Biogeosciences Discuss., 8, 7221–7256, 2011
www.biogeosciences-discuss.net/8/7221/2011/
doi:10.5194/bgd-8-7221-2011

Biogeosciences Discussions

Nitrogen compounds emission and deposition in West African ecosystems: comparison between wet and dry savanna

C. Delon[1,2], C. Galy-Lacaux[1,2], M. Adon[1,2,3], C. Liousse[1,2], D. Serça[1,2], B. Diop[4], and A. Akpo[5]

[1]Université de Toulouse, UPS, LA (Laboratoire d'Aérologie), Toulouse, France
[2]CNRS, LA (Laboratoire d'Aérologie), UMR5560, Toulouse, France
[3]Laboratoire de Physique de l'Atmosphére, Abidjan, Côte d'Ivoire
[4]Université de Bamako, Mali
[5]Université Abomey Calavi, Cotonou, Bénin

Received: 28 June 2011 – Accepted: 13 July 2011 – Published: 21 July 2011

Correspondence to: C. Delon (claire.delon@aero.obs-mip.fr)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Surface emission and deposition fluxes of nitrogen compounds have been studied in five sites of West Africa during the period 2002 to 2007. Measurements of N deposition fluxes have been performed in IDAF sites representative of main west and central African ecosystems, i.e, 3 stations in dry savanna ecosystems (from 15° N to 12° N), and 2 stations in wet savanna ecosystems (from 9° N to 6° N). Dry deposition fluxes are calculated from surface measurements of NO_2, HNO_3 and NH_3 concentrations and simulated deposition velocities, and wet deposition fluxes are calculated from NH_4^+ and NO_3^- concentration in samples of rain. Emission fluxes are evaluated including simulated NO biogenic emission from soils, emissions of NO_x and NH_3 from biomass burning and domestic fires, and volatilization of NH_3 from animal excreta. This paper is a tentative to link the variability of rain and the intra and inter annual variability of emission and deposition processes, and to compare these evolutions between dry and wet savanna ecosystems. In dry savanna ecosystems where the rain season lasts mainly from June to September, the occurence of rain correlates with the beginning of emission and deposition fluxes. This link is less obvious in wet savanna ecosystems (wet season mainly from May to October), where the surface is less submitted to drastic changes in terms of water content. Whatever the location, the natural variability of rain from year to year does not exceed 15 %, and does not induce a strong impact on emission and deposition magnitude. Due to the scarcity of available data on the African continent, it is of first importance to combine data from different origins (surface measurements, satellite and modelling) to document the atmospheric Nitrogen cycle in these tropical regions.

1 Introduction

The flux of a trace gas between the soil and the atmosphere is the result of three basic processes: production, consumption and transport. In the case of reactive nitrogen (N_r), production leads to emission of N compounds such as NO and NH_3, and

BGD
8, 7221–7256, 2011

consumption is preceded by transport and deposition of compounds such as gases (NH_3, NO_2, HNO_3, Peroxy Acetyl Nitrates,...), and particles (pNH_4^+, pNO_3^-). The atmospheric nitrogen budget depends on emission and deposition fluxes both as reduced and oxidized compounds. The deposition flux is both driven by wet and dry deposition processes. Direct methods (eddy covariance, gradient method) and indirect methods (inferential method combined to concentrations measurements) are available to determine dry deposition fluxes. The DEBITS (DEposition of Biogeochemically Important Trace Species) committee in charge of deposition studies in IGAC (International Global Atmospheric Chemistry) has decided to use indirect dry deposition fluxes determination in tropical sites, because of difficulties to operate sophisticated direct methods of fluxes measurements in remote sites (Wolff et al., 2010, Sutton et al., 2007). Recording to this, dry deposition fluxes are here estimated using the inferential method, that is combining measurements of concentrations and modelling of deposition velocity according to the resistance analogy (Wesely (1989), Zhang et al. (2003) and references therein).

IDAF (IGAC/DEBITS/Africa) network started in 1995 in tropical Africa. The major objectives of IDAF are to measure wet and dry deposition fluxes, to identify the relative contribution of natural and anthropogenic sources, and the factors regulating these fluxes. The 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 (http://idaf.sedoo.fr).

Natural emissions from soils are produced by microbial processes (nitrification/denitrification), and several modelling approaches have allowed global and regional estimates of NO (Yienger and Levy, 1995, Butterbach-Bahl et al., 2001, Stohl et al., 1996; Steinkamp and Lawrence, 2011) and NH_3 (Bouwman et al., 1997) emissions from soils. A dynamic approach of caculating NO fluxes from soils, has been developed (Delon et al., 2007) and applied in the Sahel region (Delon et al., 2008). Compared to existing inventories, the main advantage of this approach is to be coupled on line with meterological parameters.

BGD
8, 7221–7256, 2011

Anthropogenic emissions of N compounds in the atmosphere have been widely studied, often in temperate regions. At the global scale, reactive N production continues to increase every year, dominated by agricultural activities, fossil fuel energy, as well as biofuels, leading to a global estimate of 187 Tg yr^{-1} in 2005 (Galloway et al., 2008). In tropical regions, the estimate of biogenic N emitted and deposited is less easy to calculate, because of a lack of data.

In this study, a first attempt at estimating the nitrogen emission and deposition fluxes is made for the period 2002 to 2007, through measurements and simulations at five stations of the IDAF network situated in dry and wet savanna ecosystems, as shown in Fig. 1.

In a preceeding work detailed in Delon et al. (2010), the Sahel atmospheric nitrogen budget for the year 2006 was made through measurements and simulations. The method used in the present study is the same as the one detailed in Delon et al. (2010), but applied on a longer period (2002 to 2007) and for 2 more sites located in wet savanna areas. The present work allows a comparison between two different nitrogen emitting ecosystems, for different years and different points at the local scale. The present paper in actually a continuation of Delon et al. (2010), as it uses the same methods for the calculation of fluxes, with the integration of the description of wet savanna emission and deposition processes.

In the following, we will give details on the calculation of N compounds emission fluxes from biomass burning (NO_x and NH_3), domestic fires (NO_x and NH_3), soils (NO) and volatilization (NH_3), and on N compounds dry (HNO_3, NO_2, NH_3) and wet deposition fluxes (NH_4^+ and NO_3^-). In a second time, we will try to give an analysis of inter and intra annual variation of emission and deposition fluxes along a latitudinal transect (from 6.1° N to 15.1° N), for the period 2002 to 2007.

We recall however that all the description of materials and methods have already been written in Delon et al. (2010), and only a brief summary will be given in the following paragraphs.

BGD
8, 7221–7256, 2011

2 Materials and methods

2.1 Presentation of measurement sites

The study will focus on 5 stations of the IDAF network: 3 stations are located in dry savanna: Agoufou, Mali (15.3° N, 1.5° W), Banizoumbou, Niger (13.3° N, 2.4° E), Katibougou, Mali (12.5° N, 7.3° W) and 2 stations are located in wet savanna: Djougou, Benin (9.7° N, 1.7° E) and Lamto, Ivory Coast (6.1° N, 5° W). In Agoufou and Djougou, data have been collected from 2005 till today (but presented here from 2005 to 2007), and in Banizoumbou, Katibougou and Lamto, data are available from 1998 till today (but presented from 2002 to 2007). Comprehensive descriptions of the stations can be found in Mougin et al. (2009) for Agoufou, in Galy-Lacaux and Modi (1998) for Banizoumbou, in Adon et al. (2010) for Katibougou and Djougou, and Yoboué et al. (2005) for Lamto. Fig. 1 gives the geographical location and the dominant ecosystem type of the IDAF stations.

