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ENVIRONMENTAL RESEARCH
LETTERS

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To cite this article before publication: Adama Bakayoko *et al* 2021 *Environ. Res. Lett.* in press https://doi.org/10.1088/1748-9326/abe25c

Dominant contribution of nitrogen compounds in precipitation chemistry in the Lake Victoria catchment (East Africa)

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Abstract

This work provides a complete chemical characterization of rains collected in the tropical rural site of Mbita (Kenya) on the shores of Lake Victoria (annual rainfall 1259.3 mm). We present a wet nitrogen deposition budget including inorganic and organic dissolved nitrogen in relation with atmospheric sources of gases and particles, precipitation rate and air mass transport. A unique two-year monitoring data set (2017-2019), providing 183 rain samples was collected and analyzed according to international standards (WMO/GAW). Considering that precipitation represents the largest contributor of water to the Lake Victoria (80%), this study gives new insights in the seasonality of nutrients wet deposition inputs in the unique natural resource represented by Lake Victoria and its catchment.

Four main contributions to the chemical composition of precipitation, were identified: (1) a 28% terrigenous contribution related to crustal and biomass sources (2) a 14% marine contribution related to Indian ocean air masses intrusion, (3) a 15% organic contribution due to volatile organic carbon emissions from biomass burning and vegetation and (4) a predominant nitrogenous contribution of 39% due to livestock and fertilizers, biomass burning and neighboring agricultural fires. Ammonium and nitrate volume weighed mean concentrations are 36.75 and 8.88 μeq L^{-1}, respectively. Rain in Mbita is alkaline (pH=5.8) highlighting neutralization by heterogeneous chemistry. Total nitrogen wet deposition is 8.54 kgN ha^{-1} yr^{-1}, 58,760 tN yr^{-1} for the entire lake, with 26% attributed to dissolved organic nitrogen. A total atmospheric deposition of 15 kgN ha^{-1} yr^{-1} is estimated taking into account dry deposition estimate from literature, showing that the Lake Victoria ecosystem is exposed to eutrophication. An extensive and regular monitoring of wet and dry nitrogen deposition is highly

recommended both in-shore and off-shore to help improving the efficiency of nitrogen use in agricultural areas and reduce nitrogen losses around Lake Victoria.

Keywords: Precipitation chemistry, wet deposition, nitrogen wet deposition budget, Lake Victoria basin, Kenya.

1. Introduction

Determining atmospheric budgets of key chemical compounds is crucial to understand the functioning of ecosystems and biogeochemical cycles. In these budgets, the importance of the chemical composition of wet deposition as a source of nutrients, e.g, nitrogen (N), sulphur (S) or carbon (C) is widely recognized (Duce *et al* 2009), and shows a high spatial and temporal variation. This composition reflects various interacting physical and chemical mechanisms in the atmosphere including biogenic and anthropogenic sources of pollutants, atmospheric transport and chemical transformation processes as well as removal processes (Galy-Lacaux *et al* 2009). The comprehensive global assessment on precipitation chemistry and biogeochemically important trace species deposition by Vet *et al* (2014) emphasized that the African continent is under-sampled and lacks quality controlled measurements.

80% of African countries are affected by soil nitrogen deficiency because of inadequate use of fertilizers, and insufficient quantity and poor quality of organic inputs (Masso *et al* 2017). In Africa it is expected that nitrogen (N) deposition to ecosystems will increase by 50% until 2100 (Lamarque *et al* 2013), especially through wet deposition, due to increasing demography and changes in climate, land uses and atmospheric concentrations. The impact of nitrogen management and nitrogen deposition in Africa has become a major societal challenge related to food security, global change and biodiversity loss (Zhang *et al* 2020). Sub-Saharan Africa, considered as a "too little" N area, would benefit from a reduction of reactive N pollution to help limited available N sources to go further in supporting food production (Sutton *et al* 2019). The conversion of atmospheric N compounds back to N atmospheric deposition is an important determinant of N availability for ecosystems, but currently, experimental studies conducted on the African continent remain scarce and scattered.

Lake Victoria is the second largest freshwater lake in the world with a lake surface area of 68,800 km^2 and a total basin area of 195,000 km^2. Precipitation is the largest contributor of water (80%) to the lake (Sutcliffe and Petersen 2007, Tamatamah *et al* 2005), leading some authors to describe the lake

