northernmost position bringing humid air and intense precipitation in the Sahel. As mentioned in Yienger and Levy, 1995; Jaegle et al., 2004; Butterbach-Bahl et al., 2004 and references cited therein, NO emission is principally driven by soil moisture in tropical regions, and the most intense emissions occur in the Sahel when the first rains fall on the very dry soils. As shown in Fig. 1b, soil emissions in the Sahel reach their strongest values during the rainy season from June to September. In this study, we will only focus on the Sahelian region (10° N:20° N, 15° W:10° E) and the magnitude of fluxes in wet savanna or forests areas or other tropical ecosystems will not be discussed here.

### 2.2.2 NH<sub>3</sub> emission by volatilization

Ammonia is formed in soils from biological degradation of organic compounds and ammonium, as represented by the Eq. (1):



Where (aq) stands for aqueous, (g) for gaseous and (atm) for atmospheric loss. NH<sub>4</sub><sup>+</sup>(aq) depends on soil cation exchange reactions, soil moisture content and net min-

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eralization. The concentration of  $\text{NH}_3(\text{aq})$  depends on ammoniacal N, temperature and pH. The rate of gas dispersion in the natural environment depends on the temperature gradient, and wind speed (Vlek, 1981). After  $\text{NH}_3$  is emitted to the atmosphere, each nitrogen atom can participate in a sequence of effects, known as the nitrogen cascade in which a molecule of  $\text{NH}_3$  can, in sequence, impact atmospheric visibility, soil acidity, forest productivity, terrestrial ecosystem biodiversity, stream acidity, coastal productivity (Galloway and Cowling, 2002), soil acidification and eutrophication (Bouwman and Van Der Hoek, 1997).

The current annual  $\text{NH}_3$  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  $\text{NH}_3$  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  $\text{NH}_3$  in intensive grasslands, upland crops and wetland rice, respectively. However, the difficulty of obtaining reliable data concerning the use of animal manure, and management practices in tropical countries leads to uncertainties in estimating the  $\text{NH}_3$  loss, particularly in semi-arid regions like the Sahel. However, one can suppose that better conditions are encountered in the Sahel region which favours  $\text{NH}_3$  volatilization, such as high temperatures, low soil moisture and bare soil surfaces. As a consequence, 50% of loss rate has been applied to the input of N by animal manure previously prescribed for the calculation of NO emissions. This leads to a N- $\text{NH}_3$  volatilization estimated at  $6 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  in southern Mali, and  $5 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  in southern Niger. Bouwman et al. (1997) estimate the emission from domesticated animals in the Sahelian band between  $0.5$  and  $5 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  for the year 1990.

### 2.2.3 $\text{NO}_x$ and $\text{NH}_3$ emission from biomass burning

Global biomass burning inventories for gases and particles are available from January 2005 to December 2006 on the Laboratoire d'Aérodologie website (<http://www.aero.obs-mip.fr:8001/>). The available spatial resolution is 1 km/1 km, and the time scale is

daily. These global inventories use the L3JRC burnt area product based on the SPOT-VGT vegetation satellite and Global Land Cover (GLC) vegetation map, together with data on biomass densities and burning efficiencies. Such data, which were not available for the GLC2000 vegetation types but for the UMD (Hansen et al., 2000) global land cover product (Michel et al., 2005) have been established from correspondences between the 13 UMD vegetation classes and GLC2000 vegetation classes (Mieville et al., 2009). Emission factors for gaseous species were chosen following Andreae and Merlet (2001). In the present study, we used the inventories for NO, NO<sub>2</sub> and NH<sub>3</sub> species in the region of interest (−5 to 20° N, −20 to 30° E). Monthly means and averages 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.

Long range transport of nitrogen emissions from remote fires over southern Africa are not considered here due to the northward position of the stations in dry savanna areas. Indeed, Mari et al. (2008) and Sauvage et al. (2007) have shown that the impact of fire emissions occurring in the Southern Hemisphere from June to September was limited to coastal regions in the gulf of Guinea, and did not influence sites to the north.

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

Combustion of biofuel is mainly used for cooking in the Sahel. Biofuel use provides a constant emission all year long and is a potential source of trace gases. Ludwig et al. (2003) (and references therein) show that 0.5 TgNO/yr is emitted over the African continent due to biofuel consumption. In the present study, NO<sub>x</sub> and NH<sub>3</sub> emission from domestic fires uses the methodology from Junker and Liousse (2008), for the most recent existing year (2003). Due to a lack of knowledge of the emission factor as a function of countries, fuels, activity, technology/norm combinations, this methodology is based on a lumping procedure to account main factors of vulnerability. Nine emission factors are defined for each fuel category, and 3 main activities are considered (traffic, domestic, industrial), with 3 levels of technology linked to each country's

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development level (developed, semi-developed, developing). Reference  $\text{NO}_x$  emission factors values were given by Andreae and Merlet (2001), and on the 2 following web-sites: <http://www.naei.org.uk/reports.php>, and <http://www.transport.govt.nz/>.

$\text{NH}_3$  emissions factors are also given in Andreae and Merlet (2001). Consumption data are given by the United Nations database. Annual emissions are then calculated country by country, and then gridded at 25 km/25 km resolution (Assamoi and Liousse, 2009). 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.

### 3 Results and discussion

As mentioned in Sect. 2.1, deposition fluxes (dry + wet) are estimated from concentration measurements at the IDAF stations. All deposition fluxes are calculated in  $\text{kgN ha}^{-1} \text{yr}^{-1}$  and compared to emission fluxes in the same unit. Oxidized and reduced N compounds will be first treated separately. Then, a total budget will be estimated at the annual scale.

