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Trends and seasonal variability of ammonia across major biomes inferred from long-term series of ground-based and satellite measurements

Money Ossohou[1,2]*, Jonathan E. Hickman[3], Lieven Clarisse[4], Pierre-François Coheur[4], Martin Van Damme[4,5], Marcellin Adon[2,6], Véronique Yoboué[1], Eric Gardrat[7], Maria Dias Alvès[7] and Corinne Galy-Lacaux[7]

[1]Department of Physics, University of Man, Man, Côte d'Ivoire
[2]Laboratoire des Sciences de la Matière, de l'Environnement et de l'Energie Solaire, Université Félix Houphouët-Boigny, Abidjan, Côte d'Ivoire
[3]NASA Goddard Institute for Space Studies, USA
[4]Université Libre de Bruxelles (ULB), Spectroscopy, Quantum Chemistry and Atmospheric Remote Sensing (SQUARES), Brussels, Belgium
[5]Royal Belgian Institute for Space Aeronomy, Brussels, Belgium
[6]Laboratoire des Sciences et Techniques de l'Environnement, Université Jean Lorougnon Guédé, Daloa, Côte d'Ivoire
[7]Laboratoire d'Aérologie, Université Toulouse III Paul Sabatier, CNRS, France

**Correspondence to*: Money Ossohou (ossohoumoney@gmail.com)

Abstract. Ammonia (NH_3) is the most abundant alkaline component in the atmosphere. Changes in NH_3 concentrations have important implications for atmospheric chemistry, air quality, and ecosystem integrity. We present a long-term ammonia (NH_3) assessment in the Western and Central Africa region within the framework of the International Network to study Deposition and Atmospheric chemistry in Africa (INDAAF) program. We analyze seasonal variations and trends of NH_3 concentrations and total columns densities along an African ecosystem transect spanning dry savannas in Banizoumbou, Niger and Katibougou, Mali, wet savannas in Djougou, Benin and Lamto, Côte d'Ivoire, and forests in Bomassa, Republic of Congo and Zoétélé, Cameroon. We use a 21-year record of observations (1998-2018) from INDAAF passive samplers and 11-year record of observations (2008-2018) of atmospheric vertical column densities from the Infrared Atmospheric Sounding Interferometer (IASI) to evaluate NH_3 ground-based concentrations and total column densities, respectively. Annual mean ground-based NH_3 concentrations are around 5.7-5.8 ppb in dry savannas, 3.5-4.7 ppb in wet savannas and 3.4-5.6 ppb in forests. These results suggest that NH_3 emissions from precipitation-induced pulses and volatilization from animal excreta are important emission sources in dry savannas, and biomass burning and agricultural sources are important sources in wet savanna and forest ecosystems. NH_3 total column densities clearly show that the biomass burning source is the most important source in the Lamto wet savanna ecosystem. Annual IASI NH_3 total column densities are 10.1-11.0x10^{15} molec cm^{-2} in dry savanna, 16.5-21.4x10^{15} molec cm^{-2} in wet savanna and 14.3-15.1x10^{15} molec cm^{-2} in forest stations. Non-parametric statistical Mann-Kendall trend tests applied to annual data show that ground-based NH_3 concentrations increase at Bomassa (+2.56% yr^{-1}), but decrease at Zoétélé (-2.95% yr^{-1}) over the 21-year period. The 11-year period of IASI NH_3 total column density measurements show yearly

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increasing trends at Katibougou (+3.98% yr^{-1}) and Djougou (+2.24% yr^{-1}). At Zoétélé, we calculated an increasing trend of leaf area index associated to a significant anticorrelation with ground-based NH_3 concentrations. Leaf area index increase could enhance deposition processes and could contribute to the decrease of ground-based NH_3 concentrations.

1 Introduction

Atmospheric nitrogen (N) compounds play an important role in all compartments of the critical zone (biosphere-atmosphere-hydrosphere) at the global scale. Since 2002, (Bouwman et al., 2002a) had claimed that in the new future, both acidification and eutrophication risks due to excess of N could significantly increase in Asia, Africa and South America, but decrease in North America and Western Europe. Reactive nitrogen (Nr) in the atmosphere, either reduced (NH_x = NH_3 and NH_4^+) or oxidized (NO_x) forms, has a very different role. Ammonia (NH_3), the inorganic form of Nr typically produced through the deprotonation of NH_4^+, is the most abundant alkaline component in the atmosphere (Behera et al., 2013). In the atmosphere, NH_3 influences the abundance and chemical composition of sulfate particles, primarily from dimethyl sulfide (DMS) emissions arising from planktonic algae (Bouwman and Van Der Hoek, 1997). In the lower troposphere, NH_3 neutralizes a great portion of the acids produced by oxides of sulfur and nitrogen (Adon et al., 2010) and forms fine particulate matter (PM2.5) (Malm et al., 2004). Through wet or dry deposition to the surface, NH_3 can be detrimental over time due to an increased toxicity toward sensitive species of plants (Behera et al., 2013; Galloway et al., 2004), ecosystems (Erisman et al., 2013) and soils (Stevens et al., 2018). Different sources contribute to NH_3 emissions on the African continent, which in turn influence the seasonality of atmospheric concentrations and deposition of NH_3.

Soil emissions over north equatorial Africa (2.2 TgN/year) account for almost 70% of African soil emissions, because of the vast areas covered by dry ecosystems (Jaeglé et al., 2004). In the Sahel region, NH_3 emissions can represent an important N flux in natural ecosystems, cropland, grazed soils (Hickman et al., 2018) and bacterial decomposition of urea in animal excreta (Adon et al., 2010). Indeed, many organisms in soils involved in the decomposition of organic matter excrete NH_3 directly or N compounds that readily hydrolyze to NH_x (Bouwman et al., 1997). A minimum level of soil moisture is required for the microbial activities, such as urea hydrolysis, that generates NH_3 (Warner et al., 2017). Atmospheric NH_3 has been reported to be influenced by meteorological and physical parameters such as the presence of plants. Due to high temperatures, low soil moisture and bare soil surfaces conditions, the process of volatilization from soils remains the dominant NH_3 loss in the West African Sahel region (Delon et al., 2010) and Africa contributes to 14% of the global source of NH_3 (Bouwman et al., 1997). Likewise, NH_3 volatilization potential from soil/vegetation systems nearly doubles with every 5 °C increase in air temperature (Sutton et al., 2013; Pinder et al., 2012). However, the capture of NH_3 at the external surface of the leaf and transport into the leaf interior can be an important sink of atmospheric NH_3 (Van Hove et al., 1987).

According to Giglio et al. (2010), ~250 Mha area burned in the Northern Hemisphere and Southern Hemisphere Africa for the time period 1997 through 2008. This value represents on average 70% of the global area burned each year. Many scientific

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papers have shown that biomass burning represents the major source of NH_3 occurring in African savanna and forest ecosystems (Shi et al., 2015; Van Damme et al., 2018). Biomass burning emissions tend to drive seasonal variation in NH_3 total column densities in West Africa, with the largest emissions occurring late in the dry season and early rainy season (Hickman et al., 2021).

Satellite measurements of NH_3 provide a means to monitor atmospheric composition globally (Clarisse et al., 2009; Warner et al., 2017) and is a powerful tool for understanding atmospheric composition particularly for regions like Africa, where other types of measurements are scarce (Hickman et al., 2018). During the year 2008, Hickman et al. (2018) found elevated total columns of NH_3 from the Infrared Atmospheric Sounding Interferometer (IASI) in the Sahel during March-April mainly due to the Birch effect. Through recent improvements in retrieval algorithms, Van Damme et al. (2021) used the version 3 of the IASI-NH_3 total columns dataset to characterize the evolution of atmospheric NH_3 at global, national and regional scales over the 11-year period (2008-2018). Using a statistical trend method based on least squares regression and bootstrap resampling, (Van Damme et al., 2021) found large increases of NH_3 in several subcontinental regions over the last decade, especially in western and central Africa (29.0 ± 2.3 %.decade^{-1}).

Based on a 10-year period of ground-based measurements within the framework of the International Network to Study Deposition and Atmospheric Chemistry in Africa (INDAAF) network, Adon et al. (2010) documented surface concentrations and seasonal cycles according to the atmospheric sources of NH_3 in West and Central Africa. INDAAF has been a long-term monitoring measurement network since 1995 to document atmospheric chemistry and deposition fluxes in Africa. This program is part of the European Aerosol, Clouds and Trace Gases Research Infrastructure-France (ACTRIS-FR) and of the International Global Atmospheric Chemistry / Deposition of Biogechemically Important Trace Species (IGAC/DEBITS) activity. In addition, it is a labeled contributing network to the Global Atmospheric Watch/ World meteorological Porgram (GAW/WMO) program. Here we provide updated analyses of these long-term records, complemented with satellite retrievals, to better understand 21st century NH_3 dynamics in Africa.

Specifically, in the framework of the INDAAF program, this study aims to improve long-term NH_3 assessment in the Western and Central Africa region. We first compare the monthly and seasonal patterns in ground-based NH_3 concentrations (1998/2005-2018) and IASI NH_3 total columns (2008-2018) measured at three major African ecosystems: dry savannas, wet savannas and forests. Monthly and seasonal evolutions allow us to highlight the main sources and factors influencing atmospheric NH_3 levels in the tropical African ecosystems. Secondly, we use non-parametric statistically robust tests to assess long-term trends of NH_3 from surface and satellite measurements over the ecosystemic transect, and discuss results according to the analysis of sources seasonality and meteorological data trends.

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2 Material and methods

2.1 Presentation of sampling sites

Figure 1 shows the location of the 8 labeled INDAAF monitoring stations situated in West and Central Africa. Each site represents an African regional ecosystem with its own characteristics in terms of emission sources and its sensitivity to climatic, ecological, and anthropogenic changes. Thus, the sites are distributed by pairs according to latitudinal bands with significant different rainfall patterns to represent dry savanna (Banizoumbou in Niger, Katibougou in Mali), wet savanna (Djougou in Benin, Lamto in Côte d'Ivoire) and equatorial forest (Bomassa in Republic of Congo, Zoétélé in Cameroon) ecosystems. Additional details on the monitoring sites can be found in the literature (Abbadie, 2006; Adon et al., 2010; Akpo et al., 2015; Delmas et al., 1995; Diawara et al., 2014; Le Roux et al., 2006; Ossohou et al., 2019; Ouafo-Leumbe et al., 2018; Yoboué et al., 2005). To date, measurements of atmospheric and meteorological physico-chemical parameters are continuing at all the INDAAF sites. These measurements are referenced in the INDAAF database (http://indaaf.obs-mip.fr) and in the WMO OSCAR database (https://oscar.wmo.int/surface/#/).