as "atmosphere controlled" (Kizza *et al* 2009, Tate *et al* 2004). The Lake is a great socio-economic resource of the East African Community partner states for fisheries, tourism, transport, water, and energy among others, and a core to East Africa regional integration and development (Nyilitya *et al* 2020). Anthropogenic inputs of N to Lake Victoria through runoff or waste management were described in several studies (Bootsma and Hecky 1993, Kayombo and Jorgensen 2006, Masso *et al* 2017, Nyilitya *et al* 2020, Zhou *et al* 2014), where the authors also point out that N wet deposition is missing in the budget estimation, due to the lack of available measurements. Indeed, very few studies on precipitation chemistry and wet deposition of nutrients in Great Lakes exist (Lakes Victoria, Malawi, Tanganyika, Kivu) and give an incomplete view of the chemical content of rain (Bootsma *et al* 1996, Gao *et al* 2018, Vuai *et al* 2013), highlighting the necessity to accurately quantify this input. Assessing wet deposition fluxes and their seasonal variability to Lake Victoria has become important for understanding the bioavailability of nutrients in this ecosystem. Furthermore, assessing the organic part of nitrogen wet deposition with accuracy is a major challenge, while many authors report very few existing data in Africa on this topic (Cape *et al* 2011, Cornell 2011).

This study is part of the East African demonstration area of the Global Environmental Facility – United Nations Environmental Program (GEF-UNEP) International Nitrogen Management System (INMS) project, and of the French program Cycle de l'Azote entre la Surface et l'Atmosphere en afriQUE (CASAQUE). This work is based on a two year monitoring period (May 2017 – April 2019) of wet deposition on the shore of the Lake Victoria to assess the chemical composition of rain in the area, as well as the sources of chemical compound emissions influencing the precipitation chemistry content. We include a particular focus on the N contribution (inorganic and organic) in rain. The climatic and hydrological projections to the lake Victoria basin show an intensification of future annual rainfall by 25% in the eastern and 5–10% in the western part of the basin (Olaka *et al* 2019). In this context, this study will provide (1) a unique quantification of important biogeochemically species wet deposition and (2) a comprehensive atmospheric wet deposition nitrogen budget to the Lake Victoria and its catchment.

2. Sampling site and methods

2.1. Site description

The study site is located at the Centre of Insect Physiology and Ecology (ICIPE) in Mbita (Fig. 1). Mbita is a tropical agricultural area along the shores of Lake Victoria in East Africa. It is located near the Southwestern border of Kenya and Uganda at the latitude and longitude of 0.44˚S, 34.18˚E

respectively and 1125 m above mean sea level - MSL (Fig. 1a). Mbita is located at 71 km as the crow flies from the third largest city in Kenya, Kisumu City with a population of 409,928 inhabitants (KNBS 2010**)**. The Homa Bay county is divided into eight sub counties including Mbita as one sub county. The total population of the Homa Bay county is 1 131 950 inhabitants with a low density of 307 inhabitants per km2 (ADP 2019). Mbita sub county, with 125 000 inhabitants, represents the second lowest contribution of the Homa Bay total population in 2019. Two main relief regions are found, the lakeshore lowlands and the upland plateau. The upland plateau starts at 1219 m above sea level and has an undulating surface resulting from erosion (Fig. 1b). The Kondera natural forest borders Mbita. Mbita is part of the lower midland and its surrounding areas are influenced by biomass burning, mixed and anthropogenic sources, and dust aerosols (Boiyo *et al* 2018b, 2017b, Makokha and Angeyo 2013). In addition to local sources, the long-range transport of aerosols towards the site could significantly influence the aerosol load (Boiyo *et al* 2017a, Gatebe *et al* 2001). Agriculture plays a crucial role to food and nutrition security in Homa bay county. Most of the income of Homa bay county is derived from crop, livestock and fisheries activities but 50% of the population is food insecure (GoK 2013). Mbita is charasteristic of a mixed crop site with maize, sorghum, beans, millet and agroforestry systems (GoK 2014). Large scale farms are found in Mbita sub county where large stocks of livestock are kept (zebu cattle, red massai sheep, goat and chicken) (MoALF 2016). In Mbita and on the borders of Lake Victoria, the density of cattle and small ruminants represents more than 100 cattle per square kilometer (FAO 2005).

