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Dry deposition of nitrogen compounds (NO_2, HNO_3, NH_3), sulfur dioxide and ozone in West and Central African ecosystems using the inferential method

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Abstract

This work is part of the IDAF program (IGAC-DEBITS-AFRICA) and is based on the long term monitoring of gas concentrations (1998–2007) established on seven remote sites representative of major African ecosystems. Dry deposition fluxes were estimated by the inferential method using on one hand surface measurements of gas concentrations (NO_2, HNO_3, NH_3, SO_2, and O_3) and on the other hand simulated dry deposition velocities (V_d). V_d were calculated using the big-leaf model of Zhang et al. (2003b). In the model of deposition, surface and meteorological conditions specific to IDAF sites have been adapted in order to simulate V_d representative of major African ecosystems. The monthly, seasonal and annual mean variations of gaseous dry deposition fluxes (NO_2, HNO_3, NH_3, O_3, and SO_2) are analyzed.

Along the latitudinal transect of ecosystems, the annual mean dry deposition fluxes of nitrogen compounds range from 0.4 ± 0.0 to $0.8 \pm 0.2\,kg\,N\,ha^{-1}\,yr^{-1}$ for NO_2, from 0.7 ± 0.1 to $1.0 \pm 0.3\,kg\,N\,ha^{-1}\,yr^{-1}$ for HNO_3, and from 2.3 ± 0.8 to $10.5 \pm 5.0\,kg\,N\,ha^{-1}\,yr^{-1}$ for NH_3 over the study period (1998–2007). The total nitrogen dry deposition flux ($NO_2 + HNO_3 + NH_3$) is more important in forests (11.2–11.8 $kg\,N\,ha^{-1}\,yr^{-1}$) than in wet and dry savannas (3.4–5.3 $kg\,N\,ha^{-1}\,yr^{-1}$). NH_3 dominated nitrogen dry deposition, representing 67–80 % of the total. The annual mean dry deposition fluxes of ozone range between 11.3 ± 4.7 and $17.5 \pm 3.0\,kg\,ha^{-1}\,yr^{-1}$ in dry savannas, 17.5 ± 3.0 and $19.2 \pm 2.9\,kg\,ha^{-1}\,yr^{-1}$ in wet savannas, and 10.6 ± 2.0 and $13.2 \pm 3.6\,kg\,ha^{-1}\,yr^{-1}$ in forests. Lowest O_3 dry deposition fluxes in forests are correlated to low measured O_3 concentrations, lower of a factor of 2–3, compared to others ecosystems. Along the ecosystem transect, annual mean of SO_2 dry deposition fluxes present low values and a small variability (0.5 to 1 $kg\,S\,ha^{-1}\,yr^{-1}$). No specific trend in the interannual variability of these gaseous dry deposition fluxes is observed over the study period.

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1 Introduction

Deposition of chemical species onto the earth's surface plays an essential role in controlling the concentration of gases and aerosols in the troposphere. The study of deposition thus allows for tracing the temporal and spatial evolution of atmospheric chemistry and is a pertinent indicator for evaluating natural and anthropogenic influences. The deposition of atmospheric nitrogen (N) species constitutes a major nutrient input to the biosphere. On a long-term scale, the increase of N inputs into terrestrial or aquatic ecosystems leads to important environmental consequences such as a loss of biodiversity, eutrophication, acidification of semi-natural ecosystems, leaching of nitrate into groundwater, and increased carbon storage (Vitousek et al., 1997; Rodhe et al., 2002; Bobbink et al., 1998; Bouwman et al., 2002b; Liu et al., 2013). N deposition on the terrestrial surface thus impacts both atmospheric chemistry and ecosystem dynamics. Ammonia (NH_3), nitrogen dioxide (NO_2) and nitric acid (HNO_3) are the most important contributors to N dry deposition (Trebs et al., 2006). Sulfur dioxide (SO_2) is one of the important species considering the acid deposition issues, and it is also the precursor of sulfate. Dry deposition estimation of SO_2 is essential to assess ecological impact research, crop growing and air quality research (Tsai et al., 2010). Tropospheric ozone (O_3) is known to harm human health, damage vegetation and lead to deterioration of materials. The dry deposition of O_3 is one of the most important sinks in the boundary layer ozone budget (Rummel et al., 2007).

Monitoring networks have been established around the world to measure wet and dry deposition. The international program DEBITS (Deposition of Biogeochemically Important Trace Species) was initiated in 1990 as part of IGAC/IGBP (International Global Atmospheric Chemistry/International Geosphere-Biosphere Programme) "core project" in order to study wet and dry atmospheric deposition in tropical regions (Lacaux et al., 2003). The DEBITS network collects data from 25 stations that are distributed within the tropical belt in Africa, Asia and South America, and results are presented in the new IGAC structure or DEBITS II (Pienaar et al., 2005; Bates et al., 2006;

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http://debits.sedoo.fr). For tropical Africa, the IDAF (IGAC/DEBITS/AFRICA) project started in 1994 and was implemented in partnership with INSU (Institut National des Sciences de l'Univers, in France) and the CNRS (Centre National de la Recherche Scientifique, in France) as part of the Environmental Research Observatory (ORE, in France) networks.

The main 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. IDAF activity is based on high quality measurements of atmospheric chemical data (gaseous, precipitation and aerosol chemical compositions) on the basis of a multi-year monitoring (http://idaf.sedoo.fr). Within the framework of IDAF, several studies of precipitation chemical composition representative of great African ecosystems have been recently published (Galy-Lacaux and Modi, 1998; Galy-Lacaux et al., 2001, 2009; Al-Ourabi and Lacaux, 2002; Lacaux et al., 1993, 2003; Sigha et al., 2003; Yoboue et al., 2005; Mphepya et al., 2004, 2006; Laouali et al., 2012).

To complement these studies, it is appropriate to study and quantify dry deposition fluxes. Direct methods (eddy correlation, chamber method) and indirect methods (inferential method, gradient method) are available to determine dry deposition fluxes (Seinfeld and Pandis, 2006). The DEBITS committee in charge of deposition studies in IGAC has decided to use indirect dry deposition fluxes determination in tropical sites, because of difficulties to operate sophisticated direct methods of flux measurements in remotes sites (Wolff et al., 2010; Sutton et al., 2007). In this study, dry deposition fluxes are estimated using the inferential method, that is a combination between gaseous concentration measurements and modeling of deposition velocities according to the resistance analogy (Wesely, 1989; Zhang et al., 2003b; and references therein). The authors are however aware that the inferential method is not the best one to evaluate NH_3 and NO_2 fluxes. Bidirectional exchange of NH_3 and NO_2 have been frequently observed over different canopies (Dorsey et al., 2004; Trebs et al., 2006; Walker et al., 2006; Wichink Kruit et al., 2007). There have been some effort in the development of bidirectional exchange models (e.g., Sutton et al., 1998; Flechard et al., 1999; Trebs

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et al., 2006; Massad et al., 2010; Zhang et al., 2010; Wichink Kruit et al., 2012; Bash et al., 2013; Hamaoui-Laguel et al., 2013); however, none of these models can be directly applied over the different canopies of African ecosystems owing to the lack of necessary input parameters.

In a recent paper, Adon et al. (2010) have presented the long term monitoring of ambient gaseous concentrations within the IDAF program. Our study presented an original database of ten years of measurements (1998–2007) of five important atmospheric gases (NO_2, HNO_3, NH_3, O_3, SO_2) obtained within the IDAF African network at seven sites in West and Central Africa.

In the present paper, realistic dry deposition velocities according to the sites and the species involved are calculated in order to estimate dry deposition fluxes. The big-leaf model of Zhang et al. (2003b) is used to simulate dry deposition velocities representative of major African ecosystems. The results will be compared to previous estimates developed in Delon et al. (2010, 2012). Then, we present an estimate on a long term basis (10 yr) of dry deposition fluxes of gases (NO_2, HNO_3, NH_3, O_3 and SO_2) at the scale of major African ecosystems. The monthly, seasonal and annual mean variations of gaseous dry deposition fluxes are analyzed.

2 Sites description and methodology

2.1 Presentation of measurement sites

Figure 1 presents the location of the 7 IDAF measurement stations displayed on the map of African biomes adapted from the land cover product of Mayaux et al. (2004). The IDAF sites of West and Central Africa are located to represent a transect of ecosystems, i.e., dry savannas (Agoufou, Banizoumbou, Katibougou) - wet savannas (Djougou, Lamto) - equatorial forests (Zoetele, Bomassa). The geographical, ecological and climatic characteristics of the study sites are presented in Table 1. Dry savannas are characterized by a long dry season from October to May and a short wet season

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from June to September. The mean wet season extends from April to October in wet savannas and from March to November in forests; other months are the dry season. A detailed description of IDAF monitoring stations can be found in Adon et al. (2010).

Monitoring of gases at Banizoumbou, Katibougou, Lamto, Zoetele and Bomassa began in 1998. NO_2, NH_3 and HNO_3 have been monitored since 1998, while measurement of O_3 started in 2001 and that of SO_2 in 2002. As part of the Long term Observation Period of the AMMA (African Monsoon Multidisciplinary Analysis) program, the Djougou and Agoufou sites started operating in 2005. All measurements are still continuing at all the IDAF sites.

2.2 Dry deposition estimate

The inferential method, i.e., an atmospheric species dry deposition flux being estimated as a product of its air concentration by modelled dry deposition velocity, was employed in this study to estimate the dry deposition fluxes of different gaseous species. The inferential technique has been widely used in other studies for different types of ecosystems (Shen et al., 2009; Pineda and Venegas, 2009; Jin et al., 2006; Zhang et al., 2005, 2009; Delon et al., 2010, 2012; and Pan et al., 2012). This approach is more suited when routine monitoring data are available but the values of the derived fluxes are clearly dependent on the validity of the dry deposition velocity calculation.

2.2.1 Atmospheric concentration measurements

Atmospheric concentrations of NO_2, HNO_3, NH_3, O_3, and SO_2 are measured by passive samplers on a monthly basis as part of the IDAF network. Passive sampling observations at the West and Central African sites have been performed over a ten year period from 1998 to 2007. The sampling procedure and chemical analysis of samples, as well as the validation method according to international standards, have been widely detailed in Adon et al. (2010). To give an indication of the precision of this sampling technique, the covariance of all duplicate samples over the studied period were found

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to be 20 %, 9.8 %, 14.3 %, 16.6 % and 10 % for HNO_3, NO_2, NH_3, SO_2 and O_3 respectively. Furthermore, Adon et al. (2010) present the evolution of NO_2, HNO_3, NH_3, O_3, and SO_2 concentrations along the period 1998–2007 for each station. Table 2 presents a synthesis of the mean annual gas concentrations for the IDAF sites of West and Central Africa (Adon et al., 2010).

2.2.2 Modeling of the dry deposition velocity for each IDAF site

Dry deposition velocities (V_d) were calculated using the big-leaf dry deposition model of Zhang et al. (2003b). The main parameterizations of the resistances in the model are briefly presented in Appendix A. The main input parameters of this updated deposition model are physiological (parameters of land use categories), biophysical (LAI) and meteorological data.

In general, parameters of inferential models have been largely derived from European and North American studies and may not necessary be adequate for African (or tropical) climate, vegetation and soil conditions (Fowler et al., 2009). Based on ecological and climatic characteristic of African ecosystems, we tried to adapt the parameters of land-use categories (LUC) described in Zhang et al. (2003b) to IDAF specific sites for the calculation of canopy resistances, i.e., cuticle resistance (R_{cut}), stomatal resistance (R_{st}) and in-canopy aerodynamique resistance (R_{ac}) in the big-leaf model (Table 3). Note that the types of LUC described in Zhang et al. (2003b) have one vegetation layer while the savannas (shrub, tree or woody) have two vegetation layers. As a first approximation, the shrub and tree savannas were assigned to grassland LUC (long grass), which is the dominant vegetation type in these areas. In the model of Zhang et al. (2003b), values for all parameters required for calculating R_{st} are taken from literature including Brook et al. (1999) which describes a type of savanna vegetation. Thus, for the tree and shrub savannas (Lamto, Djougou and Katibougou), we included the savanna LUC parameters of Brook et al. (1999) to calculate R_{st} in the deposition model. These parameters are r_{smin} (minimal stomatal resistance), b_{rs} (empirical light response coefficient), b_{vpd} (water-vapour-pressure-deficit constant), Ψ (leaf water

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potential), T_{min}, T_{max} and T_{opt} (minimum and maximum temperatures at which stomatal closure occurs and optimum temperature for maximum stomatal opening).