#### 3.1 Monthly evolution of Nitrogen oxidized compounds

Nitric oxide (NO) and nitrogen dioxide ( $\text{NO}_2$ ) react rapidly in the atmosphere and are referred to jointly as  $\text{NO}_x$ . In turn,  $\text{NO}_x$  can be incorporated into organic compounds such as peroxyacetyl nitrate (PAN) or alkyl nitrate, or further oxidized to  $\text{HNO}_3$ . Gas-phase  $\text{HNO}_3$  can be converted to aerosol nitrate ( $\text{NO}_3^-$ ) (e.g., by reaction with ammonia). PAN can convert back into  $\text{NO}_2$ , and in hot temperature conditions its lifetime is short (few hours), so concentrations may remain low despite abundant photochemical radicals that promote PAN formation (Munger et al., 1998). We can therefore reasonably consider that the emission of NO (both biogenic and anthropogenic) at the surface is the beginning of the formation of all other reactive oxidized nitrogen compounds in the at-

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mosphere. Non reactive N compounds ( $N_2$ ,  $N_2O$ ,  $N_2O_5$ ) are not included in the budget, because emission fluxes of these compounds in arid regions are negligible. According to Meixner and Yang (2004),  $N_2O$  fluxes from semi-arid soils are expected to be low, and those from arid soils are very low, due to the fact that  $N_2O$  emission is favoured by denitrification processes at high soil moisture content. Galbally et al. (2008) (and references therein) suggest, however, that 5% of the global soil emissions of  $N_2O$  from agricultural disturbances (i.e. land conversion to agriculture) may occur in semi arid and arid zones. This fraction is considered to be low, hence  $N_2O$  emissions are not considered. Organic compounds such as PAN are also omitted from this budget, assuming that they are converted into  $NO_2$  and that their concentrations are negligible in non polluted areas (around 100 ppt, Seinfeld and Pandis, 1998). As a consequence in this study, the total  $NO_x$  emission flux is defined as the sum of biogenic  $NO$  soil emission flux+biomass burning  $NO_x$  flux+domestic fire  $NO_x$  flux. It is compared to deposition flux of oxidized components, defined as the sum of dry deposition of  $NO_2+HNO_3$  in the gas phase, + $NO_3^-$  wet deposition flux.

Figure 2 presents the monthly evolution of emission and deposition oxidized N fluxes in Agoufou, Banizoumbou and Katibougou. Emission and deposition fluxes are similar during the wet season. Mean fluxes for each station at each season are reported in Table 1. During the dry season, emission fluxes are larger than deposition fluxes, but this is not representative of a real physical effect and can be easily explained: the emission module is calibrated such that it never attains a null emission, which leads to an overestimated numerical background noise and an overestimate of emission during the dry season. This default should be corrected in a future version of the emission module.

The maximum emission value is close to the maximum deposition during the wet season at all stations, and the second maximum observed in Katibougou and Banizoumbou in November corresponds to fire emissions, which imply an increase in  $NO_x$  fluxes. This second maximum does not appear in deposition fluxes, because  $HNO_3$  deposition fluxes, which are superior to  $NO_2$  fluxes, do not increase with fire emission



increase. The similar orders of magnitude observed between emission and deposition both in wet and dry seasons leads to the conclusion that  $\text{NO}_2$  and  $\text{HNO}_3$  deposition velocities have been correctly estimated for the region.

However, note the time lag between the beginning of emission and deposition fluxes at all stations except in Katibougou. Strong deposition fluxes begin at the very beginning of the wet season (usually around May in dry savanna areas, depending on the station) due to the pulse emission flux produced over very dry soils when the first rains fall (Johansson et al., 1988; Yienger and Levy, 1995; Otter et al., 1999; Yan et al., 2005; Delon et al., 2008). Measurements in the 3 stations show that the  $\text{NO}_2$  concentration at the surface has a first maximum at the beginning of the rainy season (C. Galy-Lacaux, personal communication, 2009). In the model, emissions begin with a one month lag (compared to deposition) in Agoufou and Banizoumbou, and on time at Katibougou, where the beginning of simulated emissions corresponds to that of the measured deposition, but with less intensity.

The reason for this time lag resides in the minimum rainfall amount needed by the emission module to initiate  $\text{NO}$  fluxes. Indeed, if this threshold (close to 10 mm for one rain event) is not reached, the soil is not wetted enough to produce sufficient emissions. The quantity of rainfall is crucial for the module to initiate strong emissions at the beginning of the rain season. In situ measurements show that such a high threshold is not necessary, and the sensitivity of the emission module should be modified (i.e. threshold decreased). This modification will be considered in a further version of the module). In the forcing, the quantity of rainfall is respected, but it remains difficult to compare point rainfall measurements to a satellite-based product at a 0.5 degree spatial scale, especially because of the mostly convective nature of the rainfall which results in low spatial correlation length scales over this region (e.g. Ali et al., 2003). Furthermore, an effect of smoothing could reinforce the problem. A sensitivity test has been made to give some proofs to this statement, and we have tried to reduce the sensitivity of the biogenic emission flux algorithm to the quantity of rainfall by a factor 2, i.e. we have doubled the precipitation.

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Figure 3 shows the result of this test. Compared to Fig. 2, it is obvious that this test does not have any realistic justification, because the surface energy budget will be drastically changed and will have non expected consequences on the results (for example, if the quantity of rain increases, the surface temperature will decrease and the fluxes will also change). However, this test helps to conclude that if the quantity of rain is high at the beginning of the rainy season, the emissions will begin early. That is verified in Banizoumbou and Katibougou. In Agoufou, the northernmost position, a time lag of 1 month subsists. The quantity of rainfall decreases when the latitude increases, and when reaching the northern limit of the ITCZ (Inter Tropical Coverage Zone) position, the emission module has less rainfall available to ensure an immediate emission flux. As stated before, an adjustment of the threshold in the emission module is necessary to provide sufficient NO emission from soils at the beginning of the rainy season.