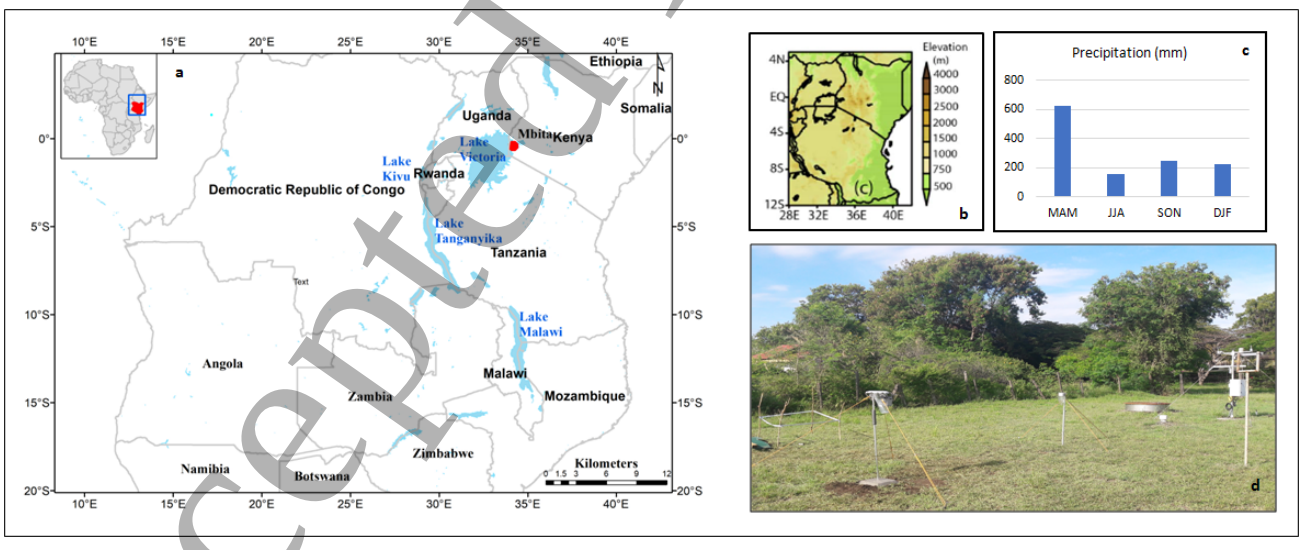

Figure 1: (a) Map of the study area on the African Continent (shown top left in the insert). Location of the site of Mbita (red circle). Blue labeling represents the major African lakes and ocean. (b) Topographic map within the study domain. (c) Mean precipitation at Mbita according to seasonal climatology for the period 1985-2019 with March

April May (MAM), June July August (JJA), September October November (SON) and December January February (DJF). (d) Photo of the Mbita measurement site located at the ICIPE center.

2.2 Climatology

A long term database of key meteorological parameters is available at ICIPE center for the period 1985 - 2019. It includes ambiant air temperature for the period 1985-2013, air relative humidity for the period 1999-2013 and pluviometry for the period 1985-2019, with a gap in 1997 and 1998 (Fig. 2a) (for more details, see supplementary material (Sup. Mat.) section meteorological parameters). Mbita is characterized by a low annual variability of ambient air temperature for the period 1985-2013, with a mean annual value of 24.33 ± 0.80 °C (Fig. 2a) and a mean annual relative humidity of 80.74 ± 3.96%. Monthly means of relative humidity do not present any distinct seasonal cycle and vary from 76.46 ±13.30 % during the dry season in August to 83.09 ±4.83 % in the wet season in May (Fig. 2a). Wind speed data show a relatively low mean annual value with 2.03 ± 0.14 $m.s^{-1}$. Monthly mean calculations show higher values in the main wet season with a maximum in May (2.30 ± 0.23 $m\ s^{-1}$) and a minimum in the dry season in February (1.83 ± 0.09 $m\ s^{-1}$). These results are similar to the ones presented in other studies in East Africa (Boiyo *et al* 2017b, Ogwang *et al* 2015, Yang *et al* 2015).