For the soil resistance (R_g), Zhang et al. (2003b) suggested some reference values based on previous studies and on a review of published measurements in the literature. A R_g value of 200 s m^{-1} is given for O_3 for all vegetated surfaces and 500 s m^{-1} for non-vegetated surfaces (like desert). For SO_2, R_g values depend on the soil wetness (rain, dew or not) and the values of the soil resistance over land surfaces where no dew or rain (R_{gd}) has occurred can be found in Zhang et al. (2003b). In our study, we used the values of soil resistance suggested by Zhang et al. (2003b) for IDAF sites excepted for the Sahelian domain. Considering the semi-arid climate of Sahel, near the Sahara (desert) and the steppe vegetation, we assumed a value of 400 s m^{-1} for R_g (O_3) and R_{gd} (SO_2) for the sites of Agoufou and Banizoumbou and a value of 300 s m^{-1} for the site of Katibougou. For the wet savannas (Djougou, Lamto), we admitted a value of 200 s m^{-1} for R_g (O_3) and R_{gd} (SO_2) and for the forested sites (Zoetele and Bomassa), a value of 200 s m^{-1} for R_g (O_3) and 100 s m^{-1} for R_{gd} (SO_2) has been set. Values of 50 and 100 s m^{-1} are assigned to the soil resistances when rain or dew has occurred respectively for all sites.

Roughness length (Z_0) is needed for calculating friction velocity, which subsequently affects aerodynamic, quasi-laminar and non-stomatal resistances (Zhang et al., 2003b). Rather than taking approximate values from the literature for the IDAF savanna sites, we have used Z_0 values already simulated on these specific sites in the frame of a previous study made with the SVAT (Soil Vegetation Atmosphere) model ISBA (Interactions between Sol, Biosphere and Atmosphere, Noilhan and Mahfouf (1996), Noilhan et al., 1989) and explicited in Delon et al. (2010).The mean Z_0 values simulated are 0.02 m for Agoufou, 0.06 m for Banizoumbou, between 0.13 and 0.16 m for both Katibougou and Djougou, and between 0.14 and 0.2 m for Lamto. For forested ecosystems (Zoetele and Bomassa), we used a Z_0value equal to 2.5 m for tropical forest taken from Zhang et al. (2003b).

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LAI is an important parameter for calculating canopy resistances. For the IDAF sites, LAI was obtained from MODIS (MODerate Resolution Imaging Spectroradiometer) satellite data for the period 2000–2007. To that end, we used the 1 km MODIS LAI values which are processed over an 8 day period (Yang et al., 2006). The MODIS LAI product has already been validated with field measurements on many sites over the western African region (Fensholt et al., 2004; Samain et al., 2008). The mean seasonal variations of the monthly LAI during 2000–2007 for IDAF sites are shown in Fig. 2. The mean monthly LAI range from 0.2 to 1 $m^2 m^{-2}$ for Sahelian sites (Agoufou and Banizoumbou), from 0.5 to 2 $m^2 m^{-2}$ for both Katibougou and Djougou sites and from 2 to 4 $m^2 m^{-2}$ for the Guinean savanna site of Lamto. For the forested site represented by Bomassa, the monthly LAI fluctuates around the average value of 5 $m^2 m^{-2}$. A strong seasonality is observed in the savannas and high values of the standard deviation of monthly LAI value occur on dense canopies of forest and wet savanna of Lamto. A sensitivity test showed a low variability of monthly V_d when the value of LAI increases by 0.5 or 1 according to the sites (Adon, 2011). The MODIS LAI of IDAF specific sites are well comparable to the ones of the corresponding land covers of the ECOCLIMAP database (Kaptue et al., 2010). For our simulation of dry deposition velocity, we used the monthly LAI averaged over 8 yr (2000–2007) for savanna sites and a constant value of 5 $m^2 m^{-2}$ for tropical forest sites. We note that Zhang et al. (2003b) and Brook et al. (1999) have used a constant LAI of 6 $m^2 m^{-2}$ and 4.5 $m^2 m^{-2}$ respectively to simulate dry deposition velocity for tropical forests.

Meteorological data required for the simulation are wind speed and temperature at a reference height in the surface layer, surface temperature, solar irradiance at the surface, precipitation, surface pressure, relative humidity and cloud cover fraction. Excepted cloud cover fraction, meteorological conditions are provided by the forcing developed in the frame of ALMIP (AMMA Land Surface Model Intercomparison Project) from satellite data, described in Boone et al. (2009). The spatial resolution of this forcing is 0.5° × 0.5° with a 3 h temporal resolution. For the simulation of dry deposition velocity, we used a database of 6 yr from 2002 to 2007. Validations of surface temperature

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and moisture have been made within the ALMIP project (Boone et al., 2009; De Rosnay et al., 2009). However, the forcing in ALMIP is available at 10 m, whereas the concentrations of gases are measured at around 2 m for the savanna sites and 3 m for the forested ecosystems within IDAF network. As a first approximation, a logarithmic decrease of the wind forcing from 10 m to 2 m, depending on the rugosity of the site, has been applied, to calculate deposition velocities at 2 m for the savanna sites, as described in Delon et al., (2010, 2012). This approach to reduce the wind from 10 m to the height of 2 m is based on the constant flux assumption (Baldocchi, 1988; Zhang et al., 2009). However for the forested ecosystems, the constant flux assumption cannot be applied below the canopy due to extra sink terms; hence another approximation is done and described below in Sect. 2.2.3.

Surface wetness controls non-stomatal resistances for soluble trace gases. The big-leaf model of Zhang et al. (2003b) predicts surface wetness semi-mechanistically and distinguishes dew from rain, based on precipitation data for rain and on night-time cloud cover and friction velocity for dew formation (Janssen and Romer, 1991; Brook et al., 1999). In this study, as the cloud cover information was missing, we performed sensitivity tests for the presence of dew by using the constant relative humidity (RH) threshold method for estimating dew condensation (Sentelhas et al., 2008). Various RH thresholds have been used as proxies to determine canopy wetness; Tsai et al. (2010) used a constant value of RH = 90 % as the threshold for wetness presence over evergreen broadleaf forest in Taiwan, while for grassland Wichink Kruit et al. (2008, 2010) suggest a smaller threshold at 71 %. Flechard et al. (2011) chose canopy wetness on the basis of a wet-dry threshold of 81 % when key meteorological variables were missing. Thus, we have performed sensitivity tests using a threshold of 90 % for forest, 81 % for wet savanna and 71 % for dry savanna to test the presence of dew. Results showed that the variation of monthly V_d is negligible when it is assumed that dew has occurred; for example the RMSE (Root Mean Squared Error) of V_d (SO_2) is 2 %, 1 %, 4 % for dry savanna, wet savanna and forest, respectively (Adon, 2011). Therefore in this study, we do not take into account the assumption of the occurrence of dew. In addition, V_d

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simulations under dew or dry conditions in Zhang et al. (2003b) showed that V_d under dew conditions are not necessarily larger than under dry conditions.

Previous studies of nitrogen budget in wet and dry savanna ecosystems have proposed an estimate of N compounds deposition velocities (Delon et al., 2010, 2012). In those studies, V_d were calculated according to Wesely (1989) resistance analogy in the Soil Vegetation Atmosphere Transfer (SVAT) big leaf model ISBA, with modifications of the cuticle and ground resistances adapted from Zhang et al. (2003b). The results were obtained at the regional scale at 0.5° resolution, for the period 2002–2007, and focused on the reactive nitrogen compounds budget (with estimates of emission and deposition (dry + wet) fluxes). The present study is focused on dry deposition only, and integrates all the parameterizations developed by Zhang et al. (2003b), which gives a different but consistent and coherent estimate of V_d at the local scale, and for more species (same N compounds + O_3 and SO_2). Indeed, this study where we investigate each specific vegetal cover representative of each IDAF measurement sites leads to a more precise understanding of deposition processes at the local scale. Differences between the two approaches are mainly due to the different models resolution, and to the degree of details involved in the estimation of input parameters such as the ones detailed in the above paragraph. The previous studies of Delon et al. (2010, 2012) give a point of comparison for N compounds V_d calculated in the present work, as very few studies are available for African ecosystems.

We present in this paper the calculation, over the period 2002–2007, of monthly means (from 3-hourly values) deposition velocities for O_3, SO_2, NO_2, HNO_3 and NH_3 for the IDAF sites.

2.2.3 Corrections for within-canopy concentration data for forests

The inferential method requires atmospheric concentrations and turbulence intensity above the canopy to predict rates of dry deposition over the forest. In the IDAF network, gas concentrations are measured at about 3 m in forests. Besides, there are very few published within-canopy vertical gas concentration profiles (HNO_3, NH_3) in

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the literature for forest (Flechard et al., 2011). Thus, we have carried out a pilot experiment by measuring simultaneously gas concentrations at 10 m and 3 m (or 2 m) from ground in the forested ecosystem of Zoetele (and in the wet savanna of Lamto and dry savanna of Banizoumbou) over the period of September 2010 to December 2011.

For the forested ecosystem, NO_2, HNO_3 and SO_2 monthly concentrations measured simultaneously at 10 m and 3 m show no significant trend and are of the same order of magnitude. Over this period, mean annual concentrations, at 3 m and 10 m respectively, are 0.81 ppb and 0.80 ppb for NO_2, 0.2 ppb and 0.3 ppb for HNO_3, and 1.8 ppb and 1.6 ppb for SO_2. However for NH_3 and O_3, monthly concentrations measured at 10 m are higher and the mean ratio was 1.5 for NH_3 and 1.3 for O_3 (mean annual concentration are 3.6 ppb and 6.1 ppb for NH_3, and 5.4 ppb and 7.0 ppb for O_3 at 3 m and 10 m respectively). This observation is consistent with the approximation made in Flechard et al. (2011) showing that NH_3 concentrations measured in clearings and below canopy are consistently smaller than above treetops; they thus applied a constant correction factor of 1.3 for NH_3 concentrations measured below trees. We note that in the dry savanna of Banizoumbou, NH_3 concentrations measured at 2 m (annual mean 8.1 ppb) are higher than at 10 m (6.3 ppb), indicative of NH_3 sources in the ground and in the leaf litter in grasslands (Nemitz et al., 2009). For O_3, our observations are consistent with ozone vertical profile concentrations that decrease towards the ground, as observed by Krzyzanowski (2004) by measuring ozone concentrations at different heights in a forest canopy.

In a first approximation, for forested sites of IDAF (Zoetele and Bomassa), we thus applied a constant correction factor of 1.5 for NH_3 and 1.3 for O_3 concentrations measured at 3 m in order to calculate the dry deposition flux at 10 m. For the other gases (NO_2, HNO_3 and SO_2) we have not applied any correction. In addition, the mean covariance of samplers exposed simultaneously at 3 m and 10 m were found to be 13 %, 25.5 % and 21 % for NO_2, HNO_3 and SO_2, respectively, and these values are comparable of the mean reproducibility of IDAF passive samplers calculated from 1998 to 2007 (9.8 %, 20 % and 16.6 % for NO_2, HNO_3 and SO_2, respectively). So, the small

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difference in measured concentrations between the two heights (3 and 10 m) for NO_2, HNO_3 and SO_2 could be included in the uncertainty calculated for the passive sampler method.