In the following sections, the emission flux estimate will be deduced from the real precipitation forcing.

The comparison of these two way fluxes in Fig. 2 at the monthly scale shows a good agreement between emission and deposition magnitude, while underscoring the difficulties of analysing the nitrogen budget in such remote areas where too few measurements are available. Emission modelling presents an alternative to this lack of measurements, but many questions remain concerning, for example, the quantity of nitrogen contained in the soils, or the power of emissions after the first rainfall in semi-arid regions. However, the joint exploitation of IDAF measurements and modelling results gives a unique opportunity to provide a nitrogen budget in Sahelian ecosystems. In the following, we will try to evaluate the contribution of biogenic and fire emissions to atmospheric deposition at the annual scale, both for oxidised and reduced nitrogen compounds.

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## 3.2 Annual and seasonal budget of N oxidised compounds

Figure 4 shows the distribution of the N oxidized component emission fluxes (a) and deposition fluxes (b), for the wet season. The same repartition is given in Fig. 5a and b but for annual means in the three dry savanna stations.

It is obvious from these figures that the emission flux is dominated by the biogenic part, both in the wet season and for the annual mean. Even by decreasing the biogenic flux during the dry season (because of the numerical overestimation mentioned above), the emission flux will keep the same shape. Mean values for each season are reported in Table 1. Emission fluxes at the annual scale range from 1.7 to 3.2 kgN ha<sup>-1</sup> yr<sup>-1</sup>. These estimates are in the range of emission fluxes reported by Meixner and Yang (2004), which range from 0.02 to 34 kgN ha<sup>-1</sup> yr<sup>-1</sup>, in semi arid and arid ecosystems. In Banizoumbou, measured emission fluxes up to 1.8 kgN ha<sup>-1</sup> yr<sup>-1</sup> have been reported by Le Roux et al. (1995) and Serça et al. (1998).

In Fig. 4b and Fig. 5b, dry deposition appears to be 1/2 to 2/3 of the total deposition, with the remaining 1/3 being wet deposition. This has already been observed in dry savanna stations and other IDAF stations in West African wet savanna (Galy-Lacaux et al., 2003).

Large deposition fluxes are observed in Katibougou and Agoufou because of high wet deposition of NO<sub>3</sub>. Katibougou, which is the southernmost station, receives more rainfall than Banizoumbou and Agoufou, which could explain the high wet deposition flux. But in Agoufou, the strong deposition flux in the wet season can only be explained by significant biogenic emissions at the beginning of this season which are not very well reproduced by the model. Figures 4a and 5a show that no biomass burning contribution appears in Agoufou, due to the small quantity of biomass available during the dry season. NO<sub>2</sub> concentration measurements do not show any increase in Agoufou in that season (Adon et al., 2009). The quantity of N available in the soil is not part of the systematic measurements in IDAF stations, but measurements in the Gourma region (Diallo and Gjessing, 1999) do not reflect any strong reservoir of N. The release of

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nitrogen compounds in Agoufou specifically (because of the reduced rainfall amount) and other dry savanna stations seems to be very powerful. Preliminary explanation could lie in the N input from N-fixing crusts of cyanobacteria and lichens (Barger et al., 2005), and N-fixing plants and trees (e.g. Acacia). Meixner and Yang (2004) state that despite the fact that deserts have low plant productivity, inorganic nitrogen can accumulate in desert and semi-desert soils during long dry periods.

Generally speaking, the deposition fluxes of oxidised N compounds in dry savanna stations is the consequence of strong biogenic emission fluxes in the wet season. Indeed, the contribution of emissions from biomass burning at the annual scale is only 19% and 24% of the total oxidised N deposition flux, respectively, in Banizoumbou and Katibougou, and the contribution of domestic fires is negligible.

### 3.3 Annual and seasonal budget of reduced compounds

The dry and wet deposition of  $\text{NH}_3$  and  $\text{NH}_4^+$  will be referred to as  $\text{NH}_x$  in the following.

The calculation of  $\text{NH}_3$  volatilisation has already been developed in Sect. 2.2.2. The subsequent  $\text{NH}_3$  volatilization flux has been estimated to be  $6 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  in southern Mali, and  $5 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  in southern Niger, whatever the season. This assumption is obviously not exact because of seasonal variations in the absorption and release depending on soil humidity, wind speed, and food quality for cattle. Owing to the lack of measurements in remote Sahelian areas, the lack of statistical data on cattle repartition and nitrogen content leads to many assumptions, implying of course a high level of uncertainty in determining the exact quantity of  $\text{NH}_x$  released. But the relative level of volatilization from excreta compared to other sources of  $\text{NH}_3$  is respected, when compared to emissions at the global scale (Bouwman et al., 1997; Galloway and Cowling, 2002). Figure 6 shows the annual repartition of emission and deposition  $\text{NH}_x$  fluxes. Mean values for each station are given in Table 2. The mean  $\text{NH}_x$  emission flux is  $6.0 \pm 0.5 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  and is dominated by the volatilization flux. The mean deposition flux (dominated by dry deposition processes) is  $5.8 \pm 0.6 \text{ kgN ha}^{-1} \text{ yr}^{-1}$ . These values

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are of the same order as deposition fluxes calculated in a tropical pasture site in Brazil, giving a range of  $\text{NH}_3$  fluxes between 6 and  $12 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  (Trebs et al., 2006).