2.2 Emission fluxes

Tropical savannas may be major sources of Nitrogen Oxides (NO) (Cardenas et al. (1994), Poth et al. (1995), Delmas et al. (1991), Serça et al., 1998). However, the vast majority of existing data have been collected in Central and South American ecosystems. In contrast, only few data exist on natural NO emissions from African ecosystem (Serça et al. (1994), Meixner et al. (1997), Scholes et al. (1997), Feig et al., 2008). Furthermore, in West Africa, biomass burning contributes to increase N compounds concentrations in the atmosphere, through NOx and NH_3 emissions (Delmas et al., 1995). NH_3 volatilization is also an important and largely unstudied N loss term (Schaeffer et al. (2003), Bouwman et al., 1997), contributing to N deposition in areas downwind of sources.

BGD
8, 7221–7256, 2011

Each potential source of N compound emission to the atmosphere used in this study is detailed in this paragraph. For more details on the whole calculation procedure, the reader is invited to refer to Delon et al. (2010).

2.2.1 Biomass burning

The global biomass burning inventories for NO_x and NH_3, 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 (Liousse et al., 2010). Emission factors for gaseous species were chosen following Andreae and Merlet (2001). Monthly emissions are evaluated around each site in a 5°/5° window. All the database was exploited here to derive interannual emissions from 2002 to 2007. Note that the biomass density is not a dynamic parameter in the algorithm. The interannual variability of biomass burning is only determined by the variability of burned surfaces. The total uncertainty applied on NH_3 and NO_x fluxes is 54 %.

Long range transport of biomass burning emission compounds from the southern hemisphere does not affect northern sites of dry savanna areas (Mari et al. (2008), Sauvage et al., 2007). This is not the case for wet savanna sites that are situated further south (9.7° N for Djougou, and 6.1° N for Lamto), and that may be impacted by potential dry deposition of N compounds from biomass burning occuring in the southern hemisphere (Thouret et al., 2009) in June–July–August (dry season of the southern hemisphere). Ammonium and nitrate loads in rains in the wet savanna of Lamto has been believed to be due to regional sources (animal waste and emissions by natural soils) as well as biomass burning and domestic fuelwood burning (Yoboué et al., 2005). Biomass burning influence from the southern hemisphere should not be excluded.

2.2.2 Domestic fires

The rural population in Africa relies on wood and charcoal as the main fuels for domestic consumption (de Montalembert and Clement, 1983). In rural areas of Africa, and often among urban population also (Barnes et al., 2001), much of the biofuel is used

BGD
8, 7221–7256, 2011

for cooking all along the year without any seasonal pattern in contrast to other types of biomass burning which tend to follow seasonal patterns (Duncan et al., 2003). The source of NOx from biofuel and field burning is about 10 % of the source from fossil fuel combustion (Yevich and Logan, 2003), with strong variations at the regional scale.

Emissions of NO_x and NH_3 by domestic fires in dry and wet savannas have been evaluated at the yearly scale considering the methodology developed in Junker and Liousse (2008) and Delon et al. (2010). Yearly emissions (typical for 2003) are evaluated around each site in a 5°/5° window. Uncertainties are mainly linked to wood and charcoal consumption estimates (50 %, Assamoi and Liousse, 2010) and emission factors (31 %, Andreae and Merlet, 2001). The total uncertainty applied to domestic fires is 60 % for both compounds.

2.2.3 Biogenic Nitrogen Oxide (NO) from soils

Nitric oxide is a gaseous intermediate of both nitrification and denitrification processes. In sites that receive pulsed precipitation events or extended dry periods, wetting of a previously dry soil often results in a large flux of NO (Davidson et al. (1991), Davidson et al. (1993), Scholes et al. (1997), Martin et al., (1998), Hartley and Schlesinger, 2000).

Biogenic NO fluxes are simulated with the model ISBA (Interaction Surface Biosphere Atmosphere, Noilhan and Mahfouf (1996)), coupled with a Neural Network derived emission module (Delon et al. (2007), Delon et al. (2008), Delon et al., 2010). Emissions depend on surface WFPS (Water Filled Pore Space), soil temperature at two depths (surface and 20–30 cm), wind speed, pH, sand percentage, and fertilization rate. Soil moisture has the prominent influence in dry savanna ecosystems, because of strong pulses of emission at the beginning at the wet season, when the first rains fall on very dry soils (Jaeglé et al. (2004), Stewart et al. (2008), Delon et al. (2008)). In wet savanna, this phenomenon is reduced, due to higher soil moisture content even during the dry season. The uncertainty of the mean annual biogenic NO flux from soil is close to 20 % for each site.

BGD
8, 7221–7256, 2011

Meteorological conditions needed to run the ISBA model are provided by the forcing, derived from satellite data, and developed in ALMIP (AMMA Land surface Model Intercomparison Project), described in Boone et al. (2009).

2.2.4 NH_3 volatilization

Large pulse of NO after wetting of a previously dry soil coincides with an increase in extractable soil NH_4^+ and high rates of gross N mineralization and nitrification (Davidson et al., 1993). Similar to NO production from nitrification, rates of NH_3 volatilization are sensitive to soil conditions that influence NH_4^+ concentrations and turnover in the soil (Barger et al., 2005). An illustration of this process is given in Adon et al. (2010), who show that the seasonal cycle of ammonia and nitric dioxide are strongly correlated. Subsequently, NO and NH_3 emissions are linked and both sensitive to constraints on microbial activity (McCalley et al., 2008). For the remote sites of the IDAF network, neither industrial nitrogen sources nor nitrogenous emissions from synthetic fertilization need to be considered, due to the remoteness of sites from large cities. Bouwman et al. (1997) have stated that synthetic fertilization in West Africa is less than $0.5\,kgNha^{-1}yr^{-1}$, whereas organic fertilization from domestic animal can be much larger, as described below. The NH_3 flux released to the atmosphere by volatilization is calculated from the N input by organic fertilization. This N input is the product of the N release by livestock, in $kgN.animal^{-1}.yr^{-1}$, adapted from Mosier et al. (1998) and Schlecht and Hiernaux (2004), by the Animal population in each region of each country (Food and Agriculture Organization, FAO, Global Livestock Production and Health Atlas GLiPHA (2009, http://kids.fao.org/glipha/). A total N input of $23\,kgNha^{-1}yr^{-1}$ in Agoufou (Mali), $25\,kgNha^{-1}yr^{-1}$ in Banizoumbou (Niger), $11\,kgNha^{-1}yr^{-1}$ in Katibougou (Mali), $8\,kgNha^{-1}yr^{-1}$ in Djougou (Benin) and $8\,kgNha^{-1}yr^{-1}$ in Lamto (Ivory Coast) has been calculated.