2.3 Uncertainties in dry deposition fluxes estimates

Uncertainties in the estimated dry deposition fluxes result from combined uncertainties in measured gaseous concentrations and in modelled dry deposition velocities. In this section, rather than quantifying the total uncertainty of the inferential method in the study, we focus on addressing the uncertainties for each contribution of the dry deposition estimates. Uncertainties linked to the measurement of gas concentration using IDAF passive samplers have been given by the covariance of duplicates (reproducibility), between 10 and 20 % according to the species (Adon et al., 2010). The main uncertainty of the dry deposition velocities is mainly related to the wind forcing. The difference between the wind in the forcing and the wind measured in situ is between 5 and 35 % depending on the site. A mean rate of 20 to 30 % according the site is applied for the wind speed uncertainty, leading to an uncertainty rate for the dry deposition velocity between 10 and 20 % for all sites. Others uncertainties are related to the representation of the soil type in the model indirectly dependent of the roughness length and the soil resistances (Zhang et al., 2003b), and the choice of plant physical parameters.

In this study, concentrations are measured with passive samplers and are monthly integrated. Dry deposition velocities are simulated 3-hourly and then monthly averaged. As we use monthly means for concentrations and deposition velocities, the covariance between the two may induce an additional uncertainty (the missing covariance term), especially for species having strong diurnal variations, in the range of $\sim$ 20 % (Matt and Meyers, 1993; Zhang et al., 2005).

Bidirectional exchange of NH_3 between atmosphere and vegetation is not included in the model of Zhang et al. (2003b). It could be another uncertainty but is assumed to be low for semi natural vegetation (Zhang et al., 2009, 2010).

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Additional uncertainties are associated with different parameterizations of dry deposition used in deposition models. The physical, biological and chemical exchange mechanisms involved in deposition processes are too complex to be explicitly and completely modeled, so that parameterizations tend to be empirical in the models (Flechard et al., 2011; Schwede et al., 2011). Wesely and Hicks (2000) reported an uncertainty of about 30 % for all the dry deposition models.

Direct measurements of dry deposition fluxes are technically difficult to implement and expensive, so dry deposition velocities coupled with concentration measurements are often used. Few long-term monitoring networks measure dry deposition fluxes. However, the Canadian CAPMON, American NADP, European EMEP and Asia EANET networks use the inferential method as the IDAF network in Africa to estimate dry deposition fluxes.

3 Results and discussion

3.1 Dry deposition velocities

Monthly means (from 3-hourly values) of deposition velocities for O_3, SO_2, NO_2, HNO_3 and NH_3 have been calculated, over the period 2002–2007, in order to reproduce the seasonal cycle of the deposition processes at each site (Fig. 3). We note a fairly clear distinction between the different ecosystems and climatic domains. The monthly V_d values of each gas increase from the semi-arid savannas (Agoufou, Banizoumbou) to the equatorial forested ecosystems (Zoetele, Bomassa) indicating that the dry deposition velocity increases with the vegetation density. As a first approximation, along the African ecosystem transect, V_d gradient thus follows the climatic gradient. In fact, there exists a marked latitudinal gradient over the West and Central African zone. Areas with the highest rainfall have the greatest volume of biomass or primary productivity; as a consequence, surface deposition (cuticle) and stomatal uptake become important,

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the canopy resistance (R_c) is the main factor determining the deposition rate in vegetative zones (Tsai et al., 2010).

The monthly variation of NO_2 V_d follows the same pattern than O_3 but with slightly smaller values. The deposition of NO_2 and O_3 is mostly controlled by stomatal uptake. NH_3V_d is similar to SO_2 but slightly higher due to its higher molecular diffusivity. NH_3 and SO_2 are reasonably soluble gases in pure water and are effectively removed at higher rates under moist conditions (Erisman et al., 1993a,b; Erisman and Wyers, 1993). Even if the chemical characteristics of NH_3 are not the same than SO_2, these two gases have similar deposition characteristics (Zhang et al. 2002, 2003b). HNO_3 presents the highest V_d among all the chemical species in this study because of its high solubility and reactivity. Note that in the big-leaf model of Zhang et al. (2003b), SO_2 and O_3 are used as base species to scale the dry deposition rate for other chemical species. The monthly means of dry deposition velocities range from 0.16 to 0.84 $cm\,s^{-1}$ for SO_2, from 0.14 to 0.40 $cm\,s^{-1}$ for O_3, from 0.16 to 0.99 $cm\,s^{-1}$ for NH_3, from 0.13 to 0.37 $cm\,s^{-1}$ for NO_2 and from 0.48 to 2.56 $cm\,s^{-1}$ for HNO_3 on the transect dry savannas-wet savannas-forests. In addition, V_d are higher in the wet season for each ecosystem, especially in dry savannas where the dry season is well marked. The seasonal average V_d of NH_3 increases from 0.18 to 0.74 $cm\,s^{-1}$ in the dry season and from 0.31 to 0.87 $cm\,s^{-1}$ in the wet season, and the V_d of NO_2 increases from 0.13 to 0.31 $cm\,s^{-1}$ in the dry season and from 0.17 to 0.33 $cm\,s^{-1}$ in the wet season on the transect of ecosystems. It is considered that higher deposition velocities in the wet season are mainly caused by non-stomatal uptake of wet canopies (Matsuda et al., 2006; Tsai et al., 2010). The 7 yr mean annual V_d of nitrogen compounds, sulfur dioxide and ozone at IDAF sites are presented in Table 4. The interannual variability of V_d for the seven year period is low and range from 3 to 12 % for NH_3 and SO_2, from 2 to 11 % for NO_2 and O_3, and from 2 to 17 % for HNO_3 on the transect of ecosystems. These variations could be attributed to the spatio-temporal variations of meteorological data. The highest coefficients of variation were observed in the dry savannas with a maximum for HNO_3 (17 %) in relation to negligible canopy resistance and to wind speed variations.

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V_d values estimated in this work are lower or within a reasonable range compared to earlier studies (e.g., Hanson and Lindberg, 1991; Duyzer and Fowler, 1994; Zhang et al. 2002, 2009, Delon et al. 2010, 2012, Endo et al., 2011). For the dry savanna sites of IDAF, Delon et al. (2010) simulated monthly mean V_d from 0.1 to 0.33 cm s^{-1} for NO_2, from 0.11 to 0.39 cm s^{-1} for $NH_3$and from 0.34 to 0.61 cm s^{-1} for HNO_3, well comparable with NO_2 V_d (0.12–0.25 cm s^{-1}) and NH_3 V_d (0.16–0.51 cm s^{-1}) but slightly lower than HNO_3 V_d (0.48–1.39 cm s^{-1}) found in this study. Zhang et al. (2005) reported that the monthly averaged NO_2 values are around 0.35–0.45 cm s^{-1} in the summer months at seven eastern Canadian rural sites and HNO_3 V_d around six to nine times higher than NO_2. Flechard et al. (2011) simulated V_d of HNO_3 in the range of 1.0 to 1.2 cm s^{-1} over short vegetation in European Ecosystems, comparable to those calculated in this study for savanna sites (0.7–1.2 cm s^{-1}) although the types of vegetation and soil are completely different between the tropical sites and the European sites (Table 4). However, the modeled V_d of HNO_3over the forest estimated in this study ($\approx$ 2 cm s^{-1}) was in the lower range compared to 1.2–11.7 cm s^{-1} range in Japanese forest (Endo et al., 2011). This is due to lower wind speeds in the forcing (0.7–1.5 m s^{-1} in African forests, as a monthly average, compared to 0.6–6.7 m s^{-1} in Endo et al. 2011). Low wind speed involves high aerodynamic resistance (R_a) which is the dominant resistance for HNO_3 V_d (Wesely and Hicks, 2000). For NH_3, the 7 yr mean annual V_d on forests (0.8 cm s^{-1}) and savannas (0.2–0.5 cm s^{-1}) for the IDAF sites are well comparable with those estimated at EANET sites in Japan (0.5–0.9 cm s^{-1} for forest and 0.2–0.6 cm s^{-1} for grass) for 5 yr (Endo et al., 2011). Moreover, the mean modeled V_d of SO_2 at a Japanese cedar forest (Takahashi et al., 2001), 0.88 cm s^{-1}, is of the same order of magnitude that the one estimated at African forested ecosystems (0.7 cm s^{-1}). For $O_3$$V_d$, in order to compare our simulation with other tropical forests, we have calculated monthly V_d diurnal variations (from 3-hourly value) at the forested ecosystem of Zoetele for the year 2006. The ranges of mean O_3 V_d were estimated to be 0.54–0.61 cm s^{-1} (daytime) and 0.14–0.18 cm s^{-1} (nighttime) in the dry season, 0.62–0.70 cm s^{-1} (daytime)

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and 0.15–0.21 $cm\,s^{-1}$ (nighttime) in the wet season. These values are comparable to those observed above a tropical forest in northern Thailand using the aerodynamic gradient method: 0.37–0.39 $cm\,s^{-1}$ (daytime) and 0.12–0.13 $cm\,s^{-1}$ (nighttime) in the dry season; 0.62–0.65 $cm\,s^{-1}$ (daytime) and 0.25–0.27 $cm\,s^{-1}$ (nighttime) in the wet season (Matsuda et al., 2006). In general, the V_d diurnal variation is highly dependent on incoming solar radiation so that the daytime V_d increases with the solar radiation due to stomatal uptake (Tsai et al., 2010).

3.2 Dry deposition fluxes

Monthly dry deposition fluxes of NO_2, NH_3, HNO_3, SO_2 and O_3 were estimated using the inferential method at West and Central African sites of the IDAF network over the study period (1998–2007). By using the monthly mean V_d averaged over 6 yr (2002–2007), the monthly deposition fluxes have been calculated for each year and then averaged over the study period in order to estimate the mean range values of gaseous deposition fluxes representative of African ecosystems.

3.2.1 Nitrogen compounds (NO_2, HNO_3, NH_3)

Table 5 presents a synthesis of the mean seasonal and annual dry deposition fluxes of nitrogen compounds (NO_2, HNO_3, NH_3).

Nitrogen dioxide (NO_2)

Figure 4 presents the monthly evolution of NO_2 dry deposition flux (NO_2_dd) estimated at the dry savannas of Niger and Mali (4a: Banizoumbou, Agoufou, Katibougou), at the wet savannas of Cote d'Ivoire and Benin (4b: Lamto, Djougou) and at the evergreen equatorial forests of Cameroon and Congo (4c: Zoetele, Bomassa). Vertical bars indicate the standard deviation calculated over the study period (1998–2007). Average monthly evolution of NO_2 concentrations over the same study period is superimposed

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on the monthly dry deposition fluxes. We note a good correlation between gas concentrations and dry deposition fluxes on the transect dry savannas-wet savannas-forests.

In the dry savannas, monthly mean dry deposition fluxes of NO_2 range between 0.12 ± 0.08 and $1.27 \pm 0.29\,kg\,N\,ha^{-1}\,yr^{-1}$. Maximum flux values are observed in May/June as for concentration values (Fig. 4a). In fact, significant deposition fluxes start at the beginning of the wet season (usually around May in dry savanna areas) due to a pulse emission flux which is produced over very dry soils when the first rains fall (Johansson et al., 1988; Yienger and Levy, 1995; Laville et al., 2005; Delon et al., 2008, 2010). Adon et al. (2010) showed that the surface NO_2 concentration has its first maximum at the beginning of the rainy season. In the Sahelian savannas (Agoufou, Banizoumbou), seasonal averaged dry deposition fluxes are two times higher in the wet season ($0.75–0.93\,kg\,N\,ha^{-1}\,yr^{-1}$) than in the dry season ($0.29–0.41\,kg\,N\,ha^{-1}\,yr^{-1}$) (Table 5). In general, higher deposition fluxes in the wet season are due to both higher measured NO_2 concentrations and its higher dry deposition velocity calculated in this season at all the three dry savanna sites of West Africa. Annual average dry deposition fluxes of NO_2 over the period 1998–2007 are $0.6 \pm 0.1\,kg\,N\,ha^{-1}\,yr^{-1}$ at Banizoumbou and $0.7 \pm 0.1\,kg\,N\,ha^{-1}\,yr^{-1}$ at Katibougou, and over the period 2005–2009, $0.4 \pm 0.1\,kg\,N\,ha^{-1}\,yr^{-1}$ at Agoufou. These values calculated in these pastoral areas are in the lower range of deposition fluxes calculated for a tropical pasture site in Brazil (Rondonia) (annual NO_2 deposition flux: $0.76–2.4\,kg\,N\,ha^{-1}\,yr^{-1}$), which is considerably higher, mainly due to higher NO_2 mixing ratios observed (Trebs et al., 2006). Kirkman et al. (2002) estimated that the pasture site (Rondonia) constitutes a NO_2 sink of $0.73\,kg\,N\,ha^{-1}\,yr^{-1}$, well comparable with our estimated values ($0.4–0.7\,kg\,N\,ha^{-1}\,yr^{-1}$).