### 3.4 Annual budget of oxidized and reduced nitrogen compounds at each station, and scaled up to the regional/global scale

Figures 7 and 8 show the total deposition and emission fluxes of oxidized and reduced nitrogen compounds at the annual scale for each station. These figures allow the synthesis between Sect. 3.2 and 3.3, and show that the maximum (wet and dry) deposition and emission fluxes are dominated by the  $\text{NH}_3$  participation, with respectively 66%, 78% and 73% at Katibougou, Banizoumbou and Agoufou stations for wet and dry deposition flux, and respectively 67%, 67% and 78% at Katibougou, Banizoumbou and Agoufou for the emission. Whelpdale et al. (1997) have estimated that the contribution of oxidized nitrogen to the total deposition was about 23% in arid savannas, which corroborates our results.

The second most important emission flux is given by the biogenic NO from soils.

Table 3 summarizes the mean N emission and deposition fluxes at the annual scale for each station. The average deposition flux, attributed to dry savanna ecosystems, is  $9.4(\pm 1.3) \text{ kgN ha}^{-1} \text{ yr}^{-1}$ , and the average emission flux is  $8.5(\pm 1.0) \text{ kgN ha}^{-1} \text{ yr}^{-1}$ . These fluxes are of the same order of magnitude as deposition fluxes in temperate rural site reported by Zhang et al. (2009), ranging from 4.3 to  $11 \text{ kgN ha}^{-1} \text{ yr}^{-1}$ . Deposition fluxes are quite homogeneous from one station to another. Common characteristics (same climate regime, same type of emission sources and amplitude, same type of vegetation and soil characteristics) deduced from these 3 stations, can be attributed to dry savanna ecosystems, and may also be scaled up to the Sahelian region ( $10^\circ \text{N}:20^\circ \text{N}$ ,  $15^\circ \text{W}:10^\circ \text{E}$ , i.e.  $4.6 \times 10^6 \text{ km}^2$ ). Emission characteristics also give common features from one site to another, despite the stronger contribution of  $\text{NH}_3$  volatilization in Agoufou, due to the very weak contribution of biomass burning and biofuel fires in this remote area.

It is therefore possible from these two-way fluxes estimates to give an N annual

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budget of dry savanna Sahelian ecosystems.

Figure 9 gives the mean repartition that should be observed in the Sahel concerning nitrogen compound emission and deposition fluxes. Total deposition has been estimated to range between 3.9 and 5 TgN/yr (with a mean value of  $4.3 \pm 0.6$ , 3.1 TgN/yr attributed to reduced compounds, 1.2 TgN/yr attributed to oxidized compounds). The emission has been estimated to be between 3.6 and 4.5 TgN/yr (mean= $3.9 \pm 0.4$ , 2.7 TgN/yr for reduced compounds and 1.2 for oxidized compounds), for the Sahel region.

As reported by Loveland et al. (2000), savannas, open shrublands and grassland ecosystems are estimated to cover 12.9 millions km<sup>2</sup> in the tropical band (see Table 4 for details), excluding Europe, North America and other countries situated poleward of the tropic of Capricorn and tropic of Cancer. As a consequence, at the global scale, the global nitrogen deposition flux associated with these ecosystems could range from 11.1 to 14.1 TgN/yr (with a mean value of  $12.2 \pm 1.7$ ), with approximately 30% attributed to oxidized compounds, and the other 70% due to NH<sub>x</sub>. The global N emission flux would range from 10.1 to 12.5 TgN/yr (mean= $11.0 \pm 1.3$ ), with the same repartition between oxidized and reduced compounds as for the deposition budget. These values have to be compared to global emission and deposition estimates: global NO<sub>x</sub> emission has been estimated at 44 (30–73) TgN/yr (Galbally et al., 2008), and NH<sub>3</sub> global emission in 1990 has been estimated at 54 (40–70) TgN/yr (Bouwman et al., 1997). Galloway et al. (2008) give a 100 TgN/yr for NH<sub>3</sub>+NO<sub>x</sub> emission in 1995. Our global estimate of N compound emissions for savannas reaches 12.2 TgN/yr, which is around 12% of the estimate of global NH<sub>3</sub>+NO<sub>x</sub> emissions cited above. Furthermore, Bouwman et al. (1997) have estimated the contribution from animal excreta to NH<sub>3</sub> emissions at 21.7 TgN/yr at the global scale, plus 5.9 TgN/yr due to biomass and biofuel combustion. Our estimate of NH<sub>3</sub> emission from volatilization and combustion would give 70% of 12.2 TgN/yr=8.5 TgN/yr in dry savanna ecosystems only. As stated by Davidson and Kinglerlee (1997) and Meixner and Yang (2004), these ecosystems constitute a significant part in the emission of Nitrogen compounds, whereas our knowledge of

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these remote areas is limited by a lack of measurements and studies.

## 4 Conclusions

This study is a first and original attempt to estimate both deposition and emission fluxes of nitrogen species in dry savanna ecosystems, using simulated and calculated inventories, and in situ measurements.

In this study, we have first tried to reproduce the nitrogen oxidized compound emission and deposition evolution month by month, based on simulated NO biogenic emission from soils and measured concentrations, at three different IDAF stations in dry savanna areas during 2006. The magnitude of deposition and emission fluxes is similar, but emission begins later than deposition due to a deficit in the sensitivity of the emission module at the beginning of the rainy season. An annual budget of reduced and oxidized N emission and deposition fluxes has therefore been calculated. It gives a mean estimate of 9.4 (8.6–10.9) kgN ha<sup>-1</sup> yr<sup>-1</sup> for total deposition, dominated by dry deposition (58% of the total), and 8.5 (7.8–9.7) kgN ha<sup>-1</sup> yr<sup>-1</sup> for total emission during the year 2006, dominated by NH<sub>3</sub> volatilization (67%) and biogenic emission from soils (24%), whereas emissions from biomass burning and domestic fires accounts for 9% only. These average fluxes are considered to be representative of dry savanna ecosystems.