30 % of this N input is released to the atmosphere by NH_3 volatilization, the rest is assimilated in soils and used as fertilization rate for the biogenic NO emission in ISBA, described above. As a consequence, the N released by volatilization in Agoufou is

BGD
8, 7221–7256, 2011

7.2 kgNha^{-1}yr^{-1}, 7.7 kgNha^{-1}yr^{-1} in Banizoumbou, 3.4 kgNha^{-1}yr^{-1} in Katibougou, 2.5 kgNha^{-1}yr^{-1} in Djougou and 2.3 kgNha^{-1}yr^{-1} in Lamto.

In the present study, an intra annual variation of NH_3 volatilization has been added, with higher emissions during the wet season. The increase in soil moisture at the beginning of the wet season is correlated to the increase of NO and NH_3 concentrations in the dry savanna sites (Adon et al., 2010). Furthermore, the excreta quantity is not constant throughout the year, and presents a maximum at the end of the wet season, both in Agoufou and Banizoumbou (Hiernaux et al. (1998), Hiernaux and Turner (2002), Schlecht and Hiernaux, 2004), because animals have been better fed during the wet season. The excreta quality is also increased during the wet season, but too few data on the subject are available to be included in our study. Due to a lack of equivalent studies in Katibougou, Djougou and Lamto, we have supposed that the processes were similar than in Agoufou and Banizoumbou, but with a smoother intensity because the available vegetation for grazing is more important in Katibougou, Djougou and Lamto than in Agoufou and Banizoumbou, due to their latitudinal position. The quantity of N released from animal excreta will therefore be maximum in October and November. The combination of these two processes (increase of soil moisture at the beginning of the wet season and increase of excreta quantity at the end of the wet season) gives a monthly evolution of the NH_3 volatilization, showing 2 maxima in June and October.

2.3 Deposition fluxes

2.3.1 Dry deposition fluxes

The dry deposition flux (molec cm^{-2} s^{-1}) is the product of the measured concentration (molec cm^{-3}) by the deposition velocity (cms^{-1}). The deposition velocity is simulated in the ISBA surface model according to Wesely (1989). Cuticle and soil resistances for each site are use the parameterization developed in Zhang et al. (2003) (i.e. soil resistances depend on the type of soil and vegetation found on site). The meteorological forcing used for these simulations is the same as the one used for the biogenic NO

BGD
8, 7221–7256, 2011

emission fluxes calculation (ALMIP forcing). However, the forcing in ALMIP is available at 10 m, whereas the concentrations of gases are measured at 1.5 m. A logarithmic decrease of the wind forcing from 10 to 1.5 m, depending on the rugosity of the site, has been applied, to calculate deposition velocities at 1.5 m.

Monthly means (from 3-hourly values) of deposition velocities for NO_2, HNO_3 and NH_3 have been calculated in order to reproduce the seasonal cycle of the deposition process at each site. Monthly mean values of deposition velocities for HNO_3 range from 0.40 to 1.00 cms^{-1} in Agoufou, 0.42 to 1.11 cms^{-1} in Banizoumbou, 0.49 to 1.13 cms^{-1} in Katibougou, 0.43 to 0.91 cms^{-1} in Djougou and 0.52 to 0.85 cms^{-1} in Lamto. Maximum values are close to mean values calculated in european ecosystems over short vegetation (such as grasslands or peats) by Flechard et al. (2011), showing HNO_3 Vd in the range of 1.0 to 1.2 cms^{-1}. The types of vegetation and soil are completely different between the tropical sites considered in this study and the european sites of Flechard et al. (2011) study, and maximum values are found during the wet season, when the vegetation cover is at its maximum, whereas vegetation in european sites is less subject to such drastic seasonal changes. However, the annual means remain 2 to 3 times lower than deposition velocities calculated by Zhang et al. (2003) for deserts and short grass ecosystems. This discrepancy is mainly due to the fact that deposition velocities in Zhang et al. (2003) are calculated at 20 m above ground, whereas deposition velocities in the present study are calculated at 1.5 m. In that case, direct comparison of deposition velocities is not adequate because of large differences in the wind speeds between the two heights. NO_2 deposition velocities range from 0.13 to 0.35 cms^{-1} in Agoufou, 0.14 to 0.39 cms^{-1} in Banizoumbou, 0.14 to 0.43 cms^{-1} in Katibougou, 0.18 to 0.46 cms^{-1} in Djougou and 0.21 to 0.46 cms^{-1} in Lamto. NH_3 deposition velocities range from 0.14 to 0.41 cms^{-1} in Agoufou, 0.16 to 0.49 cms^{-1} in Banizoumbou, 0.16 to 0.53 cms^{-1} in Katibougou, 0.22 to 0.52 cms^{-1} in Djougou, and 0.26 to 0.54 cms^{-1} in Lamto.

BGD
8, 7221–7256, 2011

Although bi directional air surface exchange of NH_3 has been frequently observed over a variety of land surfaces, the compensation point concept is not applied in this study for the calculation of NH_3 dry deposition flux. Indeed, Zhang et al. (2010) have stated that semi natural vegetation are generally not exposed to large amounts of N and therefore tend to be a sink for NH_3 rather than a source, with compensation point values close to zero. In our study, deposition and emission fluxes of NH_3 are evaluated separately, giving at the end the net flux of NH_3 as it should be done using the compensation point concept.

The concentrations are measured at each site by passive samplers. The sampling procedure and chemical analysis of samples, as well as the validation method, have been widely detailed in Adon et al. (2010). Furthermore, Adon et al. (2010) present the evolution of NO_2, NH_3 and HNO_3 concentrations along the period 1998–2007 for each IDAF station.

The total uncertainty applied to the fluxes is linked to the concentration measurements, the deposition velocity, and to terms associated to the turbulence in the atmosphere and assumptions made to obtain monthly means of deposition velocities. The total rate of uncertainty applied for deposition fluxes is 70 % for NO_2, 31 % for NH_3 and 38 % for HNO_3 (Delon et al., 2010).

2.3.2 Wet deposition fluxes

An automatic precipitation collector, specially designed for the IDAF network was located in the five IDAF sites (Galy-Lacaux et al., 2009). The automatic instrument collects precipitation with a high degree of cleanliness in a single-use polyethylene bag, avoiding aerosol deposit before the onset of the rain. The surface of rain collection is 225 cm^2. After each precipitation event, 50 cm^2 of the collected precipitation is sampled in Greiner tubes. The preservation of the rainwater samples is an important problem because of microbial degradation which can modify the chemical composition of rainwater. All samples were stored in a deep freeze environment from collection to analysis.

BGD
8, 7221–7256, 2011

The wet deposition flux is the product of the ammonium and nitrate concentrations in rain (annual Volume Weighted Mean VWM) by the annual rainfall.

3 Results

3.1 Analysis of processes responsible for emission and deposition of N compounds

Fig. 2 shows the detail of nitrogen compounds emission and dry deposition fluxes, at the 5 IDAF stations, with monthly mean rainfall evolution calculated from TRMM-3B42 data in mm month^{-1}, from 2002 to 2007. The results are presented along a pluviometry gradient, from Agoufou, the northernmost position with only 482 (± 81) mm of annual rain, Banizoumbou (664 (± 86) mm), Katibougou (720 (± 117) mm), Djougou (1115 (± 150) mm), to Lamto, the southernmost position, with 1100 (± 101) mm. Annual means and standard deviations of precipitation are calculated for the 2002–2007 period, from TRMM-3B42 data in a 5° window around each site. Compared to local measurements, we found that TRMM-3B42 data overestimates local measurements by 25% in dry savanna areas, and underestimates them by 15 % in wet savannas. TRMM-3B42 precipitation data have also been used for the simulation of deposition velocities and NO emission from soils in ISBA.