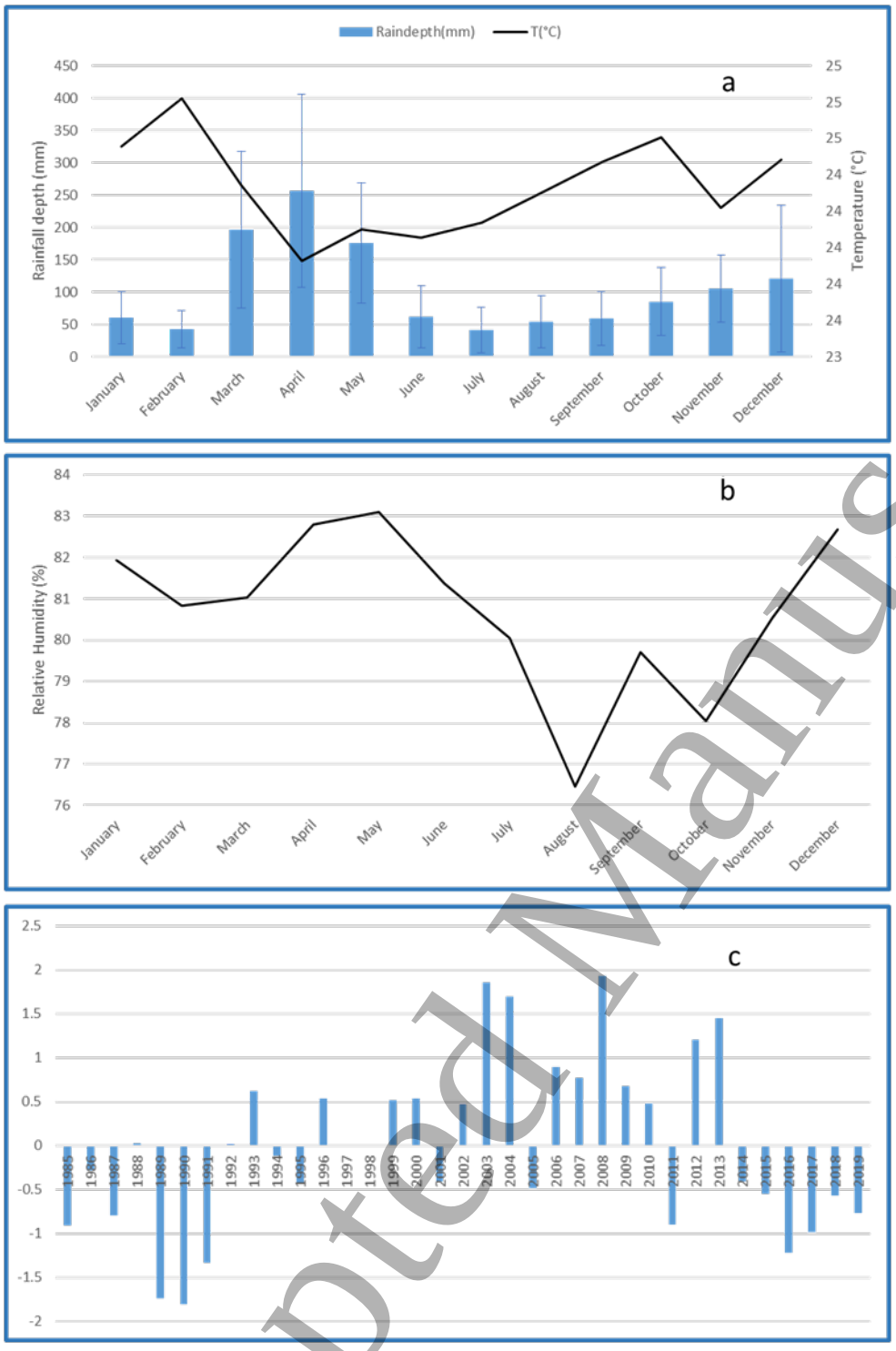

Figure 2: Monthly mean meteorological parameters measured at Mbita from 1985 to 2013. (a) Rain depth (mm) from 1985 to 2019 and ambient air temperature (°C) from 1985 to 2013, (b) air relative humidity (%) from 1999 to 2013; (c) Standardized Precipitation Index (SPI) over the period 1985-2019.

The mean annual pluviometry at Mbita is 1259.3 ± 334.6 mm for the period 1985-2019. The rainfall patterns in the study area present a bimodal distribution with a long and a short rainy season from

March to May and October to December respectively. Standard deviations indicate that precipitation can be highly variable. The atmospheric circulation caused by the presence of Lake Victoria (breeze effects) as well as the convergence zone created by the steep topography of the site (highlands) influences the spatial and temporal variability of the precipitation regime (Okech *et al* 2018). Mbita receives generally a maximum of rain in April (256.3 ±148.6 mm) and December (120.4 ±51.6 mm), and a minimum in July (41.6 ±35.3 mm) and February (42.2 ±29.0 mm). Three main periods of rainfall variability are highlighted from the Standardized Precipitation Index (SPI) analysis (see definition in the Sup. Mat.) : (I) a period in deficit from 1985 to 1992, (II) a period in excess from 1999 to 2013 and (III) a period in deficit from 2015 to 2019. The year 2008 presents a maximum SPI close to 2 and corresponds to the rainiest year (1906 mm) (Fig.2b). The driest year with an index of -1.7 is observed in 1990 with 655.7 mm. Details may be found in the supplementary material.

2.3. Sample collection and analytical procedures

Rainwater samples at Mbita were collected using an automatic precipitation collector designed for the INDAAF (International Network to study Deposition and Atmospheric composition in AFrica) network (see details Sup. Mat. section sample collection protocol). At the site, a local operator collected each rain event in 50 mL greiner tube immediately stored at ICIPE center in a deep freezer (-18°C). During the day, samples were collected just after each rainfall and when rain occurred at night, sampling wass done between 6 and 8 AM. After collection, samples were sent back for analysis to the Laboratoire d'Aérologie (Laero, Toulouse, France) in the strict respect of the cold chain.

From May 2017 to April 2019, the total rainfall amount was 1942 mm and the collected rain samples represent a total of 1814 mm with 189 samples. Table 1 presents the annual total precipitation (Pt) in mm, the percent total precipitation (%TP) and the interannual variability as a percentage relative to the mean annual rainfall for the 1985-2019 period. As defined by (WMO 2004), %TP is the ratio between the annual precipitation (Pt) and the collected precipitation (Pc). The annual and quarterly Percent Coverage Length (%PCL) is also indicated in Table 1.