In the wet savannas, monthly mean NO_2 dry deposition fluxes range from 0.17 ± 0.01 to $1.11 \pm 0.11\,kg\,N\,ha^{-1}\,yr^{-1}$. Higher values are obtained in the dry season with maximum fluxes in December at Djougou and January at Lamto, such as NO_2 concentrations (Fig. 4b). In the wet savannas, where the vegetation density is more important than in the dry savannas, biomass burning is the most important source of NO_x ($NO + NO_2$) in the dry season. Galanter et al. (2000) have shown that more than 75 %

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of the NO_x at the surface near equatorial Africa is the result of biomass burning that occurs from December to February. African fires have a well known seasonality with a northern fire belt in November–February and a southern fire belt in June–October (Hao and Liu, 1994; Cooke et al., 1996). Seasonal mean dry deposition fluxes of NO_2 are 0.5–0.6 $kg N ha^{-1} yr^{-1}$ in the dry season and 0.25–0.37 $kg N ha^{-1} yr^{-1}$ in the wet season. In the dry season, flux values are 1.5 times higher at Lamto and 2.3 times higher at Djougou than in the wet season (Table 5), due to higher NO_2 concentrations. Adon et al. (2010) reported that seasonal NO_2 concentrations were 2 and 4 times higher in the dry season respectively at Lamto and Djougou. To the opposite, the $NO_2 V_d$ was slightly higher in the wet season (0.22–0.31 $cm s^{-1}$) than in the dry season (0.16–0.23 $cm s^{-1}$). These observations indicate that ambient concentrations largely control dry deposition of NO_2 in the wet savannas. Annual average dry deposition fluxes of NO_2 are of the same order of magnitude (0.4 $kg N ha^{-1} yr^{-1}$) for the two wet savanna sites (Table 5).

In forests, monthly mean dry deposition fluxes of NO_2 range between 0.31 ± 0.15 and 1.32 ± 0.69 $kg N ha^{-1} yr^{-1}$, with higher values observed in February and March, i.e. at the end of the dry season and at the beginning of the wet season (Fig. 4c). Seasonal average fluxes are of the same order of magnitude with values of 0.6–0.8 $kg N ha^{-1} yr^{-1}$ in the dry season and 0.5–0.76 $kg N ha^{-1} yr^{-1}$ in the wet season (Table 5). As in the wet savannas, the difference in deposition fluxes between the two seasons in forests is reduced by values of NO_2 V_d slightly higher in the wet season. NO_2 concentration measurements tend to show that the biomass burning source during the dry season is equivalent to soil emissions buffered by canopy uptake in forests during the wet season (Adon et al., 2010). Annual average dry deposition fluxes of NO_2 over the ten year period (1998–2007) are 0.5 ± 0.1 $kg N ha^{-1} yr^{-1}$ at Zoetele and 0.8 ± 0.2 $kg N ha^{-1} yr^{-1}$ at Bomassa. These values are well within the range of measurements derived by Hanson et al. (1989) who showed that N deposition from NO_2 was between 0.008 and 1.9 $kg N ha^{-1} yr^{-1}$ for natural forests. They are also comparable to NO_2 deposition fluxes

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estimated by Zhang et al. (2005) that range from 0.1 to 1.5 kg N ha^{-1} yr^{-1} at seven eastern Canadian rural sites for a 1 yr period.

Nitric acid (HNO_3)

Monthly mean dry deposition fluxes of HNO_3 (HNO_3_dd) range from 0.08 ± 0.02 to 2.67 ± 0.59 kg N ha^{-1} yr^{-1} in the dry savannas, from 0.26 ± 0.08 to 1.26 ± 0.13 kg N ha^{-1} yr^{-1} in the wet savannas and from 0.57 ± 0.31 to 2.29 ± 1.41 kg N ha^{-1} yr^{-1} in the forested ecosystems over the study period (1998–2007) (Fig. 5a, b, c). As for NO_2, monthly evolution of HNO_3 dry deposition fluxes is similar to that of HNO_3 concentrations on the transect of ecosystems. In the dry savannas, HNO_3 dry deposition fluxes is very low in the dry season, especially at Agoufou and Banizoumbou, and much (4–12 more times) higher in the wet season due to higher both HNO_3 concentrations and dry deposition velocities in this season for the three sites. Seasonal average fluxes range from 0.14 ± 0.09 kg N ha^{-1} yr^{-1} in the dry season to 1.74 ± 0.80 kg N ha^{-1} yr^{-1} in the wet season (Table 5). In the wet savannas, the difference in deposition fluxes between the two seasons is low with higher values in the dry season at Lamto (0.90 ± 0.29 kg N ha^{-1} yr^{-1}) and in the wet season at Djougou (0.79 ± 0.27 kg N ha^{-1} yr^{-1}) (Table 5). The deposition fluxes are less important in the dry season at Djougou, due to the smaller $HNO_3 V_d$ in this season. In forests, seasonal average deposition fluxes of HNO_3 range between 1.66 ± 0.63 and 1.10 ± 0.17 kg N ha^{-1} yr^{-1} in the dry season, and between 0.75 ± 0.23 and 0.91 ± 0.13 kg N ha^{-1} yr^{-1} in the wet season, respectively at Zoetele and Bomassa. The difference in deposition flux values between the two seasons is low for Bomassa, but important for Zoetele where fluxes are two times higher in the dry season due to higher values of HNO_3 concentrations. Annual mean dry deposition fluxes of HNO_3 are 0.7 ± 0.3 kg N ha^{-1} yr^{-1} in the dry and wet savannas and 1.0 ± 0.3 kg N ha^{-1} yr^{-1} in forests (Table 5). Annual mean HNO_3 deposition fluxes show a low variability according to the ecosystems with seasonal differences especially pronounced in the dry savannas. Although HNO_3 concentrations (0.3–0.5 ppb) were lower

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than NO_2 (0.9–2.4 ppb) on the African ecosystems transect (Table 2), HNO_3 dry deposition fluxes were typically as important as NO_2 dry deposition fluxes owing to higher V_d values for HNO_3 (Table 4).

Few observational studies of HNO_3 dry deposition are available over tropical areas. Huebert and Robert (1985) measured the dry deposition flux of nitric acid vapor to a pasture near Champaign, Illinois during June 1982. The measured June dry deposition flux of HNO_3 was estimated to be 1.1–1.4 $kg\,ha^{-1}\,month^{-1}$ (or 2.93–3.75 $kg\,N\,ha^{-1}\,yr^{-1}$). This value is comparable to monthly mean deposition fluxes estimated at the pastoral sites of the dry savannas in the wet season where fluxes can reach the value of 2.67 $kg\,N\,ha^{-1}\,yr^{-1}$.

Ammoniac (NH_3)

On the transect of ecosystems, over the ten-year period (1998–2007), monthly mean NH_3 dry deposition fluxes range from 1.01 ± 0.48 to 7.06 ± 2.16 $kg\,N\,ha^{-1}\,yr^{-1}$ in the dry savannas, from 1.24 ± 0.60 to 5.43 ± 2.26 $kg\,N\,ha^{-1}\,yr^{-1}$ in the wet savannas, and from 6.58 ± 2.29 to 13.57 ± 4.07 $kg\,N\,ha^{-1}\,yr^{-1}$ in the forests (Fig. 6a, b, c).

In the dry savannas, seasonal mean deposition fluxes of NH_3 are about 2 times higher in the wet season (4.21–5.74 $kg\,N\,ha^{-1}\,yr^{-1}$) than in the dry season (1.66–2.54 $kg\,N\,ha^{-1}\,yr^{-1}$) (Table 5). In the two seasons, the deposition flux is more important at Katibougou (Sudano-sahelian site) due to higher $NH_3 V_d$, compared to other Sahelian sites. Dry savannas are generally pastoral areas; the main sources include bacterial decomposition of urea in animal excreta, very active in the wet season with the hydrolysis of urea, and emissions from natural soils (Schlesinger and Hartley, 1992; Bouwman et al., 1997; Bouwman et al., 2002a). In these areas, high densities of domestic animals are concentrated on the fresh natural pastures which grow during the rainy season. Delon et al. (2010) estimated NH_3 emission fluxes from domestic animal excreta for the three Sahelian sites of IDAF network. Annual mean NH_3 emission fluxes by volatilization are estimated around 3.9 ± 2.2 $kg\,N\,ha^{-1}\,yr^{-1}$ in the dry savannas.

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Bouwman et al. (1997) estimated NH_3 emission fluxes from animal source between 0.5 and 5 $kg N ha^{-1} yr^{-1}$ in the Sahelian area for 1990. Almost all emitted NH_3 is returned to the surface by deposition (dry and wet) (Bouwman et al., 2002a). These values of NH_3 emitted are comparable to dry deposition fluxes estimated in the dry savannas, knowing that a part of NH_3 emitted will be deposited by wet and particulate deposition as well.

In the wet savannas, monthly deposition fluxes appear to be of the same order of magnitude between the two seasons whereas concentrations are higher at the beginning of the dry season (January–March) (Fig. 6b). Generally, higher concentrations of NH_3 in the dry season are due to savanna fires which are a significant source of ammonia in tropical regions (Lobert et al., 1990; Delmas et al., 1995). Another source of NH_3 is domestic fuelwood burning (Brocard et al., 1996). Seasonal average deposition fluxes of NH_3 range from 3.91 ± 1.29 to 3.07 ± 0.52 $kg N ha^{-1} yr^{-1}$ at Lamto, and from 2.11 ± 0.57 to 2.45 ± 0.67 $kg N ha^{-1} yr^{-1}$ at Djougou, in the dry and wet seasons, respectively. V_d values of NH_3, 1.8 times higher at Djougou and 1.3 times higher at Lamto in the wet season, explain the lower difference in deposition fluxes between the two seasons. In the forests, no significant difference between wet and dry seasons exists. Seasonal NH_3 dry deposition fluxes are 8.86 ± 1.42 and 9.91 ± 2.35 $kg N ha^{-1} yr^{-1}$ at Zoetele and 10.04 ± 2.99 and 10.09 ± 1.71 $kg N ha^{-1} yr^{-1}$ at Bomassa, in the dry and wet seasons, respectively. Concentrations and V_d of NH_3 are also of the same order of magnitude between the two seasons in forests.

On the transect of ecosystems, annual average dry deposition fluxes of NH_3 (NH_3_dd) range between 2.7 ± 0.9 and 3.9 ± 0.6 $kg N ha^{-1} yr^{-1}$ in the dry savannas, between 2.3 ± 0.8 and 3.4 ± 1.0 $kg N ha^{-1} yr^{-1}$ in the wet savannas and between 9.7 ± 2.2 and 10.01 ± 5.0 $kg N ha^{-1} yr^{-1}$ in the forests, over the study period (1998–2007) (Table 5). The unadjusted estimates of NH_3 dry deposition fluxes in forests (without the correction factor applied for NH_3 concentrations, Sect. 2.2.3) would range between

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6.4 ± 1.5 and 6.7 ± 2.3 kg N ha^{-1} yr^{-1}. On the African ecosystems, NH_3 deposition fluxes are about three times more important in forests than in dry and wet savannas. This result can be explained by higher V_d values for NH_3 estimated in forests (0.8 cm s^{-1}), compared to savannas (0.2–0.5 cm s^{-1}) (Table 4). In fact, on the transect of ecosystems, annual mean NH_3 concentrations measured were around 6–7.5 ppb in dry savannas and 4–5 ppb in wet savannas and forests (Table 2).