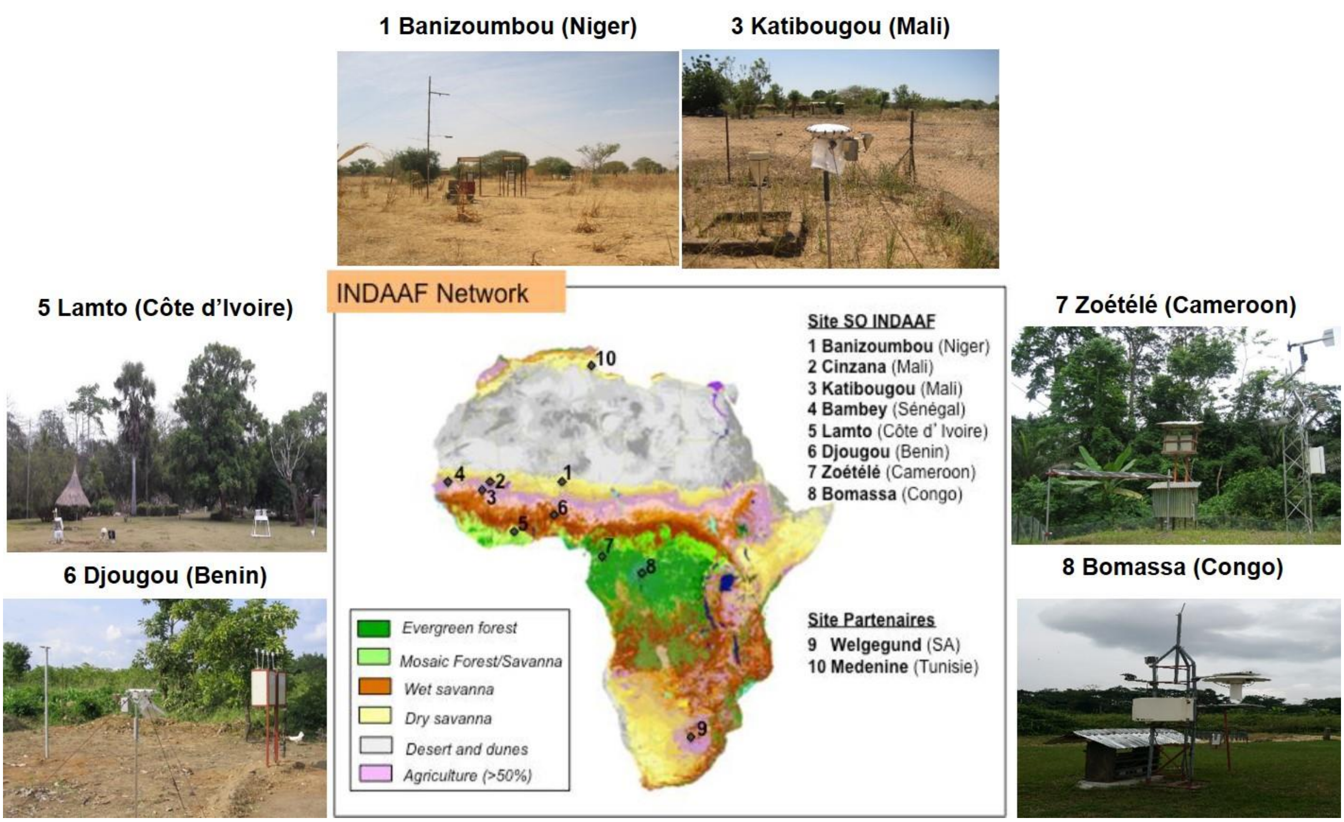

Figure 1. INDAAF measurement network composed by 10 stations across Africa location. Presentation of the stations of (1) Banizoumbou (Niger), (3) Katibougou (Mali), (5) Lamto (Côte d'Ivoire), (6) Djougou (Benin), (7) Zoétélé (Cameroon) and (8) Bomassa (Republic of Congo) stations (Adapted from Mayaux et al. (2004); Ossohou et al. (2019)).

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The geographical characteristics, soil, vegetation, climate types and the months representative of the wet and dry seasons of the western and Central African sites of interest are described in Table 1. It is important to keep in mind that dry savannas are characterized by a short wet season from June to September, whereas the wet season is longer in wet savanna and forest ecosystems extending from April to October and March to November, respectively.

Tableau 1. Site coordinates and location information (WS: wet season; DS: dry season). Dry savannas (WS: June–September DS: October–May), wet savannas (WS: April-October DS: November–March), forest (WS: March–November DS: December–February).

Ecosystems	Station	Latitude, Longitude	Type of soil and/or vegetation	Climate	Country
Dry savannas	Banizoumbou (Adon et al., 2013; Delon et al., 2012; de Rouw and Rajot, 2004)	13°31’ N, 02°38’ E	91.2% Sandy soils, Tiger bush – fallow bush	Sahelian	Niger
	Katibougou (Adon et al., 2013; Delon et al., 2012)	12°56’ N, 07°32’ W	Sandy soils, Deciduous shrubs	Sudano-Sahelian	Mali
Wet savannas	Djougou (Akpo et al., 2015; Ouafo-Leumbe et al., 2018)	09°39’ N, 01°44’ E	Ferralitic and ferruginous soil, Mosaic of dry forests and savannah	Sudano-Guinean	Benin
	Lamto (Abbadie, 2006; Yoboué et al., 2005)	06°13’ N, 05°02’ W	Ferrugineous soils, Grass, shrub and tree stratum	Guinean	Côte d’Ivoire
Forests	Bomassa (Mitani et al., 1993)	02°12’ N, 16°20’ E	Dense evergreen forest	Equatorial	Republic of Congo
	Zoétélé (Sigha et al., 2003)	03°15’ N, 11°53’ E	Dense evergreen forest	Equatorial	Cameroon

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2.2 NH_3 sampling and chemical analysis

Monitoring of NH_3 in the framework of the INDAAF program began in 1998 (2005 for Djougou). Sampling was carried out using the INDAAF passive sampler technique inspired by the work of (Ferm, 1991). The passive samplers were mounted and analyzed at the "Laboratoire d'Aérologie (LAERO)" in Toulouse (France) for all INDAAF sites.

Adon et al. (2010) give a complete overview of the sampling and analytical procedures for the INDAAF passive sampler technique. For each INDAAF site, the passive samplers were made of impregnated filter paper with a species-specific solution for adsorption of gases. Samplers are exposed during one month in duplicates to ensure reproducibility and monthly concentrations are calculated from the arithmetic mean of the duplicates. Desorbed filters are analyzed using ion chromatography (IC). The Laboratoire d'Aérologie participates twice a year in WMO's quality assurance intercomparison program. Results have always shown that the analytical accuracy of the IC realized at the LAERO is greater than 95%. The intercomparison results of the LAERO are available under the reference 700106 on the WMO website (http://qasac-americas.org/). The sampling technique using the INDAAF passive sampler method has been validated on tropical, subtropical, rural and urban sites in Africa (Adon et al., 2010; Bahino et al., 2018; Ossohou et al., 2020). INDAAF passive samplers have proven to be accurate, cheaper, easy to use and useful for air quality monitoring.

The measurement accuracy of passive samplers, evaluated through covariance with duplicates, was estimated at 14.3% for NH_3 (Adon et al., 2010). Detection limit for NH_3 was calculated from field blanks and is equal to 0.7±0.2 ppb. Values below the detection limit, as well as non-valid reproducibility values, were removed from the database. Thus, the percentages of valid data in the final database for the studied period 1998/2005-2018 were 97% for Banizoumbou, 93% for Katibougou, 90% for Djougou, 94% for Lamto, 73% for Bomassa and 93% for Zoétélé.

2.3 Biomass burning emissions of NH_3

The fourth version of the Global Fire Emissions Database (GFED4) provides monthly biomass burning emissions at 0.25° resolution since 1997 from all biomass burning sources, i.e. many sectors (agricultural waste burning, boreal forest fires, peat fires, savanna fires, grassland fires, shrubland fires, temperate forest fires and tropical deforestation and degradation). Emissions of NH_3 from biomass burning sources were downloaded for the 1° x 1° grid cell centered on each INDAAF site. The GFED4 emissions are based on the combination of satellite information on fire activity and vegetation productivity to estimate gridded monthly burned area and fire emissions, as well as scalars that can be used to calculate higher temporal resolution emissions (Giglio et al., 2013; van der Werf et al., 2017). The Global Fire Emissions Database—currently by far the most widely used global fire emissions inventory—has been widely cited in the literature, and GFED4 data can be downloaded from the Emissions of atmospheric Compounds and Compilation of Ancillary Data (ECCAD) database (https://eccad3.sedoo.fr/).

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2.4 IASI NH_3 total columns and TRMM measurements

IASI-A, launched aboard the European Space Agency's Metop-A in 2006, provides measurements of atmospheric NH_3 twice a day (9:30 in the morning and evening, Local Solar Time at the equator) (Clarisse et al., 2009). Here we use morning observations, when the thermal contrast is more favorable for infrared retrievals in the lowest layers of the atmosphere (Clarisse et al., 2010; Van Damme et al., 2014). The NH_3 retrieval product used (ANNI-NH_3-v3R) follows a neural network retrieval approach. We refer to Whitburn et al. (2016) and Van Damme et al. (2017, 2021) for a detailed description of the algorithm. Only observations with cloud cover below 10% were used. Given the absence of hourly or even daily observations of NH_3 concentrations in sub-Saharan Africa, the detection limit of IASI is difficult to determine with certainty. We regridded the Level-2 IASI NH_3 product to 0.25° × 0.25° resolution. The IASI products have been validated based on aircraft and ground-based measurements. The IASI version 3 dataset compare well with the reconstructed in-situ columns, with the reanalysed dataset used in this study presenting a better agreement (Guo et al., 2021). Previous validation work comparing older versions of the IASI product with ground-based Fourier transform infrared (FTIR) observations of NH_3 total columns has also shown robust correlations at sites with high NH_3 concentrations, but lower at sites where atmospheric concentrations approach IASI's detection limits (Dammers et al., 2017). Although FTIR observations are absent from Africa, earlier works have shown fair agreement between previous versions of IASI total column densities and INDAAF NH_3 surface observations in West Africa (Van Damme et al., 2015) and seasonal pattern (Hickman et al., 2018; Ossohou et al., 2019). In the study, the IASI NH_3 reanalysed product was used for the years 2008 —the first full year of data available—to the end of 2018.