3.1.1 Ammonia release

Oxidized (Fig. 2a–e) and reduced (Fig. 2f–j) N compounds deposition and emission fluxes in monthly means are presented separately to better understand the sources influence.

In theory, more rain will allow an increase in the vegetation available for grazing, i.e. a better quality and higher quantity of food for animals, which will induce more N in animal manure, more NH_3 from volatilization, and therefore more dry deposition of NH_3 (Fig. 2f–j). This volatilization of NH_3 will be retrieved in the rains, and scavenged

BGD
8, 7221–7256, 2011

by the wet deposition, which will further increase the NH_4^+ availability in soils, and again the NH_3 volatilization.

In dry savanna sites (Agoufou, Banizoumbou and Katibougou), Fig. 2 shows that a first maximum of emissions is found at the beginning of the rain season (both for reduced and oxidised compounds), due to the starting up of the microbial activity consecutive to higher soil moisture. A rebound is observed in NH_3 dry deposition fluxes in October, due to the increase of N content in cattle dung at the end of the wet season. A third increase (almost visible in Djougou and Lamto) is observed during the dry season, because of biomass burning emissions (except in Agoufou, where regional biomass burning sources do not affect the site). NH_3 volatilization and further deposition show a strong seasonnal variation in dry savanna areas, beacuse rates of NH_3 volatilization are sensitive to soil moisture conditions, as explained above. The interannual variability of NH_3 volatilization cannot be discussed here, because data are given for a single year and reproduced each year in the same way. In wet savannas, the ammonia release is mostly due to biomass burning emissions, discussed below. Because of a higher soil water content all along the year, seasonal variations in NH_3 volatilization and further deposition are less marked.

3.1.2 Biogenic emissions from soils and microbial activity

While different in magnitude, several processes are common to dry and wet savanna ecosystems: If rain increases during the wet season, the biogenic NO emission increases, as well as the Oxidized N dry deposition fluxes (Fig. 2a–e). The difference between wet and dry savanna ecosystem in their capacity to release biogenic NO from soils may find some explaination in microbial processes: microbial activity has been studied in Lamto and Banizoumbou by Serça et al. (1998), showing very different carbon mineralization potential and potential net accumulation of ammonium between the two sites (very low in Banizoumbou, maximum in Lamto), whereas nitrate accumulation was equivalent in the two sites. That study did not find any significant correlation between nitrate accumulation and NO emissions.

BGD
8, 7221–7256, 2011

The type of vegetation growing on site has also an important effect on biogenic NO emission from soils: nutrient poor savanna ecosystems of Agoufou and Banizoumbou (and in a less extent in Katibougou) are characterized by low denitrification potentials and relatively high nitrate accumumation. Thus substantial NO emissions in dry savanna ecosystems are most likely related to nitrification processes, whereas low potential of nitrification and denitrification rates in Lamto may be partly responsible of low NO fluxes (Leroux at al., 1995). Low nitrification rates in wet savanna ecosystem could be due to inhibition of nitrifier growth and/or functionality by specific compounds excluded by the roots of the graminea (Munro , 1966). The main herbaceaous vegetation on site in Lamto (Serça et al., 1998)) and Djougou (J. Harris and Z. Sevigona, personal communication) is *hyparrhenia*, and is considered as a nitrifier inhibitor, which could also explain the lower biogenic NO emissions during the wet season in those 2 sites compared to dry savanna emissions.

Indeed, in the five sites studied in this work, biogenic NO emission fluxes are less important in wet savanna only if the wet seasonal mean is calculated: wet seasonal means give $3.2 \pm 0.6\,\mathrm{kgNha^{-1}yr^{-1}}$ in dry savanna sites, and $2.6 \pm 0.7\,\mathrm{kgNha^{-1}yr^{-1}}$ in wet savanna sites. No pulse effect occurs in wet savannas because of higher mean soil water content. Dry season emissions are nearly 0 in dry savanna sites, whereas they keep being significant in wet savanna sites. Consequently, the annual mean for dry savanna sites is only 1.39 ± 0.28 whereas it is 2.12 ± 0.20 in wet savannas.

Surface soils in arid and semi arid ecosystems often experience large seasonal fluctuations in moisture content, and the mineralization pulse associated with soil drying/rewetting has the potential to significantly impact N release, whereas comparatively in soils maintained at equivalent mean soil moisture, the N turnover is decreased (Millet et al., 2004).

Furthermore, dry savanna ecosystems are characterized by a strong annual vegetation cycle (see FAPAR curves given for the Sahel region in Galy-Lacaux et al., 2009), whereas wet savanna ecosystems present perennial vegetation. These particular features involve different consumption and release of N in the soils, leading to

BGD
8, 7221–7256, 2011

less emissions to the atmosphere in more vegetated areas, whereas more N is lost to the atmosphere in dry savannas before the growing of the vegetation, at the beginning of the wet season. The results found in this work confirm these processes if we focus on wet seasonal means.

3.1.3 Biomass burning

If rain is sufficient, the vegetation will grow during the wet season, and NO_x and NH_3 emission from biomass burning should increase during the following dry season.

In dry savanna ecosystems, NO_x emissions from biomass burning is twice lower than NO biogenic emission from soils. Indeed, the significant pulse effect occuring at the beginning of the wet season allows high biogenic emissions, whereas in contrast low vegetation density is available in the Sahel region (almost null in Agoufou where no biomass burning emission is measured) leading to a low amount of burned biomass. Biogenic NO emissions from soils and NO_x emissions from biomass burning in annual means represent respectively $1.39 \pm 0.28\,kgNha^{-1}yr^{-1}$ and $0.61 \pm 0.52\,kgNha^{-1}yr^{-1}$ in dry savanna ecosystems.

Some particular features are observed in wet savanna ecosystems: the vegetation density is more important in Djougou and Lamto, which leads to NO biogenic emission from soils equivalent to the NO_x biomass burning emission ($2.12 \pm 0.20\,kgNha^{-1}yr^{-1}$ and $2.20 \pm 0.40\,kgNha^{-1}yr^{-1}$ respectively in annual means). Furthermore, NO emission from soil is enhanced after fire occurence, as mentionned by Serça et al. (1998), NO fluxes are the result of the product of site and fire effect. Indeed, this process could be explained by the immediate impact of fire on nitrogen cycling, with an increase of the nitrification activity (Menaut et al., 1993). Furthermore, it has been shown that fire seldom decreases total soil N pools. N volatilization losses caused by frequent burning in savannas are often balanced by N inputs in non-associative N fixation, or N deposition in rain (Coetsee et al., 2010). This may explain the higher biogenic NO emission in wet savanna than in dry savanna in annual mean.

BGD
8, 7221–7256, 2011

In dry savannas, the NH_3 biomass burning emission signal in Fig. 2f–h (around november-december, detected through NO_x biomass burning emission in Fig. 2a–c) is flooded by the volatilization signal (up to 11 $kgNha^{-1}yr^{-1}$ in Agoufou and Banizoumbou, and 5 $kgNha^{-1}yr^{-1}$ in Katibougou). The annual mean for volatilization is 6.09 ± 2.31 $kgNha^{-1}yr^{-1}$ in dry savanna, whereas the NH_3 biomass burning emission is 0.40 ± 0.35 $kgNha^{-1}yr^{-1}$.