Table 1: Rainwater collection at Mbita (2017-2019): Annual Total Precipitation (Pt, mm), Interannual variability (%), Collected precipitation (Pc, mm) and Number of collected rain event (Nc), Percent total precipitation (%TP), Annual percent coverage length (%PCL) and in brackets: %PCL for each quarter (0 and 1 means 0% and 100% respectively).

Year	2017	2018	2019
Pt (mm	930.95	1070.7	1004
Inter annual Variability (%)	-26%	-15%	-20%
Pc (mm) and Nc	592.25 (70)	1004.2 (94)	217.5 (25)
% TP (%)	64	94	22*
% PCL Annually % (quarterly)	75 (0111)	100 (1111)	25 (1000)

* %TP only concerns four months of collection (January-April)

According to the quarterly %PCL, data from April to December in 2017 and from January to April in 2019 will only be used to calculate monthly averages of Volume Weighed Mean (VWM) and Wet Deposition (WD). Annual VWM and WD for Mbita will be computed only for year 2018 where the %TP is 94% and the PCL 100% (Table 1). In reference to the WMO international standards, we assume the precipitation collection at Mbita in 2018 to be representative of the studied period according to the parameters calculated in Table 1.

2.4. Analytical methods and quality procedure

The chemical analyses of the samples were performed at the Chemical Laboratory of the Laero, in Toulouse for the major inorganic (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-}, NO_2^-) and organic ions ($HCOO^-$, CH_3COO^-, $C_2H_5COO^-$, $C_2O_4^{2-}$) using Ionic Chromatography (IC) as described in Galy-Lacaux and Modi (1998) and at the Laboratoire d'Ecologie fonctionnelle et environnement (Toulouse, France) for TDN (For detailed procedures, see Sup. Mat.).

2.5 Satellite data

Observations of atmospheric NH_3 (ANNI-NNv3) from the Atmospheric Infrared Sounding Interferometer (IASI) on board the European Space Agency's Metop-A were analyzed, providing near-daily global coverage. As for NO_2, NASA's standard IMO NO_2 v3 tropospheric column density product, on board NASA's Aura satellite, was used. (For more details, see Sup. Mat.).

2.6. Back trajectories

Back trajectories initiated at Mbita (0.44˚S, 34.18˚E, 1125 m MSL equivalent to 890 hPa pressure level) were used to identify the origin of air masses, for three initial pressure levels at 850 hPa (about 1550 m altitude), 800 hPa (2065 m), and 750 hPa (2610 m), using ERA-5 climate reanalysis from ECMWF (Hersbach *et al* 2020).

3. Results and discussion

3.1 Chemical composition of precipitation and wet deposition fluxes

The most important ions in Mbita rainwater samples in 2018 are NH_4^+ and Ca^{2+} (Table 2) representing about 52% of total cations VWM concentrations. NO_3^- and $HCOO^-$ are the most abundant anions representing 38% of total anions VWM concentrations. Concentration of cations follow a general pattern $NH_4^+ > Ca^{2+} > Na^+ > K^+ > Mg^{2+}$, while concentrations of anions follow a general pattern $NO_3^- > HCOO^- > Cl^- > SO_4^{2-} > CH_3COO^- > HCO_3^- > C_2O_4^{2-} > C_2H_5COO^- > NO_2^-$.

Table 2: Annual Volume Weighted Mean (VWM) (μeq L^{-1}) and Wet Deposition (WD) (kg ha^{-1} yr^{-1}) in 2018.

Species	VWM	WD
H^+	4.21	0.05
Na^+	6.62	1.63
NH_4^+	36.75	5,51*
K^+	5.57	2.33
Ca^{2+}	15.22	3.30
Mg^{2+}	3.01	0.40
NO_3^-	8.88	1.33*
Cl^-	7.02	2.66
SO_4^{2-}	6.94	1.20*
NO_2^-	0.07	0,01*
$HCOO^-$	8.79 (8.51)	(4.10)
CH_3COO^-	6.98 (5.71)	(3.61)
$C_2H_5COO^-$	0.32 (0.25)	(0.19)
$C_2O_4^{2-}$	1.85 (1.75)	(0.82)
HCO_3^-	5.13	3.35
pH	5.8 ± 0,6 [b]	
Pt (mm)	1070.7[c]	

VWM in brackets are dissolved part of organic acids and associated WD

* WD is calculated in kgN or S ha^{-1} yr^{-1}.