These estimates of nitrogen dry deposition from NO_2, HNO_3 and NH_3 on the transect of African ecosystems has allowed us to make a partial assessment of the total quantity of nitrogen deposited in gaseous form.

Total nitrogen dry deposition fluxes from NO_2, HNO_3 and NH_3

Monthly total N dry deposition ($NO_2 + HNO_3 + NH_3$) range from 1.3 to 9.7 kg N ha^{-1} yr^{-1} in the dry savannas, from 2.5 to 7.1 kg N ha^{-1} yr^{-1} in the wet savannas and from 7.6 to 16.0 kg N ha^{-1} yr^{-1} in forests over the period 1998–2007. In the dry savannas, seasonal total N dry deposition is more important in the wet season (6.5–7.9 kg N ha^{-1} yr^{-1}) than dry season (2.2–3.5 kg N ha^{-1} yr^{-1}). This is related to the important sources of biogenic emission for NO (microbial processes in soils) and NH_3 (hydrolysis of urea from animal urine deposited in grazing areas) in the wet season. Conversely, in the wet savannas, seasonal total N deposition is slightly higher in the dry season (5.4 kg N ha^{-1} yr^{-1}) than the wet season (4.0 kg N ha^{-1} yr^{-1}) at Lamto, and is of the same order of magnitude between the two seasons (3.2–3.5 kg N ha^{-1} yr^{-1}) at Djougou. We note that seasonal total N dry deposition in wet season is higher in the dry savannas than in the wet savannas. For the evergreen forested ecosystems, seasonal total N dry flux is of the same order of magnitude between the dry season (11.1–12.0 kg N ha^{-1} yr^{-1}) and the wet season (11.2–11.8 kg N ha^{-1} yr^{-1}) at Zoetele and Bomassa, respectively.

On the transect, over the 10 yr period (1998–2007), annual total N dry deposition is more important in forests (11.2–11.8 kg N ha^{-1} yr^{-1}) due to higher NH_3 deposition fluxes, and is of the same order of magnitude in wet savannas (3.4–4.6 kg N ha^{-1} yr^{-1})

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and dry savannas (4.0–5.3 kg N ha^{-1} yr^{-1}) (Table 5; Fig. 7a). NH_3 is the dominant species contributing to total N dry deposition over African ecosystems. On an annual basis (Fig. 7b), NH_3 contributes 67–86 %, HNO_3, 8–20 %, and NO_2, 5–16 %, to the total N dry deposition, depending on sites. Over the study period (1998–2007), the interannual variability of the dry deposition fluxes range from 10 to 35 % for NH_3, from 12 to 32 % for NO_2 and from 10 to 46 % for HNO_3 on the transect dry savannas-wet savannas-forests. No specific trend in the variability is observed, as already concluded in the study of Delon et al. (2012). The interannual variability is largely attributable to the variability of gaseous concentrations due to the potential variation of the intensity of atmospheric sources (Adon et al., 2010).

3.2.2 Sulfur dioxide (SO_2)

The monthly evolution of SO_2 dry deposition flux (SO_2_dd) on the transect of ecosystems is generally comparable to SO_2 concentrations over the period 2002–2007, following the same gradient (Fig. 8a–c). We note that the standard deviation of monthly mean fluxes is important. In fact, monthly SO_2 concentrations measured at IDAF sites are low and vary considerably from one year to another for the same month. Monthly mean SO_2 deposition fluxes range between 0.21 ± 0.16 and 1.17 ± 0.71 kg S ha^{-1} yr^{-1} in the dry savannas, between 0.38 ± 0.24 and 1.39 ± 0.42 kg S ha^{-1} yr^{-1} in the wet savannas and between 0.42 ± 0.24 and 1.57 ± 1.55 kg S ha^{-1} yr^{-1} in the forests.

If we consider the seasons, average SO_2 dry deposition fluxes on the transect are 0.85–1.0 kg S ha^{-1} yr^{-1}, 0.77–1.1 kg S ha^{-1} yr^{-1} and 0.7–1.0 kg S ha^{-1} yr^{-1} in the wet season and 0.25–0.47 kg S ha^{-1} yr^{-1}, 0.77–0.82 kg S ha^{-1} yr^{-1} and 0.8–0.9 kg S ha^{-1} yr^{-1} in the dry season, for dry savannas, wet savannas and forests, respectively. As in the case of nitrogen compounds, SO_2 dry deposition is important during the wet season in the dry savannas and is of the same order of magnitude between the two seasons for the wet savannas and forests. For the latter, dry deposition velocities, slightly higher in the wet season, reduce the difference between the two

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seasons. Adon et al. (2010) observed the same order of magnitude of SO_2 concentrations in dry and wet seasons in each ecosystem with a higher value in wet season for dry savannas. This suggests a contribution of soil emission, biosphere and biomass burning sources (Van Breemen, 1982, 1993; Macdonald et al., 2004; Bates et al., 1992; Arndt et al., 1997). However, the sulphur content of vegetation is lower compared to carbon and nitrogen elements and SO_2 emissions factors for combustion processes are lower than those for carbonaceous or nitrogen species (Lacaux et al., 1995). The 6 yr mean annual dry deposition fluxes of SO_2 are $0.5 \pm 0.2\,kg\,S\,ha^{-1}\,yr^{-1}$ at Banizoumbou, $0.7 \pm 0.3\,kg\,S\,ha^{-1}\,yr^{-1}$ at Katibougou and $0.7 \pm 0.3\,kg\,S\,ha^{-1}\,yr^{-1}$ at Agoufou in dry savannas, $0.8 \pm 0.4\,kg\,S\,ha^{-1}\,yr^{-1}$ at Lamto and $1.0 \pm 0.3\,kg\,S\,ha^{-1}\,yr^{-1}$ at Djougou in wet savannas, and $0.8 \pm 0.4\,kg\,S\,ha^{-1}\,yr^{-1}$ at Zoetele and $1.0 \pm 0.5\,kg\,S\,ha^{-1}\,yr^{-1}$ at Bomassa in forests. Along the ecosystem transect, annual mean of SO_2 dry deposition fluxes present low values and a small variability (0.5 to $1\,kg\,S\,ha^{-1}\,yr^{-1}$). The interannual variability over the 6 yr period is between 30 and 48 % for all the sites and no specific trend is observed.

Takahashi et al. (2002) simulated the dry deposition of SO_2 onto a Japanese cedar forest located in Gumma Prefecture, based on the results of 1 yr concentration measurements. The inferential estimate of SO_2 dry deposition flux was $3.6\,kg\,S\,ha^{-1}\,yr^{-1}$, which is three times higher than fluxes estimated at African tropical forests (around $1\,kg\,S\,ha^{-1}\,yr^{-1}$) although the V_d modeled are of the same order of magnitude ($0.88\,cm\,s^{-1}$ and $0.7\,cm\,s^{-1}$ in Japanese and African forests, respectively). In northern China (near Beijing), Sorimachi et al. (2003) measured sulfur dry deposition over short vegetation using the aerodynamic gradient method. The mean estimates of SO_2 dry deposition fluxes were $17.5 \pm 12\,kg\,S\,ha^{-1}\,yr^{-1}$ in late summer and $50.3 \pm 45\,kg\,S\,ha^{-1}\,yr^{-1}$ in early winter. These high values of SO_2 deposition fluxes are due to high concentrations measured (17.2 ppb in summer and 37 ppb in winter) related to the influence of anthropogenic activities. In general, dry deposition fluxes of SO_2 estimated at African ecosystems are low due to low concentrations measured (order of 1–2 ppb). Indeed,

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the remote measurement sites of IDAF network are not yet impacted by anthropogenic activities or industrial emissions and as a consequence SO_2 sources are low.

3.2.3 Ozone (O_3)

The monthly evolution of O_3 dry deposition fluxes (O_3_dd) is similar to that of O_3 concentrations on the transect of ecosystems over the period 2001–2007, as in the case of NO_2 (Fig. 9a–c). Monthly average O_3 deposition fluxes range from 2.9 ± 0.4 to $30.3 \pm 2.3\,kg\,ha^{-1}\,yr^{-1}$ in dry savannas, from 11.6 ± 4.3 to $22.2 \pm 2.2\,kg\,ha^{-1}\,yr^{-1}$ in wet savannas and from 8.2 ± 2.2 to $19.4 \pm 8.2\,kg\,ha^{-1}\,yr^{-1}$in forests. In the semi arid savannas, the seasonal cycle is clear with maximum O_3 deposition fluxes in the heart of the wet season due to higher values of both O_3 concentrations and V_d. In fact, high ozone concentrations during the wet season are the result of active photochemical production of O_3 in the boundary layer associated with high NO_x concentrations (Stewart et al., 2008). In the Sahelian region, the ozone production in the wet season is mainly related to natural biogenic precursor sources (Adon et al., 2010). The seasonal average deposition fluxes are 6.4 ± 4.1 and $21.1 \pm 2.7\,kg\,ha^{-1}\,yr^{-1}$at Agoufou, 7.2 ± 2.0 and $20.5 \pm 3.6\,kg\,ha^{-1}\,yr^{-1}$ at Banizoumbou and 13.2 ± 3.6 and $23.6 \pm 2.5\,kg\,ha^{-1}\,yr^{-1}$at Katibougou, in dry and wet seasons, respectively. These seasonal values are three times higher in the wet season than in the dry season for the Sahelian sites of Agoufou and Banizoumbou. In the wet savannas, the seasonal average O_3 deposition fluxes are of the same order of magnitude with values of 20.1 ± 1.5 and $18.5 \pm 3.3\,kg\,ha^{-1}\,yr^{-1}$at Lamto, and 16.4 ± 3.5 and $18.3 \pm 4.0\,kg\,ha^{-1}\,yr^{-1}$ at Djougou, in dry and wet seasons, respectively. The difference between the seasonal ozone depositions is reduced with higher values of V_d in the wet season and higher O_3 concentrations in the dry season as a consequence of strong regional biomass burning activities. For forested ecosystems, the mean seasonal deposition fluxes of O_3 are 19.2 ± 0.3 and $11.3 \pm 2.7\,kg\,ha^{-1}\,yr^{-1}$ at Zoetele and 10.1 ± 1.4 and $10.8 \pm 1.5\,kg\,ha^{-1}\,yr^{-1}$ at Bomassa, in dry and wet seasons, respectively. We note that these seasonal values are of the same order of magnitude

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between the two seasons at Bomassa but they are higher in the dry season at Zoetele due to higher O_3 concentrations measured in this season.