We also used the Tropical Rainfall Measuring Mission (TRMM) daily precipitation product (3B42), which is based on a combination of TRMM observations, geo-synchronous infrared observations, and rain gauge observations (Huffman et al., 2007). Independent rain gauge observations from West Africa have been used to validate this precipitation product, with no indication of bias (Nicholson et al., 2003).

For analyses of seasonal and interannual variability in each product, we used the mean monthly value (for NO_2 and NH_3) or the monthly sum (for precipitation) for a 1° x 1° grid cell containing each INDAAF site.

2.5 Trend analysis

Trend analyses were carried out by using Mann-Kendall (MK) (Kendall, 1975; Mann, 1945) and Seasonal Kendall (SK) (Hirsch et al., 1982) which are statistical non-parametric tests used to determine the increasing or decreasing trends of a random variable over some period of time. The MK and SK tests were suitable for cases with monotonic trends. The MK test allows working with no seasonal or other cycles in the data such as average annual data. The SK test follows the same principle as the MK test and is significantly robust to seasonality and was therefore applied for monthly time series. The SK test takes into account a 12-month seasonality in the time series data by computing the MK test on each dataset of "months" over the period

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1998/2005-2018 separately, and then combining the results (Tang et al., 2018). MK and SK tests allow working with non-normal data, in situations with many missing values, and are resistant to outliers (Kumar et al., 2018).

We coupled MK and SK tests respectively to Sen's Slope (SS) (Sen, 1968) and Seasonal Kendall Slope (SKS) to estimate the magnitude of the trend. These statistical tests have been widely applied and described in the literature to estimate trend in environmental parameters (Shadmani et al., 2012; Yue et al., 2002; Yue and Wang, 2004), while the application over African rural sites are limited (Ossohou et al., 2019, 2020). Two-tailed tests are conducted with the statistical software R version 4.0.4 (R Core Team, 2021) and (Addinsoft, 2022) for this study.

3 Results and Discussion

3.1 Variations of ground-based NH_3 and IASI NH_3

In the first part of this subsection, we will present the NH_3 concentration variations in the dry savanna ecosystem. In the second part, we will present the same variations for wet savanna and forest sites. Each part will show the monthly, annual evolutions and descriptive statistics of NH_3 ground-based and satellite measurements at each site. At the end of each of these two parts, we will discuss the results obtained according to the sources and the major processes that influence the atmospheric NH_3 levels.

3.1.1 Dry savanna

Figure 2 presents monthly variations of ground-based NH_3 concentrations and IASI NH_3 at the INDAAF dry savanna sites of Banizoumbou (Figure 2a) and Katibougou (Figure 2b) over 1998-2018 and 2008-2018, respectively. The monthly 21-year surface concentrations of NH_3 are in the same range at Banizoumbou and Katibougou (Table 2) with coefficients of variation of ~50%. Nethertheless, the monthly coefficient of variation of IASI NH_3 total columns appear to be larger at Banizoumbou (57%) compared to Katibougou (46%) over the 11-year period. We obtain a significant Pearson's correlation between monthly ground-based NH_3 concentrations and IASI NH_3 total columns at Banizoumbou ($r=0.30$, $p<0.001$), but not at Katibougou ($r=0.05$, $p=0.52$).

Table 2 shows that mean ground-based concentrations of NH_3 for each dry savanna site are significantly higher in wet season compared to dry season according to the t-test *($p=0.003$)*.

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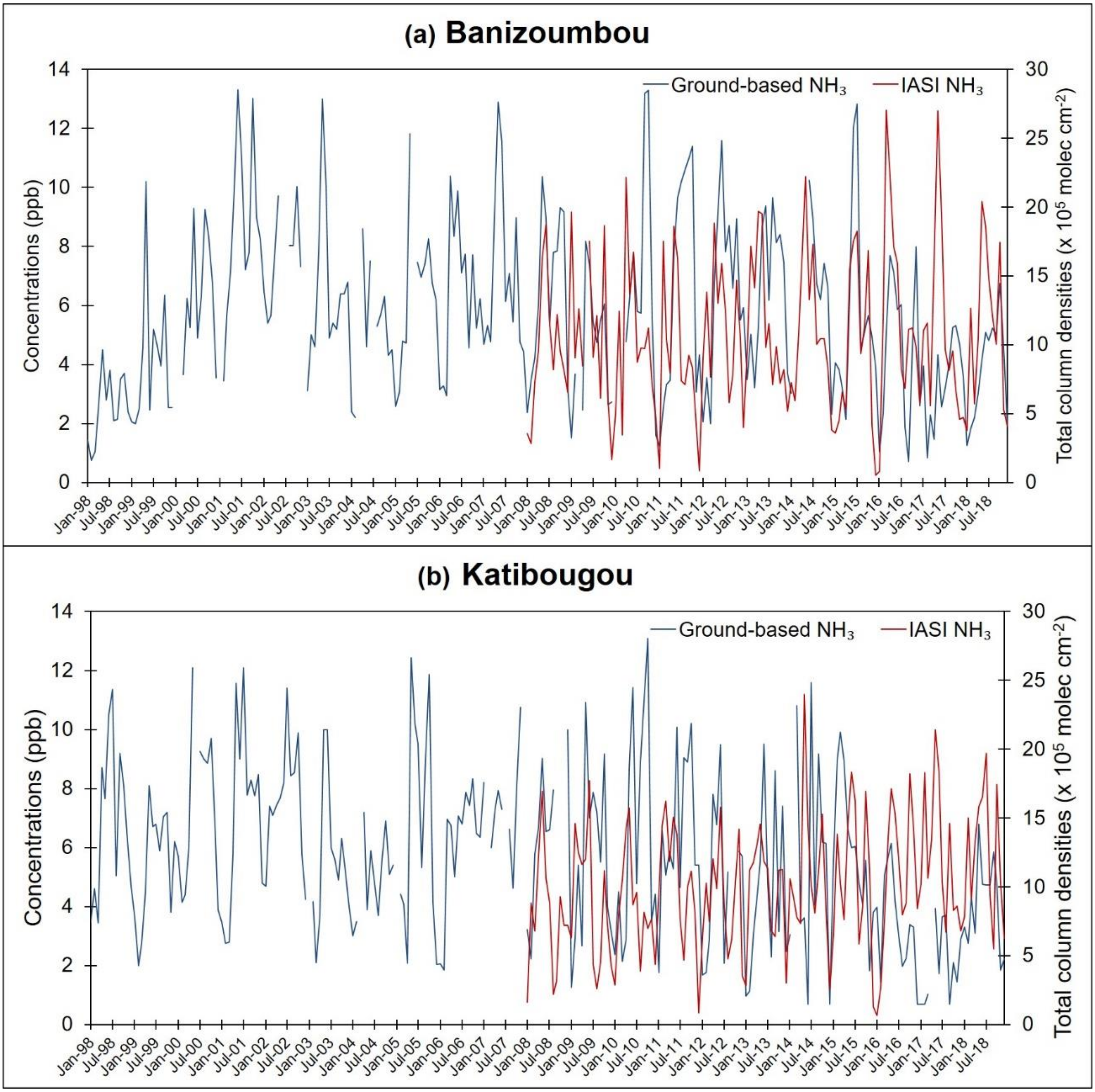

210 **Figure 2.** Monthly time-series of ground-based NH_3 concentrations over the period 1998–2018, and IASI NH_3 total column densities from 2008 to 2018 at (a) Banizoumbou, Niger and (b) Katibougou, Mali.

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Tableau 2. Minimum (Min), maximum (Max) and average (Avg) monthly, annual and seasonal ground-based NH_3 concentrations (1998–2018), and IASI NH_3 total column densities (2008–2018) at Banizoumbou, Niger and Katibougou, Mali

		Ground-based NH_3 (*ppb*)		**IASI NH_3 (*10^{15} molec cm^{-2}*)**	
		Banizoumbou	**Katibougou**	**Banizoumbou**	**Katibougou**
Monthly	Min	0.7	0.7	0.5	0.7
	Max	13.3	13.1	27.0	24.0
Annual	Avg	5.8±1.2	5.7±1.1	11.0±1.0	10.1±1.2
Wet Season	Min	2.7	2.4	9.6	6.2
	Max	10.4	9.3	14.1	12.8
	Avg	6.9±1.6	6.7±1.5	11.7±1.2	9.8±1.9
Dry Season	Min	2.5	1.8	7.1	8.8
	Max	8.1	8.0	14.9	12.2
	Avg	5.2±1.1	5.2±1.0	10.7±1.8	10.3±0.8

Figure 3 compiles the mean monthly 21-year average ground-based concentrations (solid lines) and 11-year average total column densities (dashed lines) in dry savanna ecosystems to obtain the mean annual cycle evolutions of NH_3 at the stations of Banizoumbou (a) and Katibougou (b). In both sites of the sub Saharan dry ecosystems, we observe a marked seasonal cycle with two peaks in ground-based concentrations and total columns of NH_3 appearing at the beginning (May-June) and the end (October) of the wet season (Figure 3). The lowest values of NH_3 (concentrations and densities) are generally observed during December-January and August. Mean monthly measurements vary at Banizoumbou from 2.8±1.1 ppb (January) to 8.3±2.6 ppb (June) for ground-based concentrations, and from 3.8±1.5 x 10^{15} molec cm^{-2} (January) to 20.6±5.6 x 10^{15} molec cm^{-2} (May) for IASI NH_3 total columns (Figure 3a). At Katibougou, mean monthly ground-based concentrations ranged from 3.3±1.3 ppb (January) to 7.6±2.1 ppb (June), and from 4.4±2.0 x 10^{15} molec cm^{-2} (December) to 16.1±2.7 x 10^{15} molec cm^{-2} (May) for IASI NH_3 total columns (Figure 3b). The mean annual variation coefficients are 34% and 27% for ground-based concentrations, 41% and 35% for IASI NH_3 total column measurements at Banizoumbou and Katibougou, respectively.