On the contrary, wet savanna ecosystems present a higher biomass burning emission signal of NH_3 (Fig. 2i–j), which can reach 12 $kgNha^{-1}yr^{-1}$ in Lamto and 6 $kgNha^{-1}yr^{-1}$ in Djougou as maximum values, and gives an annual mean of 1.91 ± 0.56 $kgNha^{-1}yr^{-1}$, whereas volatilization reaches 4 $kgNha^{-1}yr^{-1}$ as a maximum value in both sites (2.40 ± 0.09 $kgNha^{-1}yr^{-1}$ in annual mean). Total N emission from biomass burning in annual mean (NO_x+NH_3), is 1.02 ± 0.87 $kgNha^{-1}yr^{-1}$ in dry savanna ecosystems, and 4.11 ± 0.96 $kgNha^{-1}yr^{-1}$ in wet savannas.

Domestic fires emissions do not present any interannual variability and annual means are 0.29 ± 0.20 $kgNha^{-1}yr^{-1}$ in dry savanna, and 0.97 ± 0.02 $kgNha^{-1}yr^{-1}$ in wet savanna, which represents respectively 3 % and 10 % of the total emission budget.

All annual means emission fluxes are reported in Table 1.

3.2 Variability of emission and deposition fluxes and comparison between wet and dry savanna sites

Africa leads all other continents in annual fire emissions but with relatively small interannual variability (van der Werf et al. (2006), Liousse et al. (2010)) at the continental scale. However, in our results we consider a regional scale (5°around each local site), and a smaller time scale (7 years) than a climatic one (50 years considered in van der Werf et al. (2006) study). The availability of biomass (and consequently the quantity of resulting emissions by biomass burning) seems to be driven by the amount of rainfall during the wet season before. Indeed, a lagged correlation between monthly rainfall and monthly emission from biomass burning N compounds has been calculated, giving

BGD
8, 7221–7256, 2011

a 4 months lagged correlation rate of 0.7 in Banizoumbou, 0.6 in Katibougou and 0.6 in Djougou. In Lamto, the lagged correlation is 0.5 after 6 months. The lag is longer in Lamto, probably because of the longer duration of the rain season, and the correlation slightly lower because of the occurence of the second short wet season (around the month of October), not connected to a second burning season.

Interannual variability in soil water stress tends not to be highest in places that have the largest interannual variability in precipitation but rather at intermediate values (like in Soudano-Sahelian regions where Katibougou is situated), as water ceases to be a limiting factor with increasing rainfall despite continued increase in rainfall variability (Williams et al. (2008), their study focusing on the 1982–2003 period over the African continent). Low vegetation density and humidity stress may probably dampen the eco-physiological sensitivity to rainfall variability. Williams et al. (2008) have shown that Lamto and Djougou sites are situated in regions presenting a higher interannual variability in rainfall than in Sahelian regions, but with low impact on the interannual variability of soil water stress and vegetation cover.

At a more local scale, Mougin et al. (2009) have illustrated the fact that in the Gourma region (Mali) where Agoufou is situated, most of the inter-annual variation in annual herbaceous production result from inter-annual variation in the soil moisture regime driven by rainfall volume and distribution. Biogenic NO emissions are highly sentitive to soil water content (directly influenced by rainfall amount), which is somewhat different from soil water stress, i.e.: the soil water content gives the information on the water available for emission processes, whereas soil water stress gives an information on a threshold under which microbes do not activate N production. Biogenic NO emissions decrease/increase if rainfall decreases/increases.

In our results, no significant common tendency appears between interannual variability in rainfall intensity and interannual variability in deposition fluxes quantity. Fig. 3 shows the evolution of total dry deposition fluxes (NO_2+HNO_3+NH_3) in $kgNha^{-1}yr^{-1}$ for dry and wet savannas, together with local rain measurements (in $mm.h^{-1}$). In dry savanna, the correlation between rainfall and total deposition is quite low but remains

BGD
8, 7221–7256, 2011

significant (0.51 at Agoufou, 0.33 at Banizoumbou and 0.34 at Katibougou), but does not allow to conclude on a possible intensity effect: i.e. the occurence of rain and deposition are linked, but if the magnitude of precipitations increases, it does not necessarily involve an increase of the intensity of deposition fluxes. Indeed, as explained before, deposition depends on emission processes, enhanced by rainfall events. The change in rainfall intensity from one year to another does not lead to a direct change in deposition flux intensity.

In wet savanna, correlation between rainfall and deposition fluxes is close to 0, confirming the analysis that processes are less linked to the quantity of rainfall than in dry savanna. Once again, the interannual variability in rainfall quantity does not involve a direct response of emission and deposition processes. No interannual variability has been introduced for the volatilization of NH_3 and for the emission of NO_x and NH_3 by domestic fires (data are given for a single year), which is probably the main reason for not being able to isolate common tendencies between rainfall quantity and N emission/deposition amounts. Furthermore, the short period of study (7 years) does not help the interpretation.

Figure 4a–b presents annual means of emission and deposition contributions for the years 2002 to 2007 in Banizoumbou, Katibougou and Lamto, and for the years 2005 to 2007 for Agoufou and Djougou. Error bars give the order of magnitude of the interannual variability for each component of the budget. As mentionned above, interannual variability is not specified for NH_3 volatilization and for NH_3 and NO_x emission by domestic fires (biofuel), because for these parameters, data are given for a single year. However, it could be useful to recall that uncertainty is 50 % for NH_3 volatilization, and 60 % for biofuel emission.

In Fig. 4a, significant differences appear between sites: Sahelian sites (Agoufou and Banizoumbou) present the highest values of NH_3 volatilization, consequence of highly developed pastoralism in this region. Katibougou is located further south, and the number of cattle headskm^{-2} is less important (respectively 24, 23 and 11 cattle headskm^{-2} in Banizoumbou, Agoufou and Katibougou), leading to lower values of

BGD
8, 7221–7256, 2011

NH_3 volatilization. In wet savanna ecosystems, Djougou and Lamto (respectively 8 and 7 cattle headskm^{-2}) present moderate values of NH_3 volatilization fluxes (respectively 2.5 and 2.3 $kgNha^{-1}yr^{-1}$), comparable to other sources. Indeed, the contribution of N compounds emissions from biomass burning in Djougou and Lamto (respectively 1.9 and 2.5 $kgNha^{-1}yr^{-1}$ for NO_x and 1.5 and 2.3 $kgNha^{-1}yr^{-1}$ for NH_3) is more important than in dry savanna sites. Biomass burning and volatilization contribution to total emission flux are inverted along the latitudinal gradient from north to south, as the available vegetation is more important, and the number of cattle headskm^{-2} is less important.