The 7 yr mean annual dry deposition fluxes of O_3 are around 11–19 kg ha^{-1} yr^{-1} in dry and wet savannas (11.3 ± 4.7 kg ha^{-1} yr^{-1} at Agoufou, 12.8 ± 2.6 kg ha^{-1} yr^{-1} at Banizoumbou and 17.5 ± 3.0 kg ha^{-1} yr^{-1} at Katibougou, 17.5 ± 3.0 kg ha^{-1} yr^{-1} at Djougou and 19.2 ± 2.9 kg ha^{-1} yr^{-1} at Lamto) and 11–13 kg ha^{-1} yr^{-1} in forests (13.2 ± 3.6 kg ha^{-1} yr^{-1} at Zoetele and 10.6 ± 2.0 kg ha^{-1} yr^{-1} at Bomassa). The unadjusted estimates of O_3 dry deposition fluxes in forests (without the correction factor applied for O_3 concentrations, Sect. 2.2.3) would range between 8–10 kg ha^{-1} yr^{-1} (8.2 ± 0.6 and 10.2 ± 2.1 kg ha^{-1} yr^{-1} at Bomassa and Zoetele, respectively). The interannaul variability of O_3 deposition fluxes is between 7 and 21 % over the period 2001–2007 for all sites. This variability is mainly related to the spatio-temporal evolution of ozone concentrations which depends on various processes (precursor emissions, biogenic and anthropogenic sources) (Adon et al., 2010). Along the ecosystem transect, ozone deposition is more important in the wet savannas (Djougou and Lamto) and in the Soudano-Sahelian site of Katibougou, with a value of 18–19 kg ha^{-1} yr^{-1}. In savannas, the values of annual O_3 deposition fluxes are lower at Agoufou and Banizoumbou due to the low vegetation cover of the Sahelian zone (thus, low V_d). The low values of O_3 dry deposition in forests are correlated to low values of O_3 concentrations which were 2 to 3 times lower than those measured in the other ecosystems. The 10 yr annual concentrations range between 11–14 ppb in savannas (dry and wet) and 4–5 ppb in forests (Table 2). Several studies have shown that tropical forests appear to be a major O_3 sink, through ground and foliage deposits and loss through chemical reactions with hydrocarbons and nitrogen oxides (Rummel at al., 2007; Jacob and Wofsy, 1990; Bakwin et al., 1990; Kaplan et al., 1988).

Few observational studies of O_3 deposition fluxes for a long term period are available over tropical areas. Although measurements of ozone deposition fluxes over African tropical savannas are rare, several studies have been conducted on the tropical rain forest (Cros et al., 1992, 2000; Andreae et al., 1992; Rummel et al., 2007; Matsuda et al.,

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2005, . . .) but these studies only concern the diurnal evolution of O_3 deposition. Hourly and diurnal values of these fluxes are not comparable to monthly and annual values of O_3 deposition fluxes estimated in this study. Mikkelsen et al. (2004) have measured ozone concentrations and fluxes continuously during 5 yr (1996–2000) by the gradient method in a Norway spruce dominated forest stand in West Jutland, Denmark. The overall average concentration was 30.0 ppb during nighttime and 35.7 ppb during the day. The annual ozone deposition estimated was 125 kg ha^{-1} yr^{-1}, which is around 10 times higher than the annual mean from African forests. Zeller and Nikolov (2000) estimated fluxes above a subalpine coniferous forest in southern Wyoming (USA) by eddy covariance and model analyses and found a total ozone deposition of 29 kg ha^{-1} yr^{-1} for 1996, which is approximately 3 times higher than our estimates. In general, the low values of O_3 deposition on African tropical forest are due to lower concentrations measured, compared to other tropical sites (Adon et al., 2010).

4 Conclusions

In this study, we estimated dry deposition fluxes of nitrogen compounds (NO_2, HNO_3, NH_3), sulfur dioxide (SO_2) and ozone (O_3) in major African ecosystems, represented by IDAF sites. Monthly dry deposition fluxes have been estimated by the inferential technique, using the product of air concentrations measured monthly by passive samplers for a long term period (1998–2007) and dry deposition velocities modeled by the big-leaf model of Zhang et al. (2003b). In the deposition model, surface and meteorological conditions specific to IDAF sites have been adapted in order to simulate V_d representative of major African ecosystems. Along the transect of ecosystems, simulation results show that V_d increases with the vegetation density. Thus, the lower values of V_d for all gases have been obtained in the dry savannas and the higher in the forests. For each site, V_d values are higher in the wet season than in the dry season for all gases. For each ecosystem, the monthly, seasonal and annual mean variations of gaseous dry deposition fluxes have been analyzed. Dry deposition fluxes are more important

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in the wet season for all the gases in the dry savannas, due to higher values of both concentrations and V_d in this season. For the wet savannas and forested ecosystems, seasonal dry deposition fluxes are generally of the same order of magnitude between the dry and wet seasons for all the gases except for NO_2 in the wet savannas, and for HNO_3 and O_3 at the Zoetele forested site, where flux values are higher in the dry season due to much higher concentrations in this season.

Along the latitudinal transect of ecosystems, the annual mean dry deposition fluxes of nitrogen compounds range from 0.4 to 0.8 kg N ha^{-1} yr^{-1} for NO_2, from 0.7 to 1.0 kg N ha^{-1} yr^{-1} for HNO_3, and from 2.3 to 10.0 kg N ha^{-1} yr^{-1} for NH_3 over the study period (1998–2007). The total nitrogen dry deposition flux (NO_2+HNO_3+NH_3) is more important in forests (11.2–11.8 kg N ha^{-1} yr^{-1}) than in wet and dry savannas (3.4–5.3 kg N ha^{-1} yr^{-1}). NH_3 dominates nitrogen dry deposition, representing 67–86 % of the total. Along the ecosystem transect, annual mean of SO_2 dry deposition fluxes present low values and a small variability (0.5 to 1 kg S ha^{-1} yr^{-1}). For ozone, the annual mean dry deposition fluxes are around 11–19 kg ha^{-1} yr^{-1} in dry and wet savannas and 11–13 kg ha^{-1} yr^{-1} in forests. The lower O_3 dry deposition fluxes in forests are due to low measured O_3 concentrations despite higher V_d. Over the study period, the inter-annual variability of gaseous dry deposition fluxes showed no specific trend.

This study allowed estimates of the mean range of gaseous dry deposition fluxes representative of major tropical African ecosystems in West and Central Africa. This is one of the major scientific objectives of the IDAF program. It is based on original and unique data from remote and seldom explored regions. To improve this work, it is important to address the uncertainties in the determination of dry deposition velocities. Within the IDAF network, we suggest that an experimental determination of dry deposition fluxes by other methods (e.g. gradient method, eddy correlation) on the measurement sites would be required to compare with fluxes estimated by inferential method. Furthermore, we plan to use the model RegCM4 (Regional Climatic Model) (Giorgi et al., 2012; Shalaby et al., 2012) to simulate the regional trends of gaseous dry deposition fluxes and to compare these results to long term IDAF observations. This

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work will allow providing a high resolution map of dry deposition at regional scales of the African ecosystems useful for impact studies.

Appendix A

The dry deposition velocity (V_d) is obtained from the sum of three resistances in series as follows:

$$V_d = (R_a + R_b + R_c)^{-1} \quad \text{(A1)}$$

where R_a is the aerodynamic resistance to transfer of a species, as a result of atmospheric turbulence in the surface layer between a height Z and the surface, characterized by Z_0 (roughness length). It is computed from the following equation (Padro et al., 1991):

$$R_a = \frac{1}{k u_*} \left[0.74 \ln(Z/Z_o) - \Psi_H(Z/L)\right] \quad \text{(A2)}$$

where Ψ_H is the integrated stability function for heat, k is the von Karman constant (0.4). L is a stability parameter (Monin-Obukhov length) and can be computed jointly with u_* (friction velocity) from the basic surface layer equations, requiring only the temperatures at two different heights and the wind at one height. The conditions of application of the Eq. (A2) are discussed in Padro et al. (1991). Under very stable condition, the Richardson number is constrained to an upper limit value of 0.21. For our simulation, after a statistical analysis, the friction velocity is constrained to a lower limit value of 0.1 $\mathrm{m\,s^{-1}}$ for savannas sites and of 0.2 $\mathrm{m\,s^{-1}}$ for forest sites.

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R_b is the quasi-laminar sublayer resistance above the canopy and it is computed from the following equation (Padro and Edwards, 1991):

$$R_b = \frac{2}{k u_*} \left(v/D_j\right)^{2/3} \quad \text{(A3)}$$

where v is the kinematic viscosity of air and D_j is the molecular diffusivity of a species j in air.

R_c is the surface or canopy resistance, and characterises the surface affinity for pollutant uptake. A large part of the uncertainty of inferential method might be attributed to the parameterization of R_c. Zhang et al. (2003b) proposed a revised parameterization of R_c by including non-stomatal resistance (R_{ns}) parameterizations based on study results over 5 different vegetation types in North America (i.e., Zhang et al., 2002b, 2003a):

$$\frac{1}{R_c} = \frac{1 - w_{st}}{R_{st} + R_m} + \frac{1}{R_{ns}} \quad \text{(A4)}$$

$$\frac{1}{R_{ns}} = \frac{1}{R_{cut}} + \frac{1}{R_{ac} + R_g} \quad \text{(A5)}$$

where the sub-resistances R_{st}, R_m, R_{cut}, R_{ac} and R_g are respectively stomatal, mesophyl, cuticle, in-canopy aerodynamic and soil resistances. w_{st} is the fraction of stomatal blocking under wet conditions. One of the improvements to the model of Zhang et al. (2003b) include more realistic treatment of cuticle resistance which is parameterized as functions of leaf wetness (dry vs. wet; dew vs. rain), relative humidity, leaf area index (LAI), friction velocity and land-use specific reference value. R_{ac} is also a function of the LAI, the friction velocity and the land-use specific reference value. Note that R_g and R_{cut} are calculated for SO_2 and O_3 and then scaled for other gases. R_{st} is calculated using a sunlit/shade stomatal resistance sub-model (Zhang et al., 2002a). Thus, in this improved parameterization, R_c depends on the type of canopy, the chemical species and the meteorological conditions. The parameterizations of all

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these sub-resistances, the land use categories (LUC) and all related parameters, and more details can be found in (Zhang et al., 2003b).

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Table 1. Geographic, ecologic and climatic characteristics of the Western and Central Africa IDAF sites. WS: Wet season, DS: Dry season.

Ecosystem	station	Location Latitude Longitude	Land cover classes (GLC 2000 Africa)	Climate	Country (region)
Dry savannas	Agoufou	15° 20′ N 01° 29′ W	Open grassland with sparse shrub	Sahelian WS: June–September DS: October–May	Mali (Mopti)
	Banizoumbou	13° 31′ N 02° 38′ E	Open grassland with sparse shrub	Sahelian WS: June-September DS: October-May	Niger (Dosso)
	Katibougou	12° 56′ N 07° 32′ W	Deciduous shrubland with sparse trees	Soudano-Sahelian WS: June-September DS: October-May	Mali (Koulikoro)
Wet savannas	Djougou	09° 39′ N 01° 44′ E	Deciduous open woodland	Soudano-Guinean WS: April-October DS: November-March	Benin (Atakora)
	Lamto	06° 13′ N 05° 02′ W	Mosaic Forest/Savanna	Guinean WS: April-October DS: November-March	Cote d'Ivoire (V-Baoule)
Forests	Zoetele	03° 15′ N 11° 53′ E	Closed evergreen lowland forest	Equatorial WS: March-November DS: December-February	Cameroon (Sud, Dja-et-Lobo)
	Bomassa	02° 12′ N 16° 20′ E	Closed evergreen lowland forest	Equatorial WS: March-November DS: December-February	Congo (Sangha)

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Table 2. Mean annual concentrations (in ppb) at IDAF sites over the study period (1998 2007) (adapted from Adon et al., 2010).

Concentration (ppb)	Station (période)	NO_2	HNO_3	NH_3	O_3 [a]	SO_2 [b]
Dry savannas	Agoufou (05-09)	1.8 ± 0.4	0.5 ± 0.1	7.3 ± 0.8	11.4 ± 2.2	0.8 ± 0.3
	Banizoumbou 98-07)	2.4 ± 0.4	0.5 ± 0.2	6.3 ± 2.0	11.9 ± 2.3	0.6 ± 0.2
	Katibougou (98-07)	1.9 ± 0.3	0.4 ± 0.1	6.6 ± 1.0	12.6 ± 2.2	0.6 ± 0.2
Wet Savannas Forests	Djougou (05-09)	1.2 ± 0.1	0.4 ± 0.2	3.7 ± 1.1	13.6 ± 2.1	0.8 ± 0.3
	Lamto (98-07)	1.0 ± 0.3	0.3 ± 0.1	4.0 ± 1.2	10.9 ± 1.8	0.5 ± 0.2
	Zoetele (98-07)	0.9 ± 0.2	0.2 ± 0.1	4.2 ± 0.9	4.8 ± 1.0	0.3 ± 0.1
	Bomassa (98-06)	1.4 ± 0.4	0.3 ± 0.1	4.7 ± 1.7	4.0 ± 0.4	0.4 ± 0.2

[a] O_3 starts monitoring in 2001 and [b] SO_2 in 2002.