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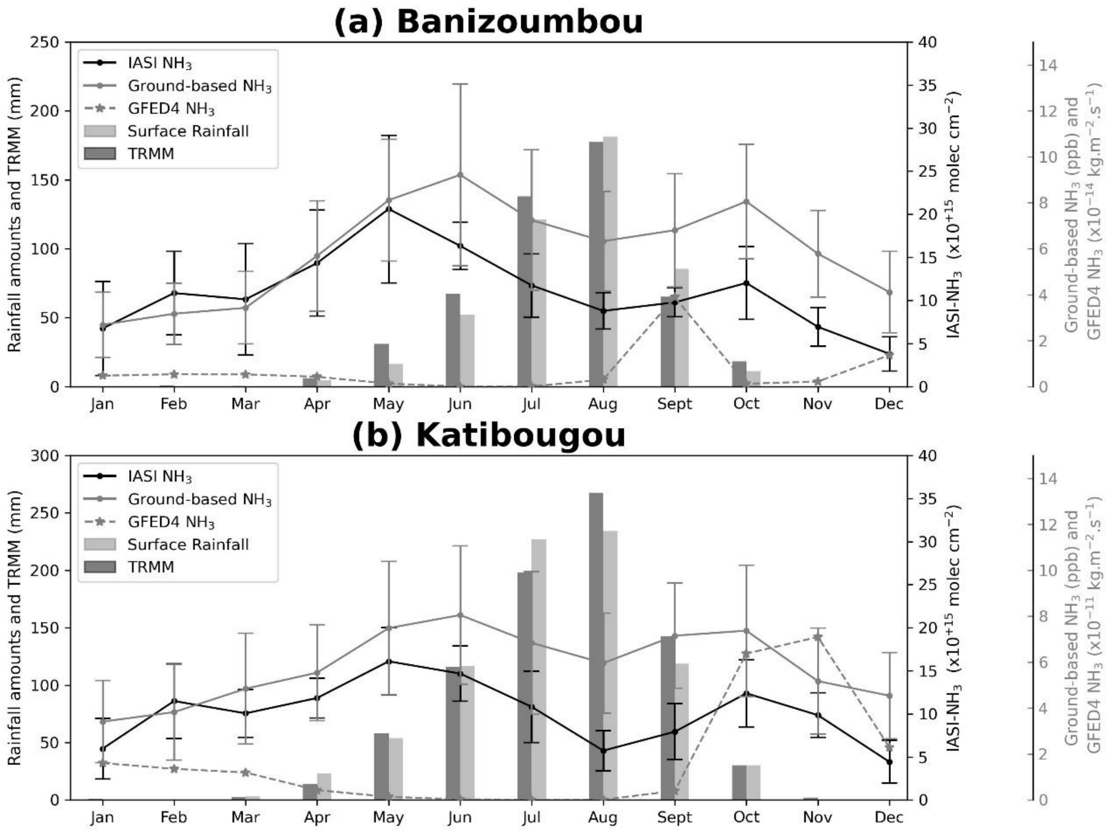

Figure 3. Mean monthly ground-based NH_3 concentrations (1998–2018), IASI NH_3 total column densities (2008–2018), rainfall amount measured by ground-based instruments (1998-2018), TRMM (2005-2018) and emissions of NH_3 from GFED4 database (1998-2018) at (a) Banizoumbou, Niger and (b) Katibougou, Mali. Error bars represent the mean absolute deviation.

In the Sahelian region, major sources of atmospheric NH_3 include bacterial decomposition of urea in livestock excreta and emission from natural or fertilized soils (Bouwman and Van Der Hoek, 1997). In addition, it has been shown in the literature that African dry savanna ecosystems, characterized by sandy soils, tiger bush, fallow bush and deciduous shrubs (Ossohou et al., 2019), tend to have alkaline soils, creating favorable conditions to NH_3 volatilization (Clarisse et al., 2019; Delon et al., 2017; Hickman et al., 2018; Vågen et al., 2016). Among other factors, rainfall pattern and amount influence NH_3 emissions considerably in drylands like Banizoumbou and Katibougou. The seasonal distributions of NH_3 concentrations look like when the weather is its most warm and dry, NH_3 concentrations are lowest meaning that during the dry season, there's not much biological activity in soils, and so NH_3 emissions are lower than in the rainy season.

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It's also important to highlight the pastoralism in the Sahel region, mainly nomadic in nature. Indeed, Sahelian agro-pastoralism appears to be very important, representing 25 to 30% of the Gross Production Product (GDP), and contributes to 10 to 15% of the GDP of Mali and Niger for example (Adon et al., 2010).

In the dry savannas, soils are often characterized by large pulses of NH_3 related to successive dryings and rewettings of dry soils (McCalley and Sparks, 2008; Soper et al., 2016). As we can see in figure 3, the first peak observed in May-June (beginning of the wet season) could be related to the optimal soil moisture to initiate bacterial activity and a flush of newly mineralized N. Our results support the conclusions of an earlier study that used satellite retrievals and in situ measurements for the year 2008 over Africa to argue that the onset of the rainy season causes pulsed emissions of NH_3 over the Sahel (Hickman et al., 2018). This study based on ground-based and satellite measurements ranging from one to two decades clearly shows a correspondence between early rainy season precipitation and NH_3 concentrations over the two dry savanna sites. Moreover, the results based on our analysis of a long-term database clearly indicates that this process is reproducible every year.

The temporal evolution of NH_3 can be associated with two most important phenomena: (1) Possible Birch effect emissions in the early and possibly late rainy season, and (2) the overall seasonal cycle of NH_3 and the reasons for this broad seasonality (separate from the Birch effect). Indeed, during the wet season (June-September), the months are wetter and cooler and give the soils more moisture than the dry season months. As a result, wet season soils are less susceptible to intense drying events than during the dry season. This consequently results in more limited NH_3 volatilization from soil drying, leading to low NH_3 levels in the wet season. However, at the end of the wet season, rainfall became erratic and led to drying soils for a few days. This erratic rainfall may explain the second observed NH_3 peak, occurring at the end of the wet season. A similar late-season pulse of nitric oxide from the re-wetted soils was observed at the regional scales in the Sahel (Jaeglé et al., 2004), suggesting that there may be some similar potential for NH_3 pulsing from re-wetted dry soils. This late-season peak appears to be of less importance than the early wet season peak, presumably because over the growing season, growing vegetation, and microbial communities that immobilized and reduced soils nitrogen pools and may continue to do so. One of the arguments for why Birch effect emissions happen at the beginning of the growing season is that there has been an accumulation of labile N in soils in the dry season. During the wet season, NH_3 is found directly in the rainwater in the form of NH_4^+, thus promoting wet deposition on the growing vegetation. NH_3 also react with some acid gases such as H_2SO_4, HNO_3 and HCl to form aerosols of atmospheric ammonium salts, such as ammonium sulphate ($[NH_4]_2SO_4$), ammonium bisulphate (NH_4HSO_4), ammonium nitrate (NH_4NO_3) and ammonium chloride (NH_4Cl). The conversion of gases to particles in the atmosphere can occur through condensation and/or direct nucleation processes (Baek et al., 2004). Condensation adds mass to pre-existing aerosols, while direct nucleation allows the formation of atmospheric aerosols from gaseous precursors. These reactions could therefore lead to a decrease in atmospheric NH_3 concentrations in the Sahelian region (Koziel et al., 2006).

We note that the GFED4 inventory shows important NH_3 emissions by all biomass burning sources from September through March in the dry savanna sites (Figure 3). Since these months correspond to the fire period in the wet savanna and forest sites, we suggest that even though the two dry savanna sites experience few fires, NH_3 columns from IASI are certainly affected by NH_3 present in the transported fire plumes.

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3.1.2 Wet savanna and forest

In the wet savanna ecosystem, we present the monthly evolutions of ground-based NH_3 concentrations (2005-2018: Djougou, 1998-2018: Lamto) and IASI NH_3 total column densities (2008-2018 for both sites) at Djougou (Figure 4a) and Lamto (Figure 4b). Monthly ground-based NH_3 concentrations range from 0.7 to 12.1 ppb at Djougou and from 0.7 to 8.9 ppb at Lamto. IASI NH_3 total column densities vary from 0.7 x 10^{15} to 36.6 x 10^{15} molec cm^{-2} at Djougou and from 0.3 x 10^{15} to 55.6 x 10^{15} molec cm^{-2} at Lamto. The results show that the maxima of ground-based NH_3 concentrations are generally the most important at Djougou (Figure 4), while those of IASI NH_3 total column densities are highest at Lamto (Figure 4). The coefficients of variation are globally high, equal to 57% and 62% for ground-based measurements, and 51% and 68% for IASI NH_3 total columns at Djougou and Lamto, respectively. For the entire period of measurements, Pearson correlation test applied to monthly ground-based NH_3 concentrations and IASI NH_3 total columns reveals no significant correlation at Djougou ($r=0.04$, $p=0.68$), but strong linear correlation at Lamto ($r=0.59$, $p<0.01$).

Table 3 presents a synthesis of monthly, seasonal and annual minimum, maximum and average ground-based NH_3 concentrations and IASI NH_3 total columns at Djougou and Lamto stations. The results show that mean annual, wet season and dry season ground-based NH_3 concentrations in Djougou are significantly higher than that in Lamto (t-test, $p<0.05$). In contrast, mean annual and dry season IASI NH_3 total columns are significantly more important (*t-test*, $p<0.01$) at Lamto compared to Djougou (Table 2).