In Fig. 4b, the dry deposition flux of oxidized and reduced compounds is more important in Agoufou, Banizoumbou and Katibougou than in Djougou and Lamto, whereas the wet deposition flux increases from dry to wet savanna ecosystems. Wet deposition fluxes are correlated to rain amount and concentrations in precipitations. Obviously, wet deposition flux is larger if the rainfall is stronger and if the concentrations are more important. Annual means of wet deposition fluxes ($NH_4^+ + NO_3^-$) are 2.35 ± 1.88, 2.14 ± 0.67, 3.40 ± 0.75, 3.53 ± 0.70, and 5.33 ± 0.97 $kgNha^{-1}yr^{-1}$ in Agoufou, Banizoumbou, Katibougou, Djougou and Lamto respectively. One should note that in spite of the large difference in precipitation quantity between the two extremes of this gradient, Agoufou (392 mm) and Lamto (1301 mm), the wet deposition flux is only twice in Lamto. Rain in Agoufou presents high concentrations in nitrate and ammonium (respectively 14 and 25 μeql^{-1}), larger than those measured in Lamto (9 and 21 μeql^{-1} of nitrate and ammonium respectively). The combination of annual rainfall depth and measured concentrations leads to a nitrogen wet deposition flux in Lamto only twice the one in Agoufou. The large quantity of rainfall, and above all the longer duration of the rain season in Lamto has a dilution effect on chemical species in rain.

The contribution of different sources of emission, together with the contribution of wet and dry deposition are presented in Fig. 5a–e for each site. In Agoufou, Banizoumbou and Katibougou (dry savannas), dry deposition is superior to wet deposition. The contribution of wet deposition increases in Djougou and Lamto, for the reasons invoked above. In Agoufou and Banizoumbou, as well as in Djougou and Lamto, emission

BGD
8, 7221–7256, 2011

fluxes are superior to deposition fluxes, by 3 %, 8 %, 6 % and 2 % respectively. In Katibougou, deposition fluxes are 7 % superior to emission fluxes. Considering the range of uncertainty on total emission (45 %) and deposition (25 %) fluxes, these differences in the budgets in all sites are not significant enough, but could however lead to the conclusion that emission and deposition fluxes are quasi equivalent in wet and dry savannas, with a slight tendency for emission fluxes to be superior to deposition fluxes. The case of Katibougou is a transitional case: Katibougou is located at 12° N, at the boundary between Sahel and Soudano-Sahelian region. The vegetation description is close to Sahelian vegetation, but the quantity of rainfall is more important. Wet and dry deposition fluxes are both substantial, and dominate sligthly total emission fluxes.

However, the variability in rainfall amount from year to year does not explain the interannual variability of wet deposition in one single ecosystem. As mentioned above, rainfall variation from year to year does not exceed 15 % in the period 2002–2007, whereas wet deposition variation exceeds this rate in all stations. The complexity and retroactions between emission and deposition contributions prevents from finding an easy and direct link from rainfall to wet deposition. As detailed in Galy-Lacaux and Modi (1998), Galy-Lacaux et al. (2009) and Yoboué et al. (2005), wet deposition annual variations are mainly related to the combination of the natural rainfall and emissions sources variability. If the annual rainfall in each ecosystem does not explain variability in wet deposition, it should be interesting to study the distribution of the rain in each year during each rain season, to have a better understanding of wet depostition distribution. But this is beyond the scope of this study.

The compilation of all components of Fig. 4 and Fig. 5 leads to Fig. 6, presenting the atmospheric reactive N budget in all stations referenced in this study. Total uncertainty is 45 % for emission fluxes, and 25 % for deposition fluxes. One has to keep in mind that this budget is not exhaustive, because measurements do not include all N compounds. Non reactive N compounds (N_2, N_2O, N_2O_5) are not included in the budget (we only consider the atmospheric reactive N compounds), and organic compounds such as PAN are also omitted from this budget, assuming that they are converted into

BGD
8, 7221–7256, 2011

NO_2 at high temperatures (Munger et al., 1998). Particulate compounds are not taken into account in the dry deposition flux calculation, because concentrations measured on sites are negligible compared to gas concentrations (Galy-Lacaux, personnal communication). However, major reactive N compounds sources and sinks are included to try to balance the budget.

Figure 6 shows that total emission and deposition quantities are in the same order of magnitude for each site, with deposition fluxes of $7.25 \pm 2.60\,kgNha^{-1}yr^{-1}$ in Agoufou, $8.16 \pm 1.84\,kgNha^{-1}yr^{-1}$ in Banizoumbou, $8.87 \pm 1.59\,kgNha^{-1}yr^{-1}$ in Katibougou, $7.07 \pm 0.98\,kgNha^{-1}yr^{-1}$ in Djougou, $9.39 \pm 1.07\,kgNha^{-1}yr^{-1}$ in Lamto. Emission fluxes are $8.29 \pm 3.88\,kgNha^{-1}yr^{-1}$ in Agoufou, $11.45 \pm 5.20\,kgNha^{-1}yr^{-1}$ in Banizoumbou, $6.58 \pm 2.52\,kgNha^{-1}yr^{-1}$ in Katibougou, $9.11 \pm 3.14\,kgNha^{-1}yr^{-1}$ in Djougou, and $10.09 \pm 4.11\,kgNha^{-1}yr^{-1}$ in Lamto. Though budgets seem equivalent from dry to wet savanna, this study has shown that a detailed description of processes of emission and deposition leads to different contribution of each component of the budget in the two types of ecosystems.

4 Conclusions

This study is based on original and unique data from remote and seldom explored regions. Satellite data, surface measurements and modelling results were merged to estimate the atmospheric reactive N cycle in West Africa, in five stations of the IDAF network: 3 stations in dry savanna ecosystem, 2 in wet savanna ecosystem, situated along a rainfall gradient from 15° N to 6° N. The N cycle in dry savanna is closely linked to the precipitation cycle: N is stored in the soil during the long dry season, and released to the atmosphere during the wet season (almost at the beginning) in the form of NO and NH_3, and further wet and dry deposited in the form of NH_3, HNO_3, NO_2, NH_4^+ and NO_3^-. Wet and dry deposition are mainly associated to biomass burning, domestic fires, biogenic NO emissions and NH_3 volatilization. In wet savanna the moisture content in soils is less variable because the rainfall amount is more important, and dry deposition is mostly influenced by biomass burning emissions. Wet

BGD
8, 7221–7256, 2011

deposition increases along the rainfall gradient from Agoufou to Lamto, mainly due to the quantity of rainfall. Fluxes are linked to the rainfall (in occurrence and quantity), and indirectly to the growing of the vegetation, through the N cycle in soils and in the atmosphere. It is therefore crucial in this part of the world to correctly represent the amount and occurrence of rain to impulse the right emissions and explain the subsequent deposition processes. No obvious tendency could have been assessed between the rainfall variability from year to year and the corresponding variability in N deposition and emission loads. Rainfall interannual variability has been evaluated to be 15 % at the most, whereas total emission flux inter annual variability is maximum 18 % (taking into account variable fluxes only), dry deposition flux variability ranges between 15 and 28 %, and wet deposition variability is around 20 % for every site. The short period of the study (6 years only), and above all the fact that the major contribution in the emission budget is held in constant fluxes (NH_3 volatilization), prevented to find a common tendency between the quantity of rainfall and the magnitude in emission and deposition fluxes across the years. Total budget gives emission and deposition loads between 6 and 11 $kgNha^{-1}yr^{-1}$, dominated by NH_3 whatever the type of ecosystem. Total emission and deposition fluxes are in the same range whatever the site, but the contribution of each emission source and each deposition process is different between dry and wet savanna. This study has allowed a comparison between emission and deposition processes responsible for N load in the atmosphere in two different types of ecocystems of west Africa, in regions where few data are available.