If we make the reasonable assumption that dry savanna (i.e.; open grassland and open grassland with sparse shrubs) is representative of the main Sahelian vegetation type (Mayaux et al., 2003), we can scale up those fluxes to the Sahelian region: mean emission in the Sahel could reach 3.9 (3.6–4.5) TgN/yr, whereas mean deposition would be 4.3 (3.9–5) TgN/yr. The difference between emission and deposition gives a net deposition of 0.4 (0.2–0.6) TgN/yr. The uncertainties are numerous, but they are linked to necessary assumptions considering the small amount of data available in this region. Uncertainties correspond to the estimate of the livestock population in the Sahel, to the measurements protocol, to the lack of NH<sub>3</sub> dry deposition

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data in Agoufou, to the deposition velocities assumptions, and to the upscaling of the ANN algorithm. Furthermore, we have assumed that long range transport of nitrogen compounds did not influence local deposition (except wet deposition) at the three studied stations. If applied to other regions, this assumption has to be carefully re-considered. Finally, estimates of emission and deposition flux of nitrogen compounds at the global scale have been calculated for dry savanna ecosystems (mean deposition 12.2 (11.1–14.1) TgN/yr, mean emission 11.0 (10.1–12.5) TgN/yr). These values are in the (upper) range of fluxes already estimated in preceding studies, which increases the robustness of our assumptions, and a high level of confidence in the reliability of the ANN module in dry savanna areas. The improvement of the ANN module for suitability in other West African ecosystems (wet savannas, forests) is foreseen in future research studies.

Furthermore, in order to improve on the assumptions given in this study, in collaboration with specialized teams, further measurements are needed to improve the biogeochemical description of the soils in arid and semi-arid regions. This is needed to better understand the strong release of N compounds at the beginning of the wet season, despite their low N content, and to improve the quantification of N release to the atmosphere.

This study, involving original and unique data from remote and seldom explored regions, will be extended to other West African ecosystems in the future.

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**Table 1.** Seasonal and annual mean emission and deposition fluxes (in  $\text{kgN ha}^{-1} \text{yr}^{-1}$ ) of oxidized nitrogen compounds in dry savanna stations.

Site	Wet season $\text{O}_x \text{ N}$		Dry season $\text{O}_x \text{ N}$		Annual mean $\text{O}_x \text{ N}$	
	Deposition	Emission	Deposition	Emission	Deposition	Emission
Agoufou	$2.6 \pm 1.2$	$2.4 \pm 0.9$	$0.7 \pm 0.1$	$1.2 \pm 0.1$	$2.9 \pm 1.2$	$1.7 \pm 0.8$
Banizoumbou	$2.3 \pm 0.8$	$3.8 \pm 1.1$	$1.0 \pm 0.7$	$2.1 \pm 1.1$	$1.9 \pm 1.0$	$2.7 \pm 1.4$
Katibougou	$2.5 \pm 0.4$	$3.4 \pm 0.8$	$1.1 \pm 0.6$	$2.9 \pm 1.6$	$3.0 \pm 0.9$	$3.2 \pm 1.2$

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**Table 2.** Annual mean emission and deposition fluxes (in  $\text{kgN ha}^{-1} \text{yr}^{-1}$ ) of reduced nitrogen compounds for dry savanna stations.

Site	Annual mean $\text{NH}_x$	
	Deposition	Emission
Agoufou	$7.9 \pm 1.4$	$6.1 \pm 0.1$
Banizoumbou	$6.7 \pm 1.8$	$5.4 \pm 0.3$
Katibougou	$5.7 \pm 1.6$	$6.5 \pm 0.6$

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**Table 3.** Total emission and deposition fluxes (in  $\text{kgN ha}^{-1} \text{yr}^{-1}$ ) of reduced and oxidized nitrogen compounds in dry savanna.

Site	Annual mean	
	Total Deposition	Total Emission
Agoufou	$10.9 \pm 2.7$	$7.8 \pm 0.8$
Banizoumbou	$8.6 \pm 2.7$	$8.1 \pm 1.5$
Katibougou	$8.7 \pm 2.5$	$9.7 \pm 1.6$
Average	$9.4 \pm 1.3$	$8.5 \pm 1.0$

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**Table 4.** Description and area of ecosystems used for N emission estimate calculated in the tropical band (between the tropics of Capricorn and Cancer).

Cover type	Area (km <sup>2</sup> )	Description
Grasslands	2 667 998	Tree and shrub cover $\leq 10\%$ , herbaceous types of cover
Open shrublands	4 397 437	Woody vegetation $\leq 2\text{m}$ with shrub canopy cover between 10–60%
Savannas	5 877 186	Lands with herbaceous and other understorey systems and with forest canopy cover between 10–30%. Forest cover height $\geq 2\text{m}$
Total	12 942 621	
Excluded countries (outside tropics)	North America, Europe, Afghanistan, Algeria, Argentina, Armenia, Aruba, Azerbaijan, Bahamas, Bahrain, Bangladesh, Bhutan, China, Egypt, Georgia, Hong Kong, Iran, Iraq, Israel, Japan, Jordan, Kazakhstan, North and South Korea, Kyrgyzstan, Lebanon, Libya, Mongolia, Morocco, Nepal, Mariana Islands, Pakistan, Saudi Arabia, South Africa, Syria, Taiwan, Tajikistan, Tunisia, Turkey, Turkmenistan, Saudi Arabia, South Africa, Syria, Taiwan, Tajikistan, Tunisia, Uruguay, Uzbekistan, Yemen.	