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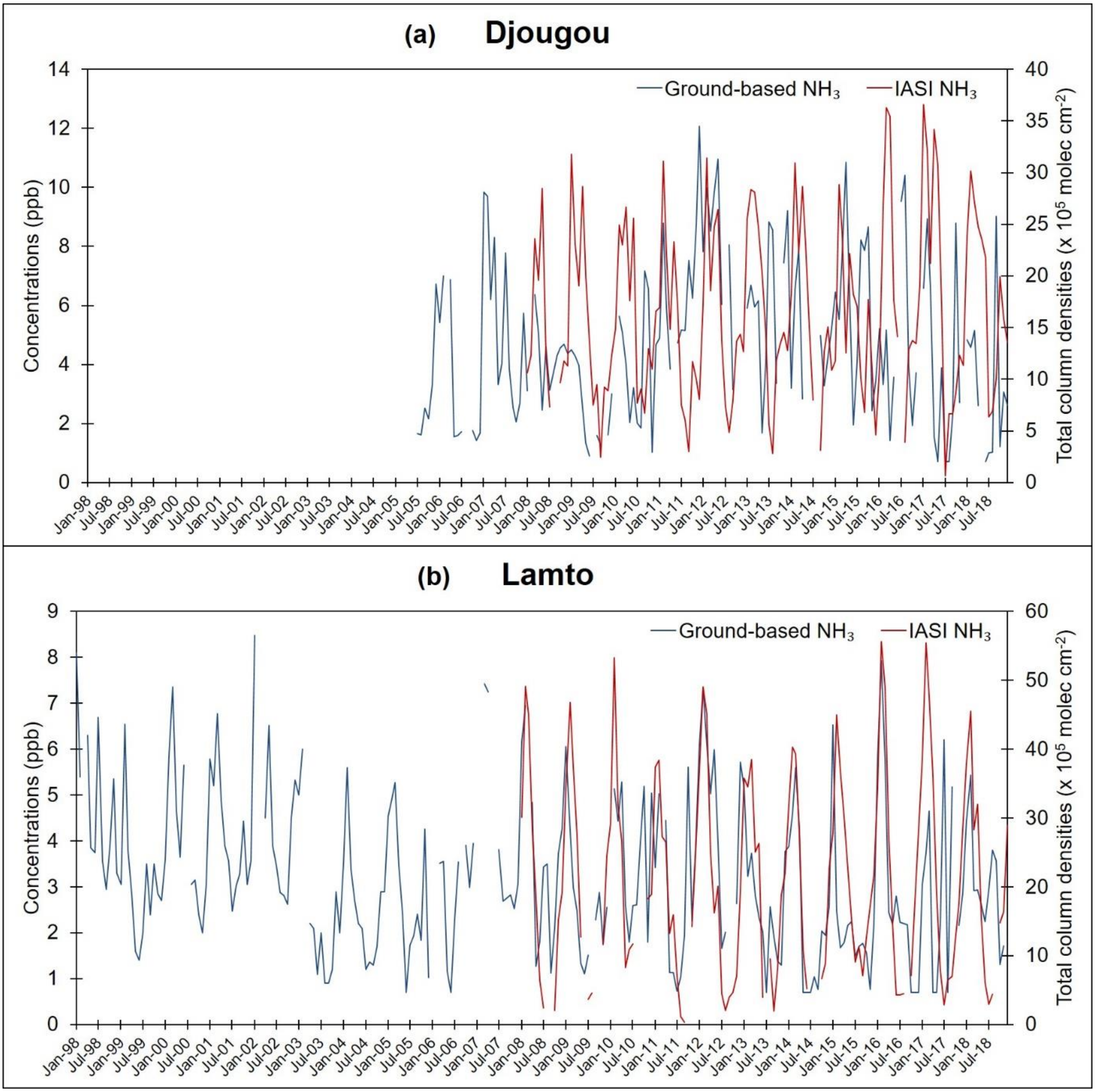

Figure 4. Monthly time-series of ground-based NH_3 concentrations over the periods 2005-2018 and 1998–2018, and IASI NH_3 total column densities from 2008 to 2018 at (a) Djougou, Benin and (b) Lamto, Côte d'Ivoire.

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295 **Tableau 3.** Minimum (Min), maximum (Max) and average (Avg) monthly, annual and seasonal ground-based NH_3 concentrations (Djougou : 2005-2018; Lamto : 1998–2018), and IASI NH_3 total column densities (2008–2018) at Djougou, Benin and Lamto, Côte d'Ivoire

		Ground-based NH_3 (*ppb*)		**IASI NH_3 (*10^{15} molec cm^{-2}*)**	
		Djougou	**Lamto**	**Djougou**	**Lamto**
Monthly	Min	0.7	0.7	0.7	0.3
	Max	12.1	8.5	36.6	55.6
Annual	Avg	4.7±1.3	3.5±0.8	16.5±1.2	21.4±1.0
Wet Season	Min	1.5	1.5	11.9	10.4
	Max	7.6	4.5	16.3	15.5
	Avg	4.1±1.5	2.8±0.9	14.6±1.1	12.5±1.2
Dry Season	Min	3.5	2.7	13.9	28.5
	Max	8.8	7.8	22.7	37.5
	Avg	5.5±1.3	4.6±1.1	19.2±2.1	31.6±2.1

Figure 5 presents the annual mean cycle of monthly ground-based concentrations and IASI NH_3 total column densities at Djougou (Fig. 5a) and Lamto (Fig. 5b) located in the wet savanna ecosystem. The results show that the annual mean ground-based and IASI NH_3 profiles have a poor covariation at Djougou (Fig. 5a), while IASI NH_3 have good agreement well the evolution of ground-based NH_3 at the Lamto site (Fig. 5b). Ground-based NH_3 concentrations and IASI NH_3 total columns exhibit a clear seasonality at Lamto station with higher values occurring in the dry season (January to February) and lower values in the wet season (May through November). Mean annual cycle of ground-based NH_3 concentrations and IASI NH_3 total column densities seasonality are less marked at Djougou (maximum from January to April) compared to Lamto station. Monthly mean concentrations and total column densities of NH_3 range from 3.2±1.4 (June) to 6.7±1.8 ppb (February) and from 6.6±2.0 x 10^{15} (August) to 27.3±4.0 x 10^{15} molec cm^{-2} (February) at Djougou (Fig. 5a), and from 2.2±1.0 (June) to 6.1±1.8 ppb (February) and from 5.4±2.6 x 10^{15} (July) to 46.6±5.4 x 10^{15} molec cm^{-2} (February) at Lamto (Fig. 5b), respectively. The mean annual variation coefficients are 23% and 41% for ground-based concentrations, 46% and 72% for IASI NH_3 total column measurements at Djougou and Lamto, respectively.

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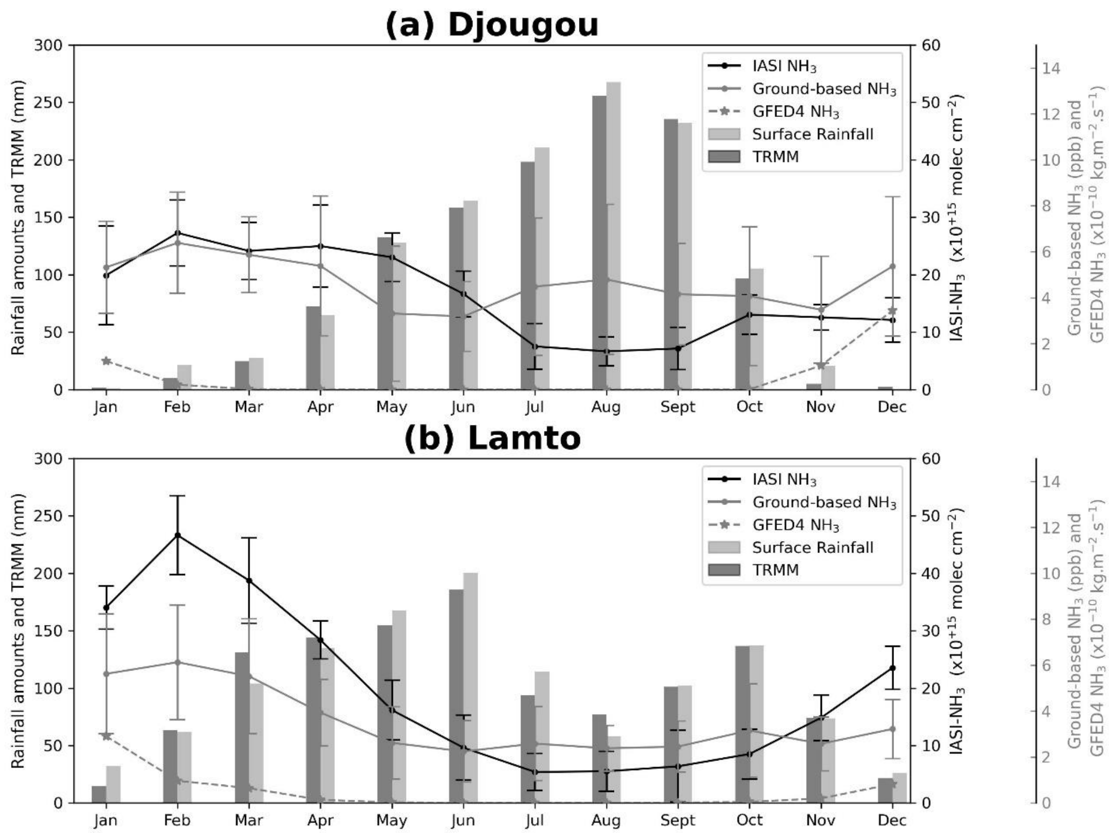

Figure 5. Mean monthly ground-based NH_3 concentrations (Djougou : 2005-2018 & Lamto : 1998–2018), IASI NH_3 total column densities (2008–2018), rainfall amount measured by ground-based instruments (1998-2018), TRMM (2005-2018) and emissions of NH_3 from GFED4 database (Djougou : 2005-2018 & Lamto : 1998–2018) at (a) Djougou, Benin and (b) Lamto, Côte d'Ivoire. Error bars represent the mean absolute deviation.

The monthly variations of ground-based NH_3 concentrations (1998-2018) and IASI NH_3 total column densities (2008-2018) over the two forested monitoring sites are presented in Figures 6. The results show that the peak values of ground-based concentrations and IASI NH_3 total column densities are larger at Bomassa (Figure 6a) compared to Zoétélé (Figure 6b). The monthly 21-year coefficients of variation of NH_3 are in the same order of magnitude at Bomassa (55%) and Zoétélé (56%). Nethertheless, the monthly coefficient of variation of IASI NH_3 total column densities are more important in the forested ecosystem compared to dry and wet savannas, i.e more than 80% at Bomassa and Zoétélé over the 11-year period. Significant

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Pearson's correlations are found between monthly ground-based NH_3 and IASI-NH_3 total column densities at Bomassa ($r = 0.23$, $p = 0.03$) and Zoétélé ($r = 0.36$, $p < 0.001$).

The monthly, seasonal and annual measurement results of ground-based NH_3 concentrations and IASI NH_3 total columns at Bomassa and Zoétélé are summarized in Table 4. According to the t-test, ground-based NH_3 average concentrations are significantly higher ($p<0.001$) at Bomassa compared to Zoétélé, but IASI NH_3 total column average densities are in the same order of magnitude for these sites. For each forested ecosystem station, the results show that the mean ground-based NH_3 concentrations are in the same order of magnitude between wet and dry seasons. However, IASI total column densities are significantly higher (*t-test*, $p<0.001$) in the dry season at Bomassa and Zoétélé compared to the wet season (Table 4).