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Table 3. Adaptation of Land use categories (LUC) used in the big-leaf model to IDAF sites for the calculation of canopy resistances (R_{ac}, R_{cut}, R_{st}) in V_d.

Sites (type of ecosystem)	Land Use Cover (LUC) (resistances)	Reference
Agoufou, Banizoumbou (grassland)	Short grass and forbs (R_{ac}, R_{cut}, R_{st})	Zhang et al. (2003b)
Katibougou (Shrubland)	Long grass (R_{ac}, R_{cut}) and Broadleaf shrubs with perennial ground cover (R_{st})	Zhang et al. (2003b) Brook et al. (2009)
Djougou, Lamto (tree Savanna)	Long grass (R_{ac}, R_{cut}) and Broadleaf trees with ground cover or savanna(R_{st})	Zhang et al. (2003b) Brook et al. (2009)
Zoetele, Bomassa (Forest)	Tropical broadleaf trees (R_{ac}, R_{cut}, R_{st})	Zhang et al. (2003b)

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Table 4. Mean annual dry deposition velocities (V_d in $cm\,s^{-1}$) of gases on the transect of African ecosystems over the period 2002–2007.

V_d (cm s)	SO_2	NH_3	O_3	NO_2	HNO_3
Dry savannas	0.21–0.29	0.22–0.32	0.16–0.22	0.15–0.20	0.68–0.99
Wet savannas	0.31–0.42	0.35–0.51	0.21–0.29	0.20–0.28	0.97–1.17
Forests	0.64–0.69	0.77–0.84	0.33–0.35	0.31–0.32	2.0–2.2

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Table 5. Mean seasonal and annual dry deposition fluxes and standard deviation ($kg N ha^{-1} yr^{-1}$) of NO_2, HNO_3, NH_3 and total N at IDAF sites of West and Central Africa over the period 1998–2007.

Dry deposition fluxes ($KgNha^{-1}yr^{-1}$)		NO_2			HNO_3			NH_3			Total N
seasons		Dry	wet	annual	dry	wet	annual	dry	wet	annual	annual
Dry savannas	Agoufou*	0.29 ± 0.12	0.75 ± 0.24	0.4 ± 0.1	0.14 ± 0.09	1.74 ± 0.80	0.7 ± 0.1	2.06 ± 0.63	4.52 ± 0.24	2.9 ± 0.3	4.0
	Banizoumbou	0.41 ± 0.23	0.93 ± 0.25	0.6 ± 0.1	0.17 ± 0.09	1.40 ± 0.43	0.7 ± 0.3	1.66 ± 0.69	4.21 ± 0.72	2.7 ± 0.9	4.0
	Katibougou	0.59 ± 0.16	0.78 ± 0.24	0.7 ± 0.1	0.36 ± 0.13	1.35 ± 0.52	0.8 ± 0.2	2.54 ± 1.01	5.74 ± 0.96	3.9 ± 0.6	5.3
Wet savannas	Djougou*	0.57 ± 0.32	0.25 ± 0.10	0.4 ± 0.0	0.54 ± 0.19	0.79 ± 0.27	0.7 ± 0.2	2.11 ± 0.57	2.45 ± 0.67	2.3 ± 0.8	3.4
	Lamto	0.54 ± 0.16	0.37 ± 0.15	0.4 ± 0.1	0.90 ± 0.29	0.59 ± 0.17	0.7 ± 0.1	3.91 ± 1.29	3.07 ± 0.52	3.4 ± 1.0	4.6
Forests	Zoetele	0.60 ± 0.19	0.52 ± 0.19	0.5 ± 0.1	1.66 ± 0.63	0.75 ± 0.23	1.0 ± 0.3	8.86 ± 1.42	9.91 ± 2.35	9.7 ± 2.2	11.2
	Bomassa	0.83 ± 0.37	0.76 ± 0.28	0.8 ± 0.2	1.10 ± 0.17	0.91 ± 0.13	1.0 ± 0.3	10.04 ± 2.99	10.09 ± 1.71	10.01 ± 5.0	11.8

* Deposition flux values of Agoufou and Djougou are averaged over the period 2005–2009.

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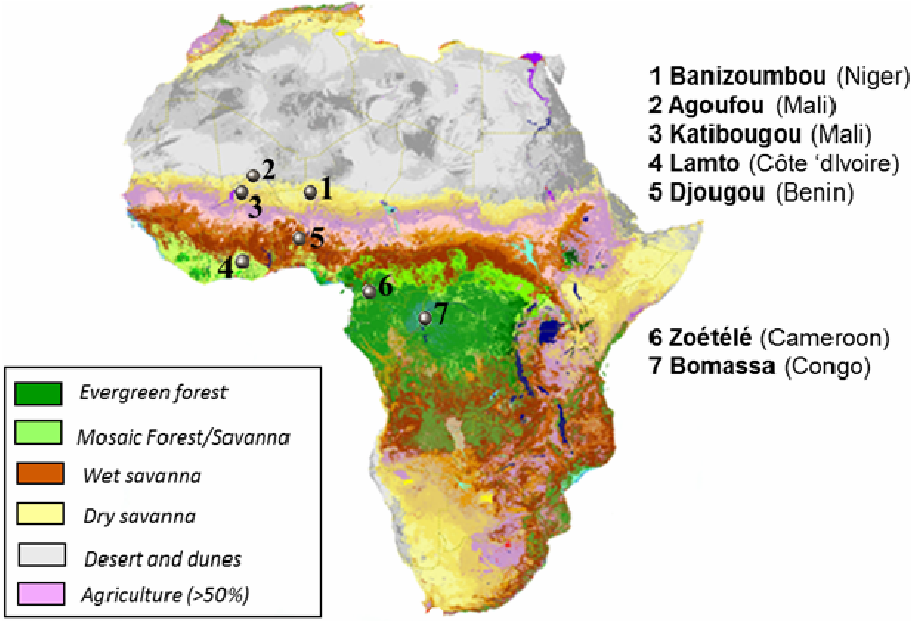

Fig. 1. Vegetation and location map of the 7 measurement stations of the IDAF network. Only the 7 IDAF stations of West and Central Africa included in the present study are represented.

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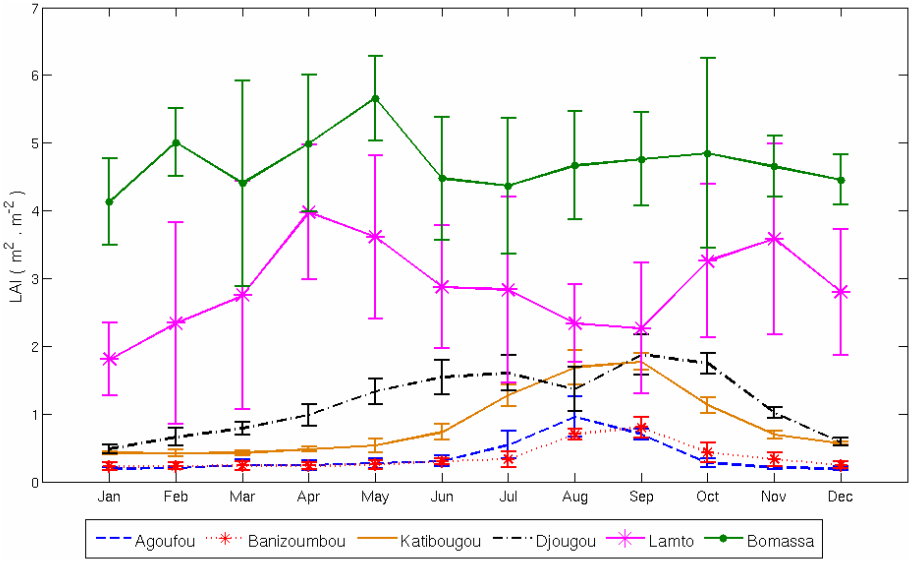

Fig. 2. Monthly variation of MODIS LAI averaged over the period 2000–2007 for IDAF sites. Vertical bars depict the standard deviation over the study period.

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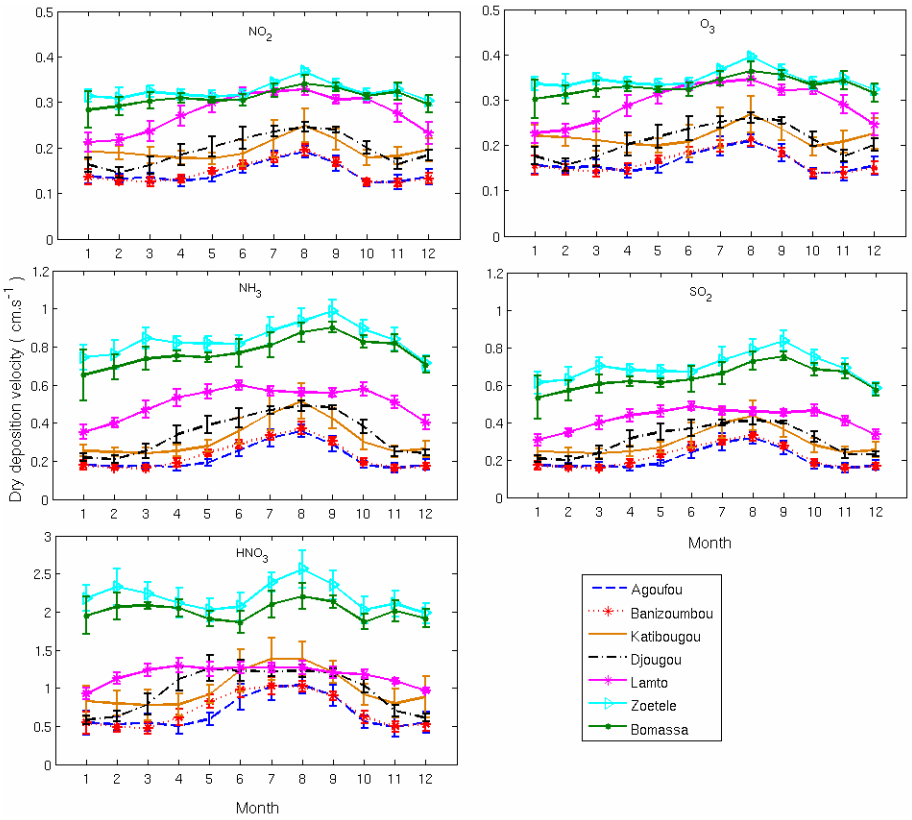

Fig. 3. Monthly variation of dry deposition velocity (V_d) of NO_2, NH_3, HNO_3, O_3 and SO_2 averaged over the period 2002–2007 for IDAF sites. Vertical bars depict standard deviation over the study period.