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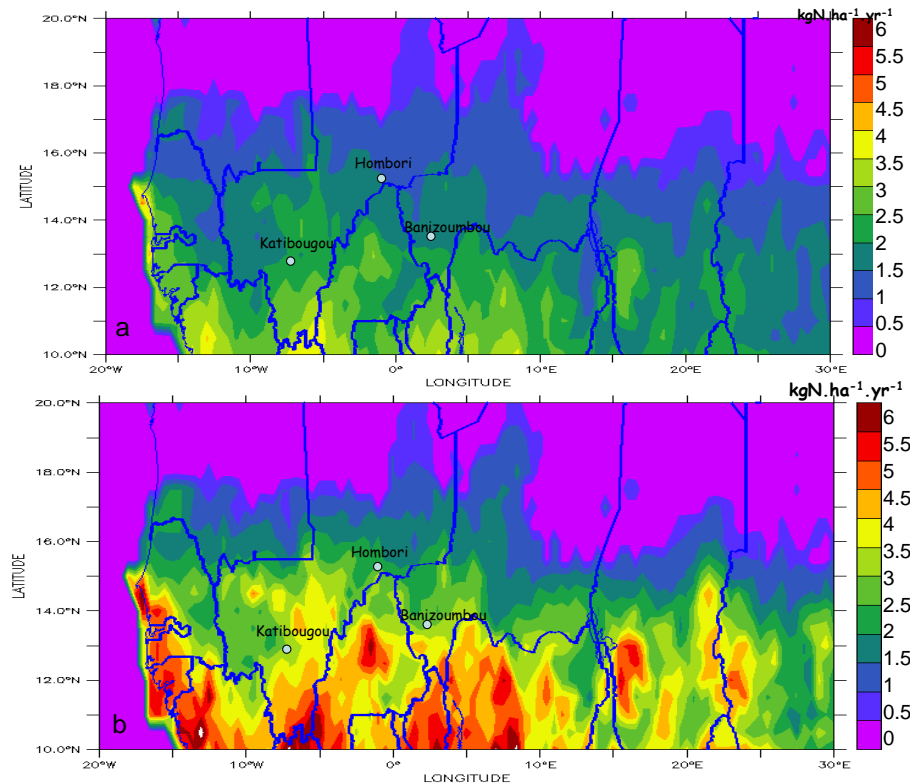
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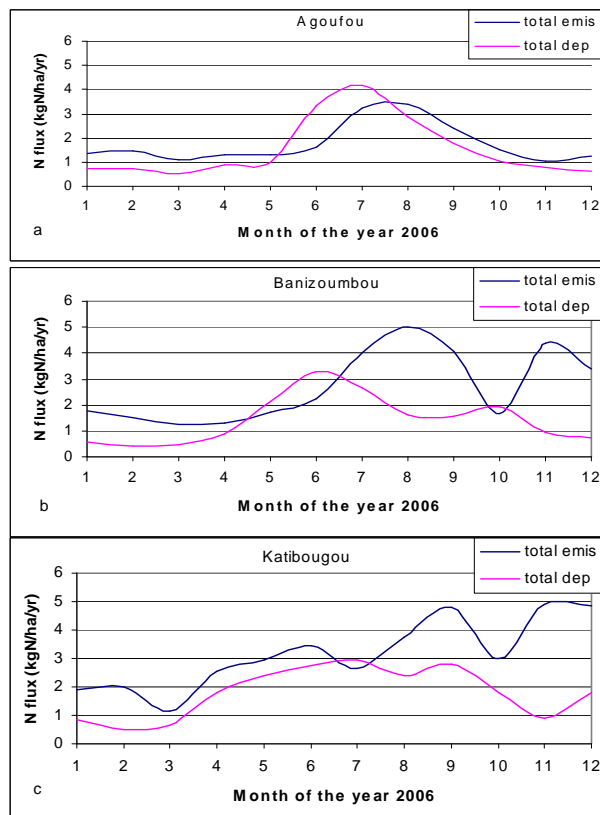


**Fig. 1.** Simulated biogenic NO flux from soils in  $\text{kgN ha}^{-1} \text{yr}^{-1}$  in North West Africa (**a**) annual mean, (**b**) JJAS mean.

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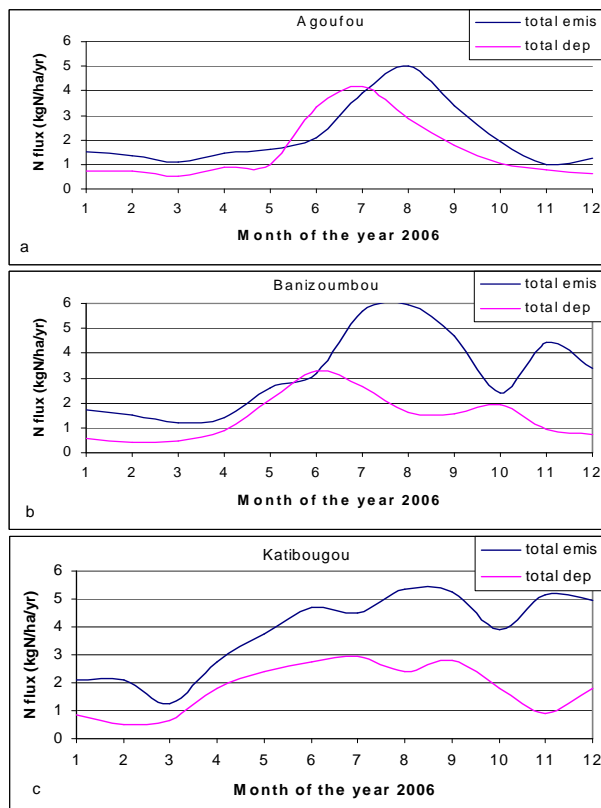


**Fig. 2.** Monthly evolution of deposition flux (purple line, dry deposition of  $\text{NO}_2$  and  $\text{HNO}_3$  + wet deposition of  $\text{NO}_3$ ) and emission flux (blue line, biogenic  $\text{NO}$  emission from soils + biomass burning flux + domestic fires flux) in  $\text{kgN ha}^{-1} \text{yr}^{-1}$  for IDAF dry savanna stations.