The publication of this article is financed by CNRS-INSU.

BGD
8, 7221–7256, 2011

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BGD
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BGD

8, 7221–7256, 2011

Table 1. Emission fluxes in annual mean, in $kgNha^{-1}yr^{-1}$ for dry and wet savanna ecosystems. BB = Biomass Burning, BF = Biofuel.

Emission flux	Dry savanna	Wet savanna
Biogenic NO	1.39 ± 0.28	2.12 ± 0.20
NO_x BB	0.61 ± 0.52	2.20 ± 0.40
Volatilization	6.09 ± 2.31	2.40 ± 0.09
NH_3 BB	0.40 ± 0.35	1.91 ± 0.56
NO_x+NH_3 BB	1.02 ± 0.87	4.11 ± 0.96
NO_x+NH_3 BF	0.29 ± 0.20	0.97 ± 0.02

BGD
8, 7221–7256, 2011

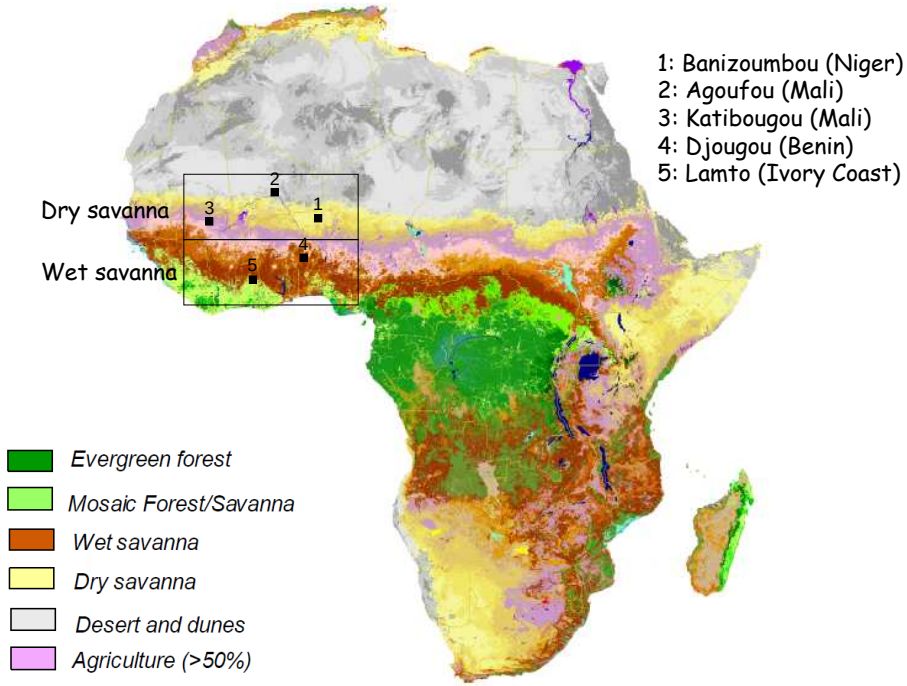

Fig. 1. Location of the stations involved in the IDAF network. Only the five first stations are included in the present study.

BGD
8, 7221–7256, 2011

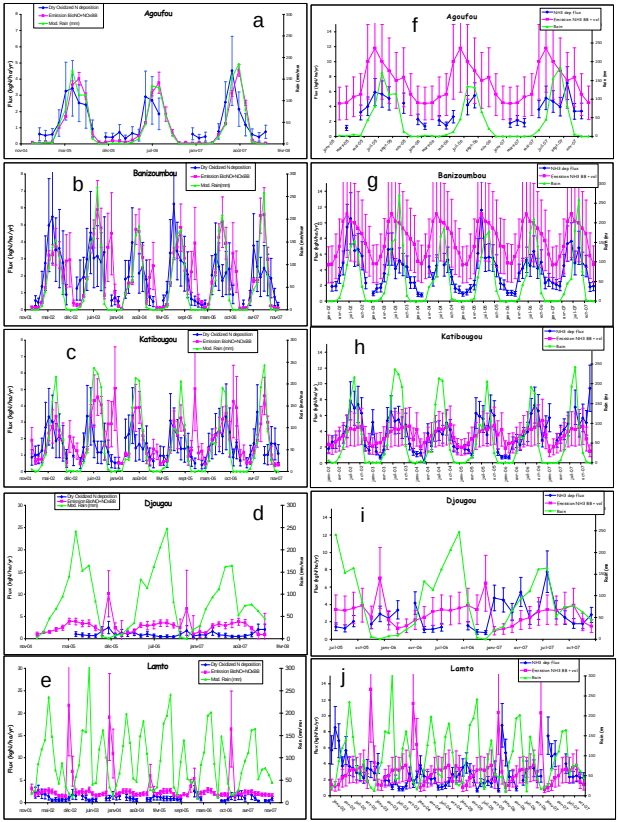

Fig. 2. Left: Interannual evolution of oxidised N compounds (Biogenic NO from soils + NO_x emission from biomass burning in pink, NO_2 + HNO_3 dry deposition in blue, rainfall in green) and Right: Interannual evolution of reduced N compounds (NH_3 volatilization + NH_3 emission from biomass burning in pink, NH_3 dry deposition in blue, rainfall in green), in Agoufou **(a, f)**, Banizoumbou **(b, g)**, Katibougou **(c, h)**, Djougou **(d, i)**, and Lamto **(e, j)**.

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8, 7221–7256, 2011

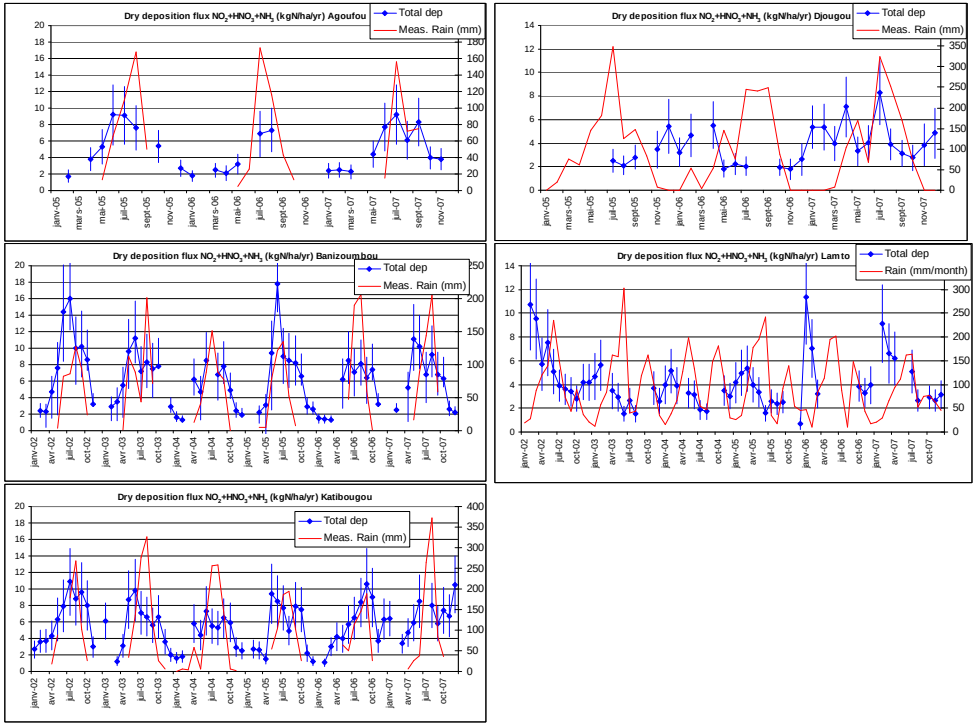

Fig. 3. Total dry deposition flux (NH_3 + NO_2 + HNO_3) in $kgNha^{-1}yr^{-1}$ and measured rain ($mmmonth^{-1}$) at Agoufou, Banizoumbou, Katibougou, Djougou and Lamto.