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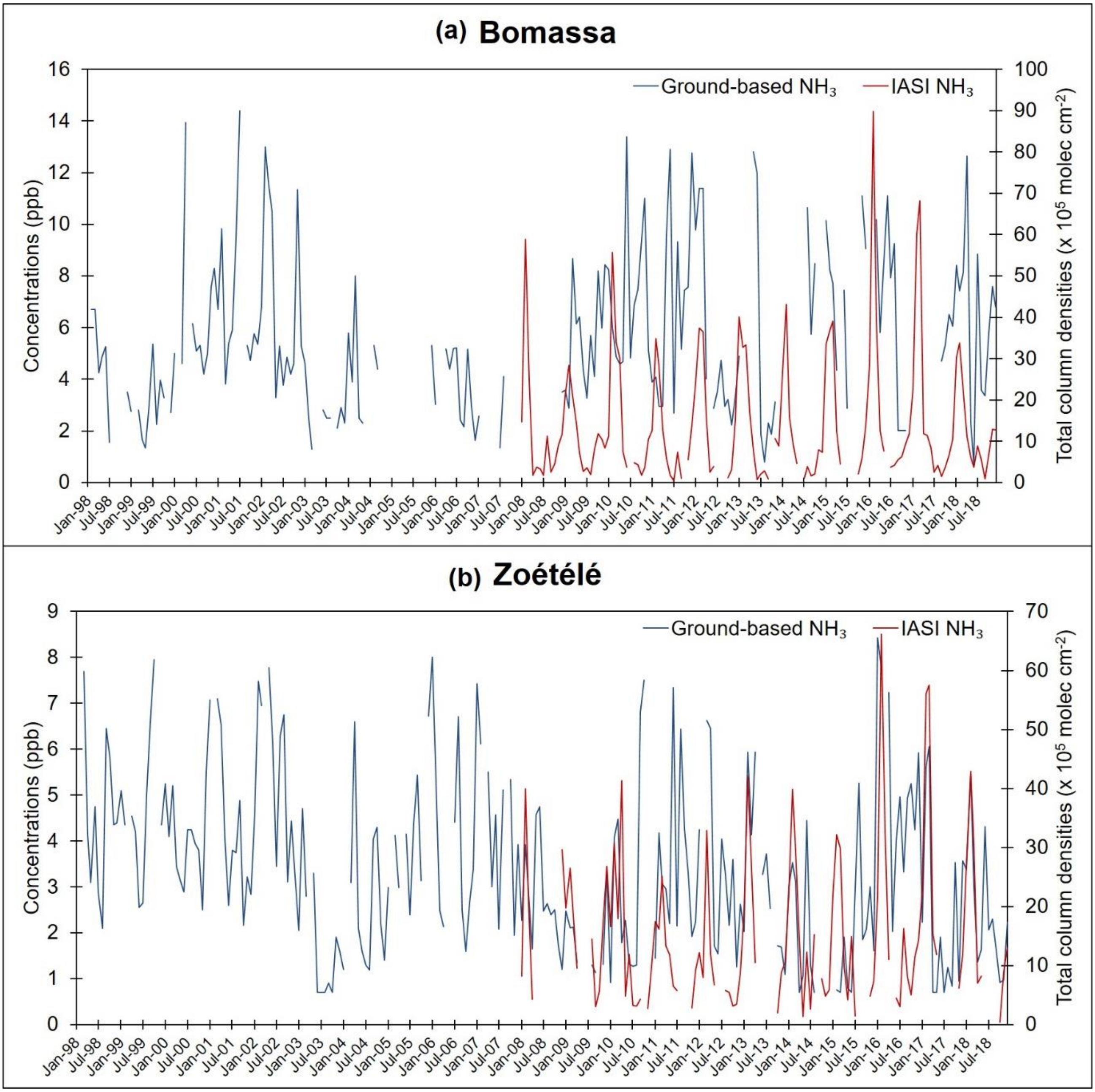

Figure 6. Monthly time-series of ground-based NH_3 concentrations over the period 1998–2018 and IASI NH_3 total column densities from 2008 to 2018 at (a) Bomassa, Republic of Congo and (b) Zoétélé, Cameroon.

Tableau 4. Minimum (Min), maximum (Max) and average (Avg) monthly, annual and seasonal ground-based NH_3 concentrations (1998–2018), and IASI NH_3 total column densities (2008–2018) at Bomassa, Republic of Congo and Zoétélé, Cameroon.

		Ground-based NH_3	IASI NH_3

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		(ppb)		*(10^{15} molec cm^{-2})*	
		Bomassa	**Zoétélé**	**Bomassa**	**Zoétélé**
Monthly	Min	0.7	0.7	0.4	0.2
	Max	14.4	8.4	89.9	66.1
Annual	Avg	5.6±1.4	3.4±0.9	14.3±2.3	15.1±2.8
Wet Season	Min	2.4	1.4	5.8	9.3
	Max	8.6	5.6	13.1	20.0
	Avg	5.6±1.5	3.3±0.9	9.4±1.6	11.2±1.8
Dry Season	Min	2.3	1.2	19.3	9.6
	Max	9.1	7.4	43.4	34.0
	Avg	5.6±1.7	3.8±1.3	27.3±3.9	22.9±5.2

We present the mean annual ground-based NH_3 concentrations and IASI NH_3 evolutions based on monthly data measured in the forested ecosystems of Bomassa (Figure 7a) and Zoétélé (Figure 7b). Ground-based NH_3 concentrations are high or low in both the dry and wet seasons, with no clear seasonality. In contrast, IASI NH_3 total column shows a well-marked seasonality, with high densities in the dry season (December to February), and low densities in the wet season (March to November) for the two sites. Mean monthly ground-based NH_3 concentrations narrowly vary from a minimum of 4.1±1.1 ppb (September) and a maximum of 7.1±3.0 ppb (March) at Bomassa (Figure 7a), from a minimum and maximum of 2.4±0.9 ppb (November) and 4.2±1.6 ppb (March) at Zoétélé, respectively (Figure 7b). NH_3 total column densities show a peak representing the annual maximum in February (46.5±14.3 x 10^{15} molec cm^{-2} for Bomassa and 36.4±12.5 x 10^{15} molec cm^{-2} for Zoétélé) and the lowest values in September (2.9±2.0 x 10^{15} molec cm^{-2} for Bomassa) and July (3.7±1.0 x 10^{15} molec cm^{-2} for Zoétélé). Mean annual coefficients of variation over the 21-year period are 16% and 19% for NH_3 concentrations, and more than 80% for IASI NH_3 over the 11-year period at Bomassa and Zoétélé, respectively.

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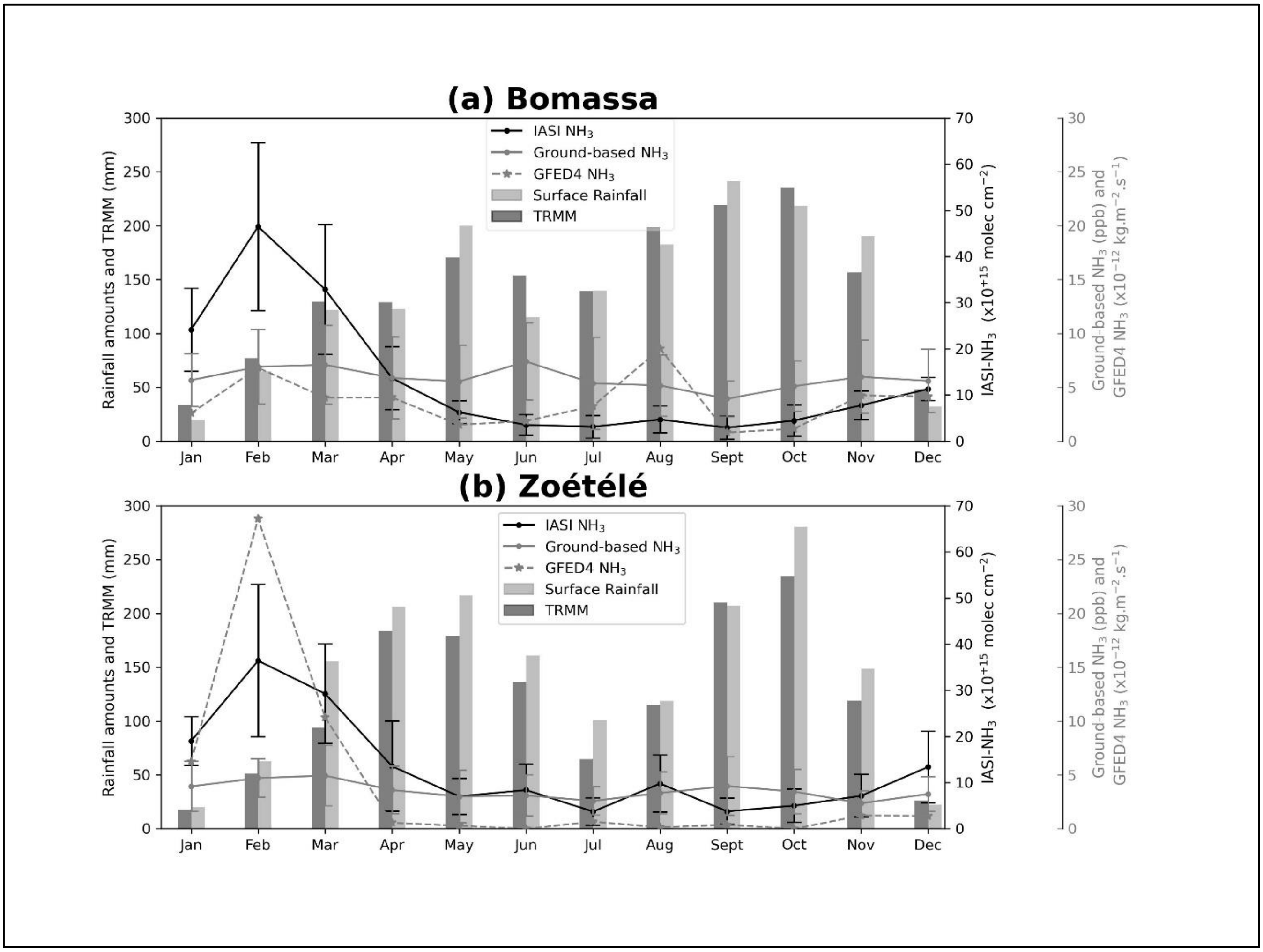

Figure 7. Mean monthly ground-based NH_3 concentrations (1998–2018), IASI NH_3 total column densities (2008–2018), rainfall amount measured by ground-based instruments (1998-2018), TRMM (2005-2018) and emissions of NH_3 from GFED4 database (1998-2018) at (a) Bomassa, Republic of Congo and (b) Zoétélé, Cameroon. Error bars represent the mean absolute deviation.