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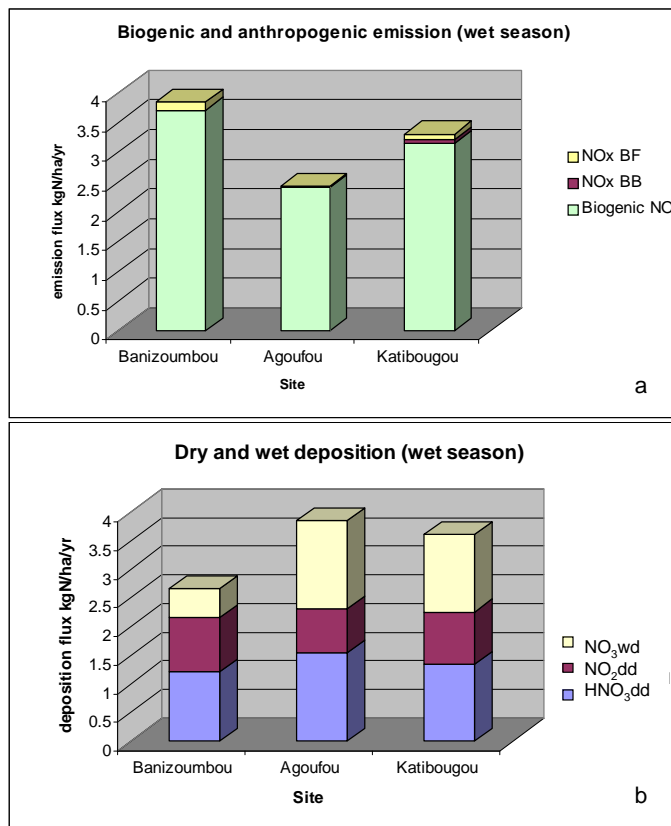


**Fig. 3.** Same as Fig. 2, but with precipitation forcing doubled.

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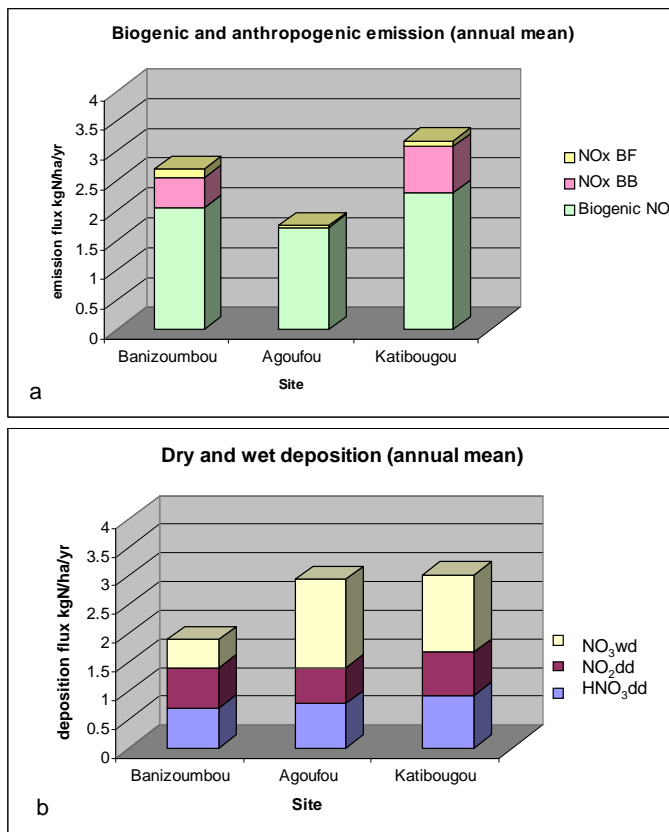
**Fig. 4.** Wet season emission (a) and deposition (b) fluxes in  $\text{kgN ha}^{-1} \text{ yr}^{-1}$  for oxidized nitrogen species at the three dry savanna stations. BF=biofuel, BB=biomass burning, wd=wet deposition, dd=dry deposition.

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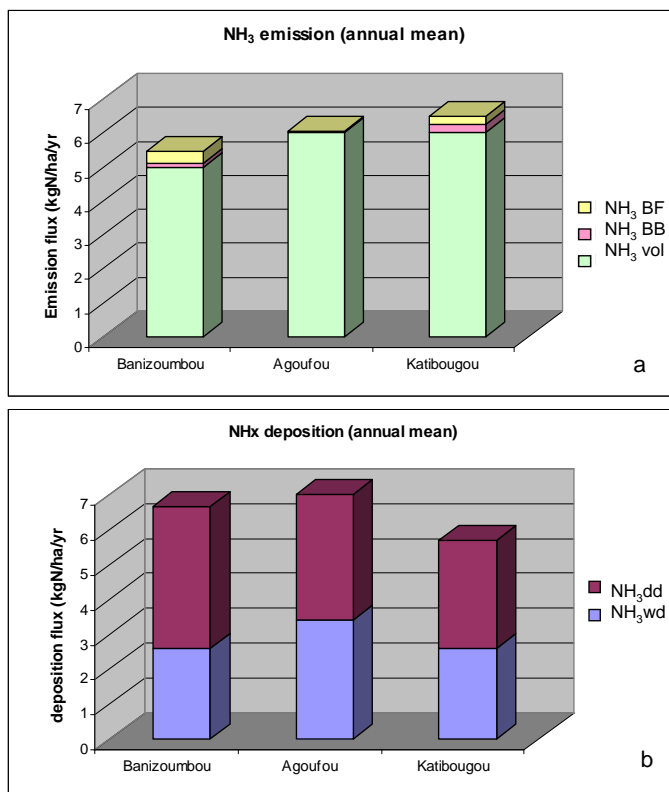


**Fig. 5.** Annual mean emission (a) and deposition (b) fluxes in  $\text{kgN ha}^{-1} \text{yr}^{-1}$  for oxidized nitrogen species at the three dry savanna stations. BF=biofuel, BB=biomass burning, wd=wet deposition, dd=dry deposition.