[b] Mean pH and standard deviation

[c] 2018 annual precipitation

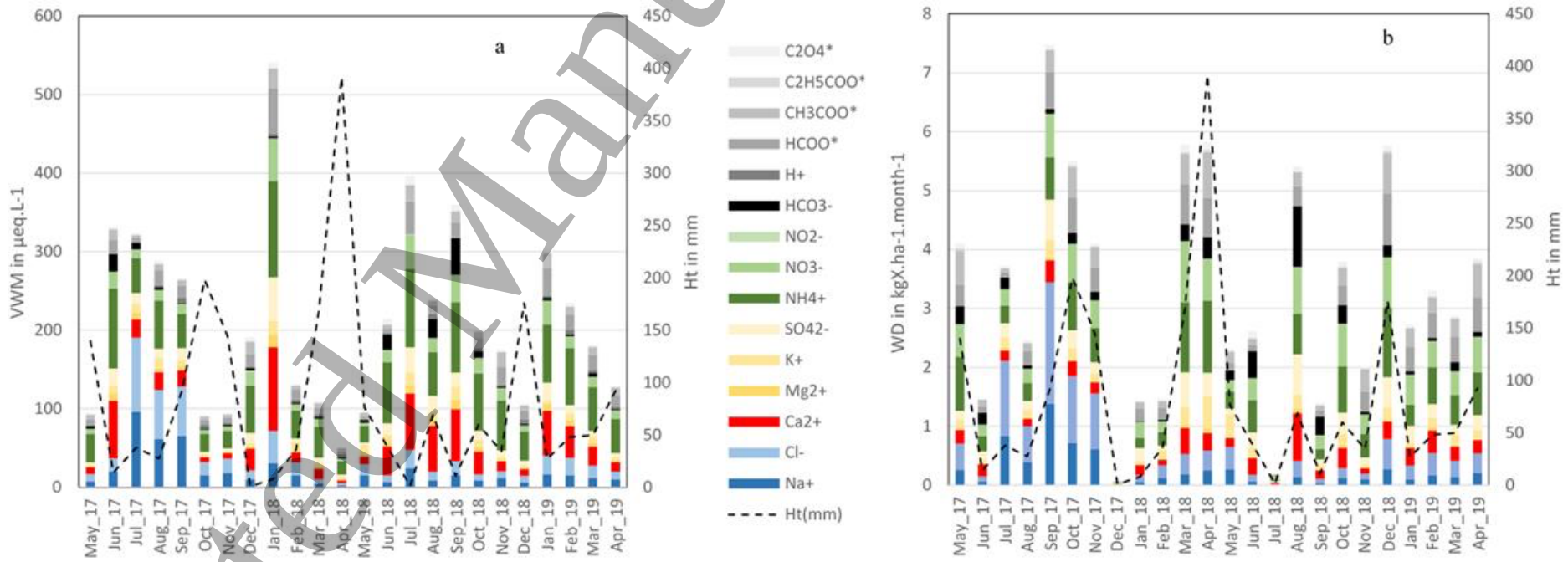

Figure 3: (a) Monthly Volume Weighed Mean (VWM) concentrations measured in precipitation at Mbita and (b) monthly Wet Deposition fluxes in kg ha^{-1} $month^{-1}$ for each species from May 2017 to April 2019. Ht stands for rain depth.

Marine and terrigenous contribution

The Cl^-/ Na^+ ratio at Mbita (1.06) is close to the seawater ratio (1.16). The high correlation between Na^+ and Cl^- (r=0.99) associated to the Enrichment Factor (EF, defined in Sup. Mat.) of Cl^- (<1) indicates that both compounds have a marine origin (Cao *et al* 2009). Lower correlations are found between Na^+ and K^+, Mg^{2+} and SO_4^{2-} (r = 0.40, 0.29 and 0.33 respectively), suggesting a non-marine additional origin for these species. Sea Salt Fractions (SSF, for Ca^{2+}, Mg^{2+}, K^+ and SO_4^{2-} represent 2%, 50%, 3% and 12% respectively.

Monthly variations of ionic VWM concentrations indicate the predominance of NH_4^+ for all months (Fig. 3a), except in July, August and September (JAS) 2017 where the marine ions Na^+ and Cl^- are the most abundant. Indeed, during the rain event of 15 July, very high contents of Na^+ (285.6 µeq L^{-1}) and of Cl^- (293.8 µeq L^{-1}) were measured. The analysis of back trajectories arriving in Mbita on 15 July 2017 1900 UTC (Fig. 4a, 4b) from 850 to 750 hPa pressure levels confirms that the air is of oceanic origin owing to a southeasterly flow which crosses the central Kenyan highlands and the Rift valley before arriving over the Lake Victoria plateau. The duration of the trip from the Indian Ocean to Mbita is about 3.5 days for the air observed at 800 and 750 hPa at Mbita. The same course took 5.5 days for the 850 hPa level with strong ascent from 1500 to 4500 m altitudes on 11 July 2017 probably in relation with cloud formation on the high terrain slopes, leading to rainfall at Mbita strongly loaded in Na^+ and Cl^-. From July to September 2017, 7 rain events of this type were registered leading to maximum monthly VWM concentrations from 65 to 96 µeq L^{-1} (Fig. 3a), clearly originating from the Indian Ocean (figures not shown) whereas the majority of rain events over the period 2017-2019 show concentrations ten times lower (1 to 50 µeq L^{-1}). The analysis of monthly Aerosol Optical Depth (AOD) measurements by Boiyo *et al* (2019, 2018b) obtained at Mbita ICIPE from 2007 to 2015 confirms this marine influence with a peak in June, July, August (JJA) associated to marine aerosols. The authors indicate that 26% of the aerosols in Mbita originate from marine sources.