Biomass burning is recognized as a significant source of atmospheric NH_3, especially in tropical regions, but also at higher latitudes (Coheur et al., 2009; Lutsch et al., 2019; Whitburn et al., 2015a). It represents the second largest source of NH_3 after agriculture (Whitburn et al., 2017) and contributes to about 13% of total NH_3 emissions (Galloway et al., 2004) at the global scale. Other major sources of NH_3 in African wet savanna and forest ecosystems include decomposition of urea from animal excreta, fertilized soils (Bouwman et al., 2002b) and domestic fuelwood burning (Adon et al., 2010; Lobert et al., 1990). In wet savannas and forests in Africa, the NH_3 concentrations represent a combination of all natural sources with the largest contribution from biomass burning sources (Adon et al., 2010).

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Our study demonstrates that highest NH_3 concentrations are recorded during the period when fires predominate (December-February), while the lowest are obtained when rainfall is high. Indeed, during the dry season, farmers take advantage of the absence of rainfall to clear land, weed and burn agricultural residues. This slash-and-burn agriculture contributes significantly to nitrogen (NO_x and NH_3) and carbon (CO and CO_2) emissions (Tiemoko et al., 2021) into the atmosphere during the dry season. Fires related to agriculture and hunting become more important in the dry season and represent respectively 64% and 6% of the economic activities of the villagers in certain areas such as Lamto (Suzanne, 2016).

In order to show the influence of the combustion source on the atmospheric NH_3 concentrations and total column densities, we conducted linear correlation study between monthly ground-based NH_3 concentrations and IASI NH_3 total column densities on the one hand, and the GFED4 (1998-2018) emission data of NH_3 from biomass burning (Giglio et al., 2013; van der Werf et al., 2017) on the other hand. These combustion sources include agricultural waste burning, forest fires, tropical deforestation and degradation, peat fires, savanna, grassland, shrubland and temperate forest fires. The results show that there are significant Pearson correlation coefficients between monthly ground-based NH_3 concentrations and NH_3 emissions at Lamto ($r = 0.33, p < 0.001$). Monthly IASI NH_3 total columns are correlated to NH_3 emissions at Lamto ($r = 0.35, p < 0.001$). These results are consistent with the study of Whitburn et al. (2015b) carried out in four regions including "Africa north of Equator (ANE)" accounting for a major part of the total affected by fires. Indeed, they found a significant correlation ($r = 0.57$) between time series of monthly NH_3 columns retrieved from IASI measurements and MODIS fire radiative power (FRP) over the period 2008-2013 (Whitburn et al., 2015b).The most likely explanation of this correlation between NH_3 (ground-based concentrations and total columns) and emission data is that NH_3 concentrations observed in this region are therefore probably the combination of both biomass burning and soil emissions at Djougou and Lamto (Adon et al., 2010, 2013; Whitburn et al., 2015b).

For the wet savanna and forested ecosystems where NH_3 seasonality is driven by biomass burning emissions, it looks like there is still an overall pattern of increasing NH_3 in the dry season, and decreasing NH_3 in the rainy season that would be expected, which is unusual at Djougou. This modest increase in ground-based NH_3 concentrations in the wet season at Djougou could be due to the Leaf Area Index (LAI) which is much lower there than in Lamto during the wet season with annual averages of about 1.2 m^2 m^{-2} in Djougou and 3.6 m^2 m^{-2} in Lamto (Ossohou et al., 2019). Indeed, NH_3 emissions during the wet season at Djougou are therefore less intercepted by the canopy via the dry deposition process. In a general way, we assume that canopy interception/bi-directional exchange could play a role in reducing the seasonal variability at the surface (Adon et al., 2013; Delon et al., 2019), but not for the total column densities while keeping in mind that the satellite observations are for 1°x1° around each site, so they are influenced by a lot of non-local dynamics.

3.2 Trends of ground-based NH3 and IASI NH_3

We conduct the long-term trend computations by using Mann-Kendall (MK) test coupled to Sen Slope (SS) for mean annual, mean wet and dry seasons for each year of ground-based concentrations (14 and 21-year periods) and total columns densities for the 1° x 1° grid cell centered around each site (11-year period). Additional trend analyses are carried out using the Seasonal

Kendall (SK) coupled to Seasonal Kendall Slope (SKS) only for monthly data over the entire period. We adopt significance thresholds of 90% ($p<0.1$) for all trend analyses, and the percent increase or decrease is based on the mean concentrations or total column densities over each period.

In section 3.2.1, we present and discuss trends results for mean annual, wet and dry season of ground-based concentrations and IASI NH_3 total column densities in the three main ecosystems using MK test coupled to SS. The section 3.2.2 focuses on long term trends based on monthly data of NH_3 ground-based concentrations and total column densities at the six stations by using the SK test coupled to SKS. In these sections, we present only the results of significant trends. In the paragraph preceding the conclusion of the paper, we present a general comment on the trends obtained for each ecosystem and explain the differences obtained between ground-based concentration and total column density measurement trends.

Reported ground-based NH_3 concentration and IASI NH_3 trends are analyzed in the light of NH_3 emissions from all combustion sources (described in section 2.3), meteorological (air temperature and rainfall) and physical (LAI) parameters when available, which influence the atmospheric level of NH_3.

3.2.1 Annual trends

Globally, results indicate decreasing annual, wet and dry season trends in ground-based NH_3 concentrations for the three ecosystems except at Bomassa, but increasing trends in IASI NH_3 total column densities. At the annual scale, results show there is no simultaneous trend for ground-based concentrations and total column densities of NH_3 at the same site.

Results indicate significant increases in IASI NH_3 total column densities at the dry savanna of Katibougou site of +0.40 molec cm^{-2} yr^{-1} (+3.98% yr^{-1}) and at the wet savanna of Djougou site of +0.37 molec cm^{-2} yr^{-1} (+2.24 % yr^{-1}) over the 11-year period. Surprisingly, for the forested ecosystem, annual ground-based NH_3 concentrations register an increasing trend at Bomassa of +0.14 ppb yr^{-1} (+2.56 % yr^{-1}) but a decreasing trend at Zoétélé of -0.10 ppb yr^{-1} (-2.95 % yr^{-1}) over 21-year period.

We also investigate potential trends by applying the non-parametric MK test coupled to SS to the annual average of wet and dry seasons (separately) at the six stations representing the great ecosystems in Sub Saharan Africa. We observe in the wet season that NH_3 concentrations decrease at Katibougou in Malian dry savanna by -0.22 ppb yr^{-1} (-3.25% yr^{-1}), and at Zoétélé in Cameroon's forest ecosystem by -0.11 ppb yr^{-1} (-3.24 % yr^{-1}) but increase at the other forested site of Bomassa in republic of Congo by +0.13 ppb yr^{-1} (+2.29 % yr^{-1}). Ground-based NH_3 concentrations in the dry season reveal decreasing trends in both dry savanna (-0.13 ppb yr^{-1} or -2.41 % yr^{-1} for Banizoumbou and -0.12 ppb yr^{-1} or -2.26 % yr^{-1} for Katibougou) and wet savanna (-0.08 ppb yr^{-1} or -1.70 % yr^{-1} for Lamto) sites. From satellite measurements, the only significant increasing trend is obtained for IASI NH_3 total column densities at Katibougou station with slopes of +0.65 molec cm^{-2} yr^{-1} (+6.66 % yr^{-1}) and +0.26 molec cm^{-2} yr^{-1} (+2.55 % yr^{-1}) in the wet and dry seasons, respectively.

To investigate the potential causes of the observed trends of NH_3 concentrations at Zoétélé, we have applied MK trend and Pearson's correlation tests to meteorological and NH_3 emission data from GFED4 databases. The results show the decreasing trend in ground-based NH_3 concentrations in the wet season at Zoétélé could be attributed to wet season-to-wet season

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increasing of the LAI (+0.69% yr^{-1}), with a 99% significant anticorrelation of -0.57 between these two variables. We do not yet know the cause of the increase in LAI from one wet season to the next in the Zoétélé forest ecosystem. However, more vegetation results in greater dry deposition rate, which would significantly reduce the observed wet season to wet season ground-based atmospheric NH_3 concentrations at Zoétélé. During the wet season, air humidity and soil moisture increase, leading to large NH_3 deposition on vegetation during wet months (Delon et al., 2019).

3.2.2 Trends accounting for seasonality

Long time series of atmospheric NH_3 could usually be affected by seasonality, which is the cyclical changes in concentrations or densities over the course of the year. The SK test is significantly robust in revealing trends in seasonal time series. In this section, we perform trend computations using SK coupled to SKS of monthly mean ground-based NH_3 concentrations and IASI NH_3 total column densities of the entire dataset. Results for only significant monthly trends ($p<0.1$) from all INDAAF sites are shown in Figure 8. In general, the statistical tests reveal significant decreasing trends for ground-based NH_3 concentrations (except at Bomassa), but increasing trends for IASI NH_3 total column densities (Figure 8).