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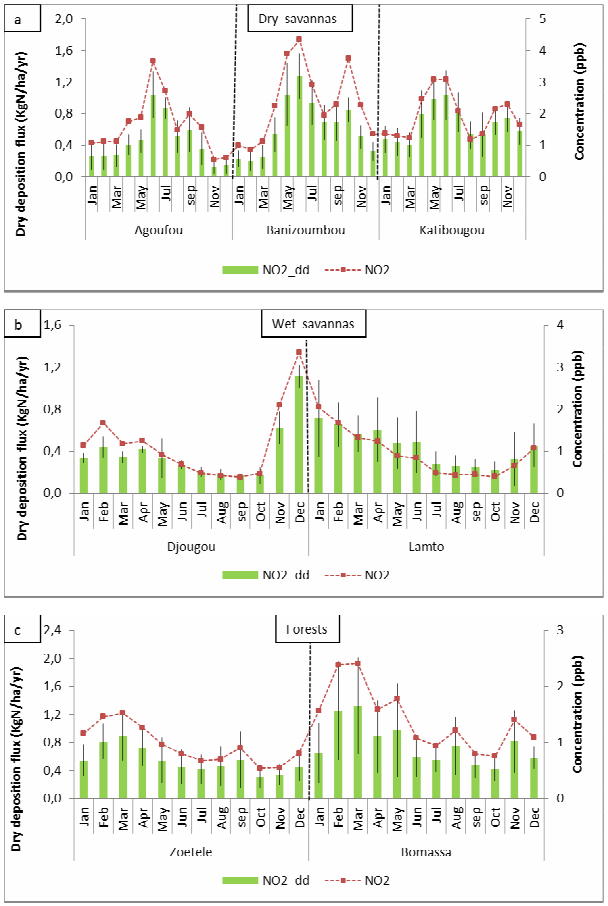

Fig. 4. Evolution of monthly dry deposition fluxes of NO_2 (NO_2_dd) in $kg N ha^{-1} yr^{-1}$ (1998–2007) on the transect dry savannas **(a)**, wet savannas **(b)**, forests **(c)**, associated to evolution of NO_2 concentrations (ppb).

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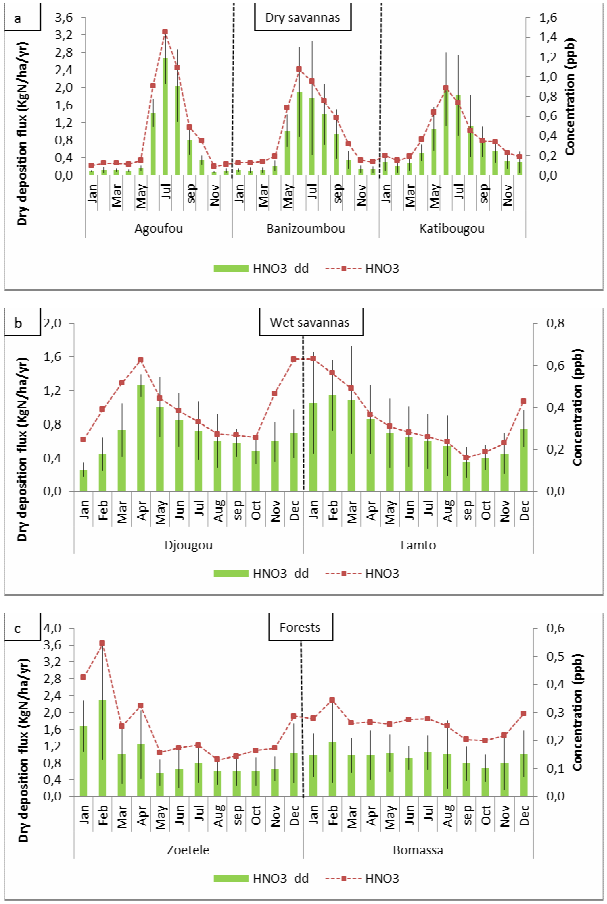

Fig. 5. Evolution of monthly dry deposition fluxes of HNO_3 (HNO_3_dd) in kg N ha^{-1} yr^{-1} (1998–2007) on the transect dry savannas **(a)**, wet savannas **(b)**, forests **(c)**, associated to evolution of HNO_3 concentrations (ppb).

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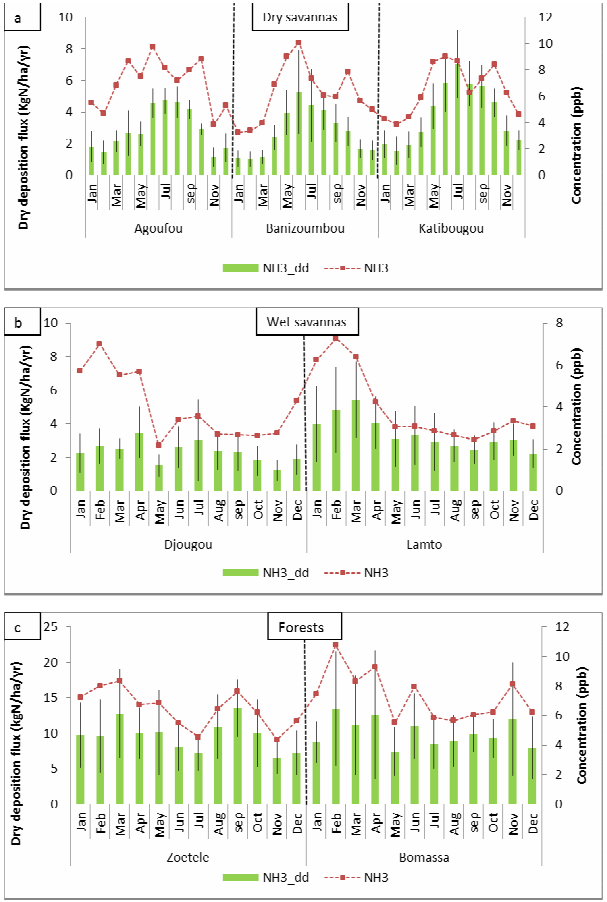

Fig. 6. Evolution of monthly dry deposition fluxes of NH_3 (NH_3_dd) in kg N ha^{-1} yr^{-1} (1998–2007) on the transect dry savannas **(a)**, wet savannas **(b)**, forests **(c)**, associated to evolution of NH_3 concentrations (ppb).

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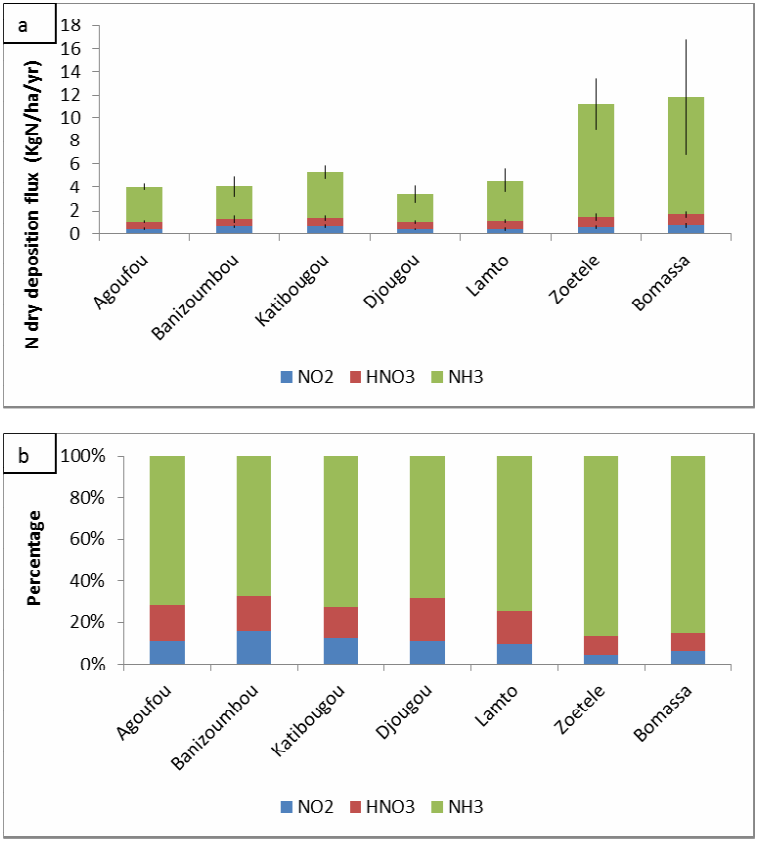

Fig. 7. Annual dry deposition fluxes in $kg N ha^{-1} yr^{-1}$ **(a)** and percentage **(b)** of total N dry deposition fluxes attributed to NO_2, HNO_3 and NH_3 over the period 1998–2007 at IDAF sites.

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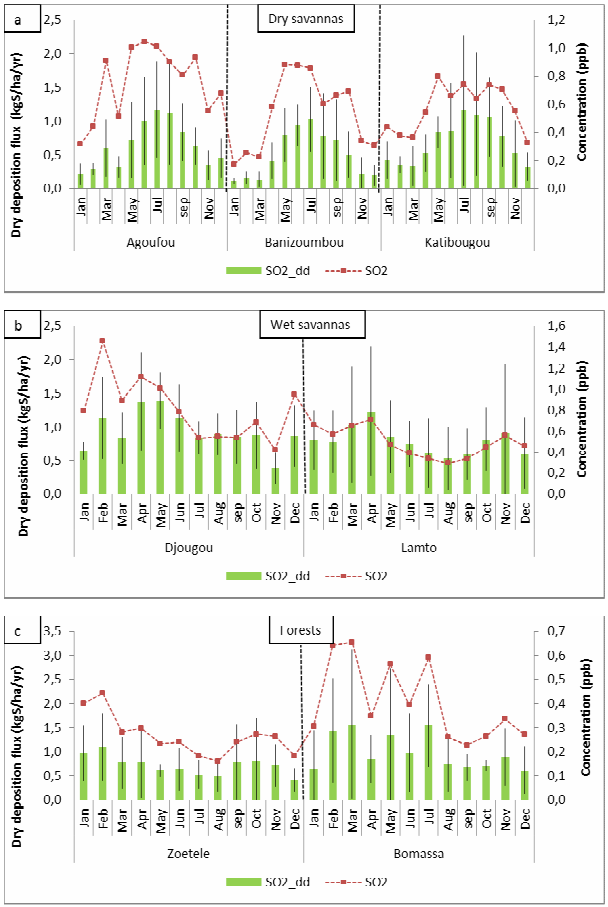

Fig. 8. Evolution of monthly dry deposition fluxes of SO_2 (SO_2_dd) in $kg\,S\,ha^{-1}\,yr^{-1}$ (2002–2007) on the transect dry savannas **(a)**, wet savannas **(b)**, forests **(c)**, associated to evolution of SO_2 concentrations (ppb).

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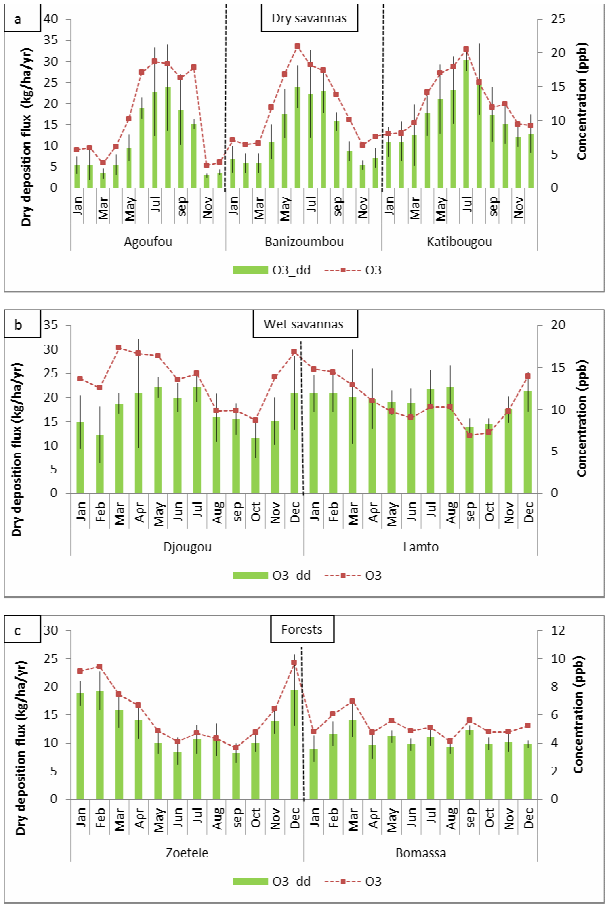

Fig. 9. Evolution of monthly dry deposition fluxes of O_3 (O_3_dd) in kg ha^{-1} yr^{-1} (2001–2007) on the transect dry savannas **(a)**, wet savannas **(b)**, forests **(c)**, associated to evolution of O_3 concentrations (ppb).

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