The Ca^{2+} contribution is the second most important after NH_4^+ (Table2). Ca^{2+} is significantly correlated to K^+, Mg^{2+} and SO_4^{2-} (r = 0.80, r = 0.97, r = 0.88) emphasizing the contribution of potential crustal (terrigenous) and biomass burning sources. Non-Sea Salt Fraction (nSSF) contributions for Ca^{2+}, Mg^{2+}, K^+, SO_4^{2-} are 98%, 50%, 97%, 88% respectively. Monthly VWM evolution of Ca^{2+}, Mg^{2+}, K+, SO_4^{2-} concentrations over the period 2017-2019 exhibit two peaks in JJA and in January (Fig. 3a). The analysis of back trajectories arriving at Mbita on 5 January 2018 0700 UTC (Fig. 4c, 4d) from 850 to 750 hPa pressure levels confirms that the air is of continental origin owing to a northerly flow

which crosses South Sudan and Uganda before arriving over the Lake Victoria plateau. During the southern hemisphere summer, high temperatures at surface and in the low atmosphere south of the Equator lead to a cross-equatorial flow which brings relatively dry and dusty air to Lake Victoria. The airflow arriving at Mbita at 800 and 750 hPa is relatively strong as it took about 24 h to cover 500 km at 5.8 m s^{-1} (Fig. 4d). The origin of air at 850 hPa is much closer as it stays very close to the surface level. Ca^{2+} is usually used as a reference element for continental crust and considered as a typical lithospheric element (Ding *et al* 2013). The positive relationship between K^{+}, Mg^{2+}, SO_4^{2-} and Ca^{2+} confirms the importance of the particles coming from arid and semiarid regions in the chemical composition of rainfall. The North African desert areas (Sahel and Sahara) as well as the Arabian Peninsula are probably the most important mineral aerosol sources (Jish Prakash *et al* 2015, Kaufman 2005). Boiyo *et al* (2019, 2018b, 2018a) show that long-range transport of aerosols from Saharan and Arabian desert enhances dust atmospheric loading during the local dry season in Mbita. Due to the partial dissolution of soil dust terrigeneous components, rain in the tropical rural site of Mbita is loaded with dissolved calcium and carbonates (calcite). In addition to calcite, dust contains dolomite and gypsum, which may explain the enrichment of Mg^{2+}, SO_4^{2-} and K^{+} (Avila *et al* 1997). Terrigenous contributions were also found in other African ecosystems and regions of the world influenced by arid areas (Celle-Jeanton *et al* 2009, Desboeufs *et al* 2010, Galy-Lacaux *et al* 2009, Kulshrestha *et al* 2009, Laouali *et al* 2012).