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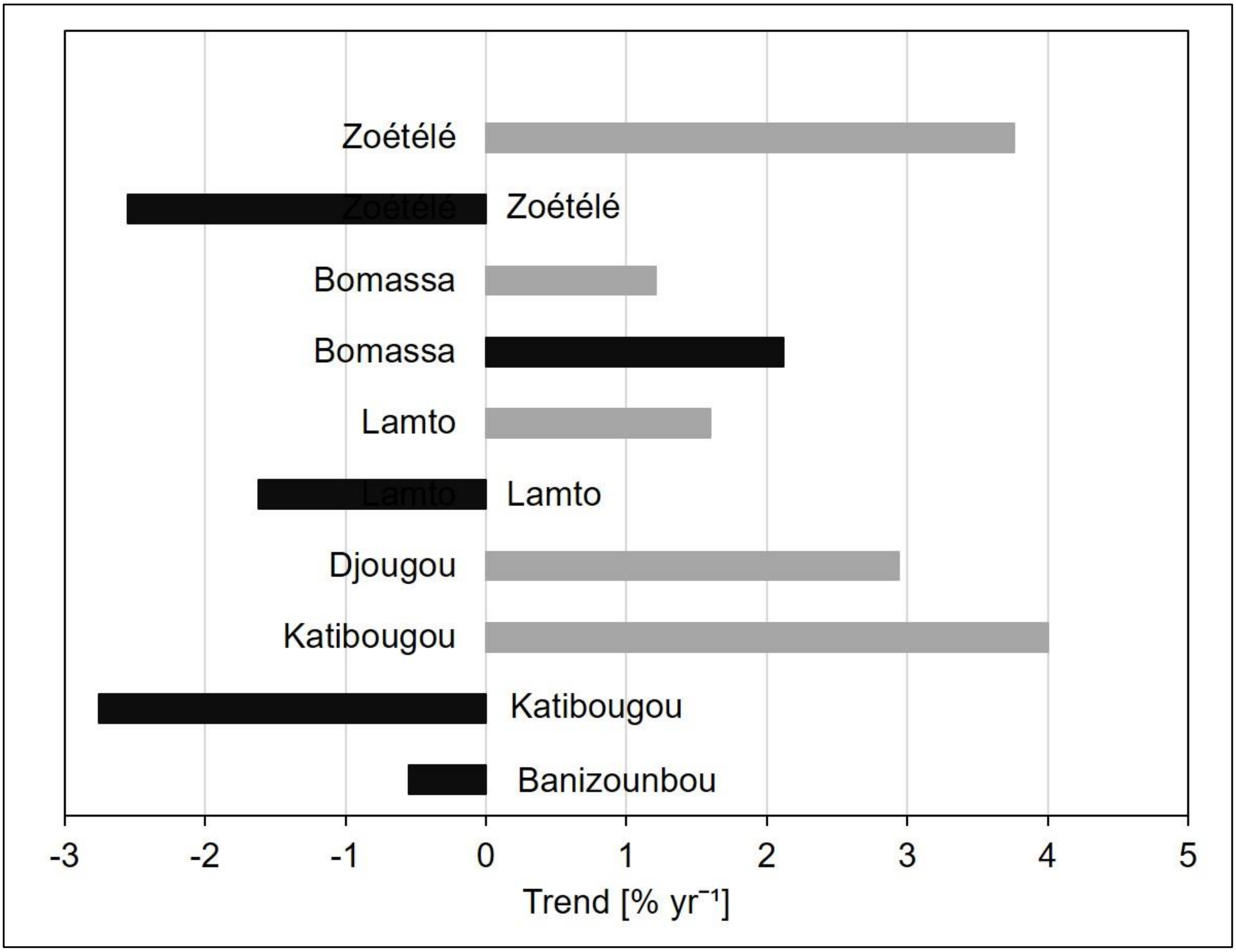

Figure 8. Estimated percentage changes in ground-based NH3 concentrations (in black) and IASI NH_3 total column densities (in grey) for INDAAF network stations where monthly trends are significant at 90%. These trends are obtained using the monthly data of the overall database.

Ground-based NH_3 concentrations decrease in the dry savannas of Banizoumbou by -0.03 ppb yr^{-1} (-0.55 % yr^{-1}) and Katibougou by -0.16 ppb yr^{-1} (-2.76 % yr^{-1}), but IASI NH_3 total column densities increase at Katibougou by +0.40x10^{15} molec cm^{-2} yr^{-1} (+4.00 % yr^{-1}). A significant decreasing trend is also found for ground-based NH_3 concentrations in the wet savanna of Lamto (-0.06 ppb yr^{-1} or -1.62 % yr^{-1}), but significant increasing trends are obtained for IASI NH_3 total column densities both at Djougou (+0.49x10^{15} molec cm^{-2} yr^{-1} or +2.94 % yr^{-1}) and Lamto (+0.34x10^{15} molec cm^{-2} yr^{-1} or +1.60 % yr^{-1}) sites. SK test applied to monthly ground-based NH_3 concentrations in the forested ecosystem sites shows significant increasing trend by +0.12 ppb yr^{-1} (+2.12 % yr^{-1}) at Bomassa, but decreasing trend by -0.09 ppb yr^{-1} (-2.55 % yr^{-1}) at Zoétélé. The increasing

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trend of IASI NH_3 total column densities at Bomassa (+0.17 molec cm^{-2} yr^{-1} or +1.21 % yr^{-1}) is three times lower than that of Zoétélé (+0.56 molec cm^{-2} yr^{-1} or +3.76 % yr^{-1}).

The SK applied to monthly data from January, 1998 to December, 2018 shows that relative humidity decreases by -0.15% yr^{-1} at Lamto. We calculate Pearson's correlation between ground-based NH_3 concentrations and relative humidity and we find a coefficient of -0.50 significant at 99%. This statistical test demonstrates that the decreasing monthly trend of ground-based NH_3 concentrations cannot be explained by the monthly relative humidity trend.

Trend studies of NH_3 concentrations and densities obtained respectively with the INDAAF passive samplers and the IASI instrument have shown significant trends depending on each biome. Overall, we obtained decreasing trends for ground-based measurements (except at the Bomassa forest site), but increasing trends for IASI total column densities of NH_3. This result was found for all ecosystems. The long-term statistical trend results for NH_3 emissions from GFED4 database are not significant, so could not explain the trends obtained for the ground-based and satellite data. A plausible explanation for the contrasting trends between surface concentrations and satellite columns could be the impact of biomass burning plumes. The latter are likely less well captured from INDAAF passive samplers, while they are very well measured by IASI (Zheng et al., 2021). It is likely that IR sounders have a higher sensitivity to fire plumes, which are located higher in the atmosphere (and so the ground-based measurements will show less sensitivity to them).

4 Conclusion

Using a 21-year period of INDAAF passive samplers and an 11-year period of IASI product, we have characterized coevolutions and trends of atmospheric NH_3 at six stations of the INDAAF network in the African dry savanna (Banizoumbou, Niger and Katibougou, Mali), wet savanna (Djougou, Benin and Lamto, Côte d'Ivoire) and forest (Bomassa, Republic of Congo and Zoétélé, Cameroon). The results showed that ground-based concentrations of NH_3 and IASI NH_3 total column densities are more important in the dry savanna and wet savanna ecosystems, respectively. Indeed, mean annual ground-based concentrations of NH_3 over periods 1998/2005-2018 period are 5.7-5.8 ppb in dry savanna, 3.5-4.7 ppb in wet savanna and 3.4-5.6 ppb in forest ecosystems. The overall mean annual IASI NH_3 total column densities for a 1° x 1° grid cell centered on each site over 2008-2018 are 10.1-11.0 x 10^{15} molec cm^{-2} in the dry savanna, 16.5-21.4 x 10^{15} molec cm^{-2} in the wet savanna and 14.3-15.1 x10^{15} molec cm^{-2} in the forest ecosystems. If we consider only ground-based measurements, the results show that NH_3 emissions from Sahelian soils and livestock in the dry savanna ecosystem are higher than those from biomass burning in the wet savanna and forest ecosystems.

We have recorded 95% significant Pearson correlation between monthly ground-based concentrations and IASI total column densities of NH_3 at Banizoumbou (*r=0.3)*, Lamto (*r=0.59*), Bomassa (*r=0.23*) and Zoétélé (*r=0.36*), showing that NH_3 abundancies at the wet savanna of Lamto show the best agreement between ground-based and satellite remote sensing. In the dry savanna sites of Banizoumbou and Katibougou, the seasonal ground-based concentrations of NH_3 are highest both at the

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beginning and the end of the wet season. Conversely, ground-based concentrations of NH_3 are highest in the dry season at the wet savanna sites of Djougou and Lamto, but no marked seasonality between wet and dry season was observed for ground-based NH_3 concentrations in the forest sites of Bomassa and Zoétélé. IASI NH_3 total column densities follow the same seasonality as ground-based NH_3 concentrations in the dry and wet savannas, while the seasonality is more marked in the forested ecosystem.

The non-parametric Mann-Kendall statistical trend test shows 90% significant mean annual increasing trend for IASI NH_3 total column densities which is the most important in the dry savanna of Katibougou (+3.98 % yr^{-1}). Ground-based NH_3 concentrations in the forested ecosystem increase at Bomassa (+2.56 % yr^{-1}), but decrease at Zoétélé (-2.95 % yr^{-1}). In both dry and wet seasons, ground-based NH_3 concentrations decrease from -3.25% yr^{-1} (Katibougou) to -1.70% yr^{-1} (Lamto), but increase in wet season at Bomassa (+2.29% yr^{-1}). IASI NH_3 total column densities increase in the wet season (+6.66% yr^{-1}) and dry season (+2.55% yr^{-1}) only at Katibougou, Mali. The seasonal Kendall test applied to monthly data over the entire periods also shows decreasing trends at all the sites, except at Bomassa (+2.12% yr^{-1}) for ground-based NH_3 concentrations. In contrast to trends calculated using ground-based observations, monthly IASI NH_3 total column densities increase for all ecosystems, ranging from +1.21% yr^{-1} (Bomassa) to +4.00% yr^{-1} (Katibougou). The increasing trends observed in dry seasons of wet savanna and forest African ecosystems could be attributed to a longer residence time of NH_3 from biomass burning and agricultural waste burning sources in the atmosphere which are the main sources of atmospheric NH_3 in this season. Decreasing trend in ground-based NH_3 concentrations in the wet season at Zoétélé could be related to wet season-to-wet season increasing of the LAI (+0.69% yr^{-1}), with a 99% significant anticorrelation of -0.57 between these two variables.

Results reported in this paper represent the unique long-term regional characterization of ground-based NH_3 concentrations in Africa. Our study allows a better understanding of the main drivers of atmospheric NH_3 level of concentrations. We conclude that the main atmospheric NH_3 sources are alkaline Sahelian soils and agro-pastoralism emissions along the dry savanna ecosystem. NH_3 variability in the wet savanna and forest ecosystems emphasized the importance of two main sources, i.e., biomass burning and agricultural waste burning.

Data availability

The INDAAF NH_3 observations are available at https://indaaf.obs-mip.fr/ upon registration. TRMM 3B42 precipitation data are available from https://pmm.nasa.gov/data-access/downloads/trmm. The IASI NH_3 data are available from The IASI https://iasi.aeris-data.fr.

Author contribution

Money Ossohou designed the study, conducted the statistical analysis, and wrote the paper.
Jonathan Hickman, and Corinne Galy-Lacaux contributed to study design and edited the paper.

Lieven Clarisse, Pierre-François Coheur, and Martin Van Damme developed the original IASI trace gas retrievals and edited the paper.
Marcellin Adon, and Véronique Yoboué edited the paper.
Eric Gardrat, and Maria Dias Alvès analysed the samples.

Competing interests

The authors declare that they have no conflict of interest.

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