BGD
8, 7221–7256, 2011

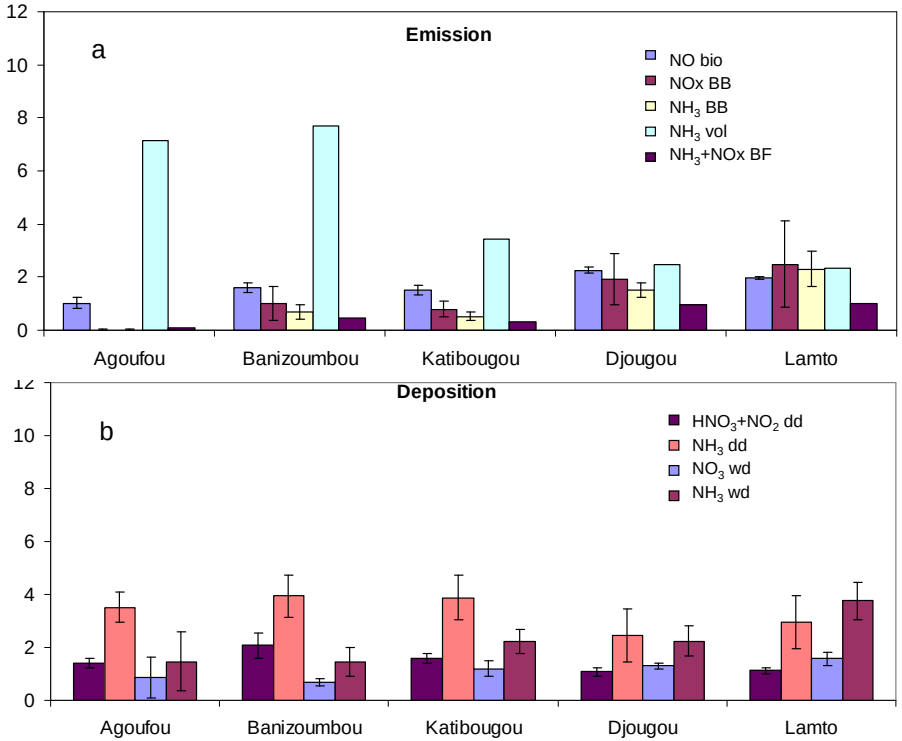

Fig. 4. Emission **(a)** and deposition **(b)** components of the atmospheric N budget in annual mean in $kgNha^{-1}yr^{-1}$ for the five IDAF stations for the years 2002 to 2007. bio = biogenic, BB = biomass burning, wd = wet deposition, dd = dry deposition, vol = volatilization, BF = biofuel. Error bars represent interannual variability. No error bars appear for NH_3 volatilization and BF because of no interannual variability available for these data.

BGD
8, 7221–7256, 2011

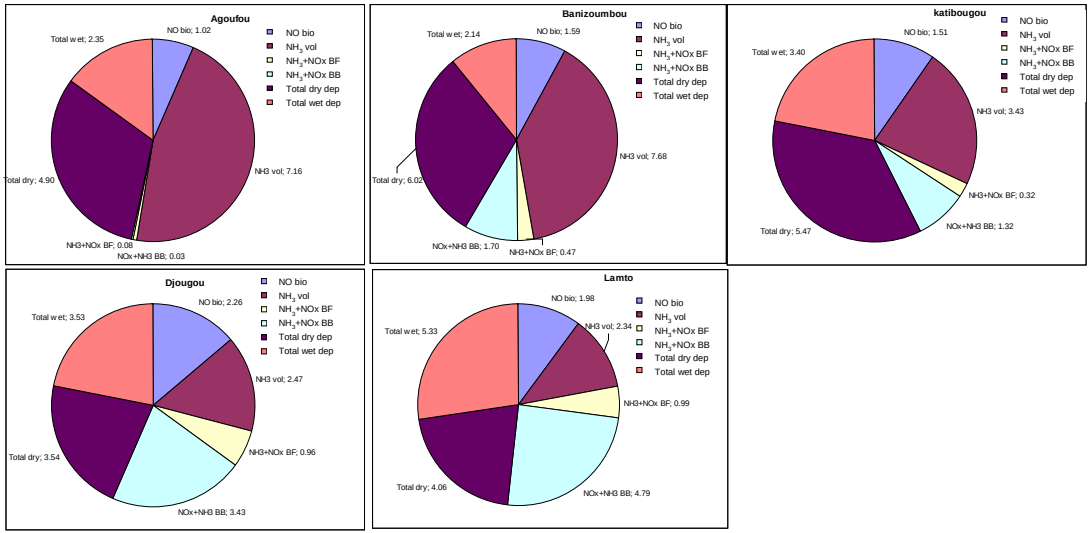

Fig. 5. Contribution of emission and deposition N fluxes in $kgNha^{-1}yr^{-1}$ for five stations in dry and wet savanna ecosystems. Relative uncertainties are specified in the text

BGD
8, 7221–7256, 2011

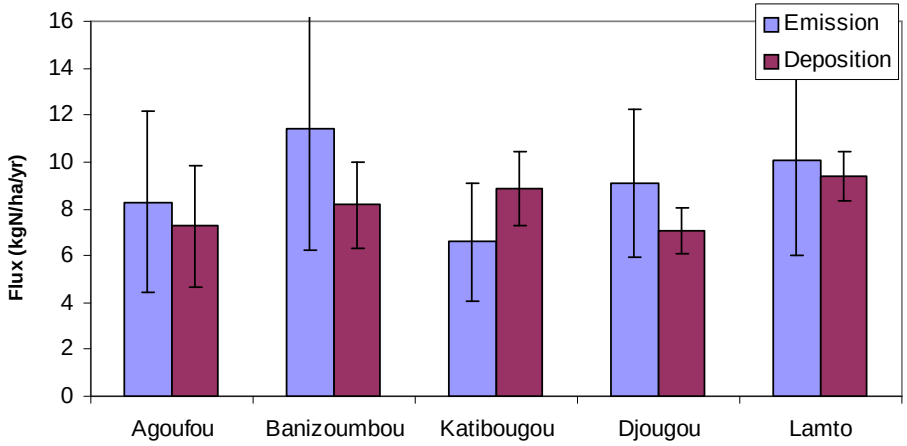

Fig. 6. Emission and deposition N budget in $kgNha^{-1}yr^{-1}$ for five stations in dry and wet savanna ecosystems. Error bars represent the total uncertainty of fluxes.

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8, 7221–7256, 2011