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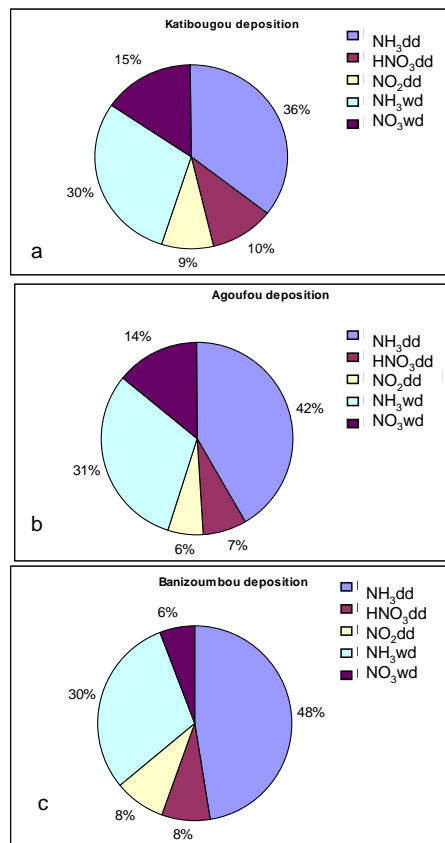


**Fig. 6.** Annual mean emission **(a)** and deposition **(b)** fluxes in kgN ha<sup>-1</sup> yr<sup>-1</sup> for reduced N compounds (NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup>) at the three dry savanna stations. BF=biofuel, BB=biomass burning, wd=wet deposition, dd=dry deposition, vol=volatilization.

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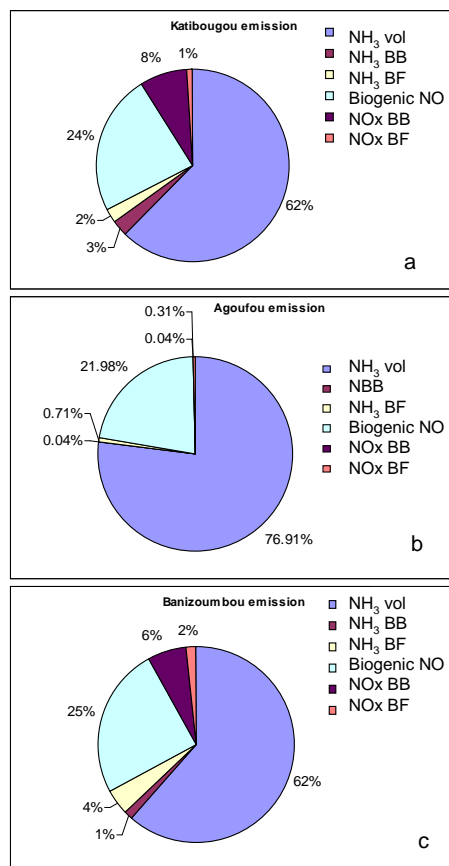


**Fig. 7.** Dry and wet deposition fluxes at (a) Katibougou (b) Agoufou and (c) Banizoumbou of oxidized (NO<sub>2</sub>, NO<sub>3</sub><sup>-</sup>, HNO<sub>3</sub>) and reduced (NH<sub>4</sub><sup>+</sup>, NH<sub>3</sub>) nitrogen compounds.

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**Fig. 8.** Emission fluxes at **(a)** Katibougou **(b)** Agoufou and **(c)** Banizoumbou of oxidized (Biogenic NO, Biomass burning NO<sub>x</sub>, Biofuel NO<sub>x</sub>) and reduced (Biomass burning NH<sub>3</sub>, Biofuel NH<sub>3</sub> and volatilized NH<sub>3</sub>) nitrogen compounds.

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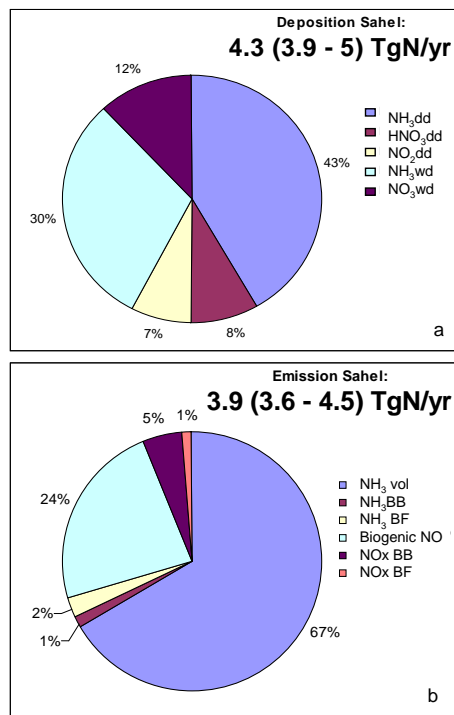
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**Fig. 9.** Mean repartition (and range) of emission and deposition (oxidized and reduced) N fluxes, scaled up at the Sahelian region (4.6 millions km<sup>2</sup>).

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