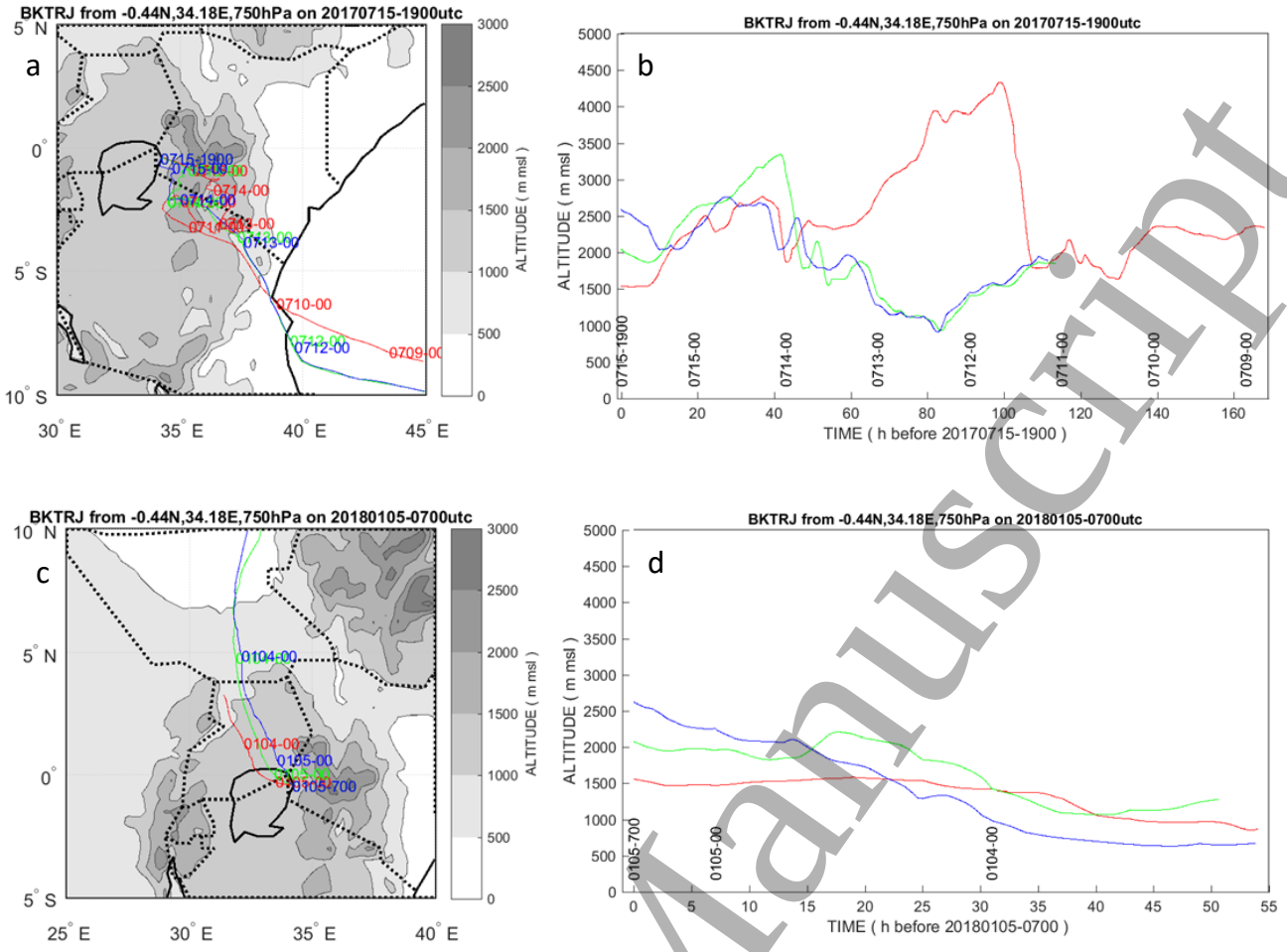

Figure 4: Back trajectories initiated in Mbita on the horizontal (left side) and vertical (right side) plans (a,b):15 July 2017 1900 UTC and (c,d) 5 January 2018 0700 UTC for 850 hPa (red line), 800 hPa (green line) and 750 hPa (blue line).

In addition to this terrigenous contribution, Ca^{2+}, Mg^{2+}, K^{+}, SO_4^{2-} particles can also be emitted to the atmosphere by biomass burning; furthermore, K^{+} in the sub-micron mode is considered as an atmospheric tracer of biomass burning (Andreae *et al* 1983, de Mello 2001) and SO_4^{2-} as a tracer of ammonium sulfate aerosols (Malavelle *et al* 2019). Maximum monthly VWM K^{+} concentrations (10 to 16 µeq L^{-1}), twice as large as the annual VWM (5.57 µeq L^{-1}), are measured in JJA 2018. The analysis of the mean monthly annual cycle of burned area from 2017 to 2019 using GFED4s fire emissions shows two periods of strong influence of biomass burning in the region: one in December, January, February (DJF) coming from the Northern part of Africa (South Sudan, Central African Republic and Democratic Republic of Congo) and one in JJA coming from the southern part of Africa (Tanzania and south of the Democratic Republic of Congo) (Fig. 5). These results are consistent with annual African biomass burning temporal dynamics described by Roberts *et al* (2009). In Mbita, Boiyo

et al (2018a) showed that high AOD in JJA was attributed to a combination of fine and coarse mode particles emphasizing the combined sources of dust and biomass burning. The fine mode of biomass burning in JJA is two times larger than in DJF, and in DJF coarse mode dust aerosols are predominant in mixture with fine combustion aerosols. Fig. 5 confirms the influence of biomass burning in the two seasons with a closer vicinity of burned surface in the south of Mbita in JJA and in the north in DJF. Mbita aerosols from biomass burning are related to biomass burning at the continental scale but also to neighboring agricultural zones and industrial-vehicular emissions due to the proximity of urbanized regions and roads (Boiyo *et al* 2017b, Makokha and Angeyo 2013). The main source contributing the most to aerosols from biomass burning are local activities linked to agriculture. Indeed, every year, bush fires are lit by farmers in different parts of the lake basin in order to prepare the land for cultivation (Makokha and Angeyo, 2013; Boiyo et al., 2017b). During March, April, May (MAM) and DJF, Mbita experiences significant local activities (land clearing and biomass burning) (Boiyo *et al* 2017b). The mixed type of aerosols combining marine and terrigenous contributions, combustion aerosols from biomass burning and anthropogenic activities is present in all seasons at Mbita, with the highest contribution in JJA as confirmed by maximum monthly VWM concentrations of marine, terrigenous and biomass burning tracers in JJA (Fig. 3).

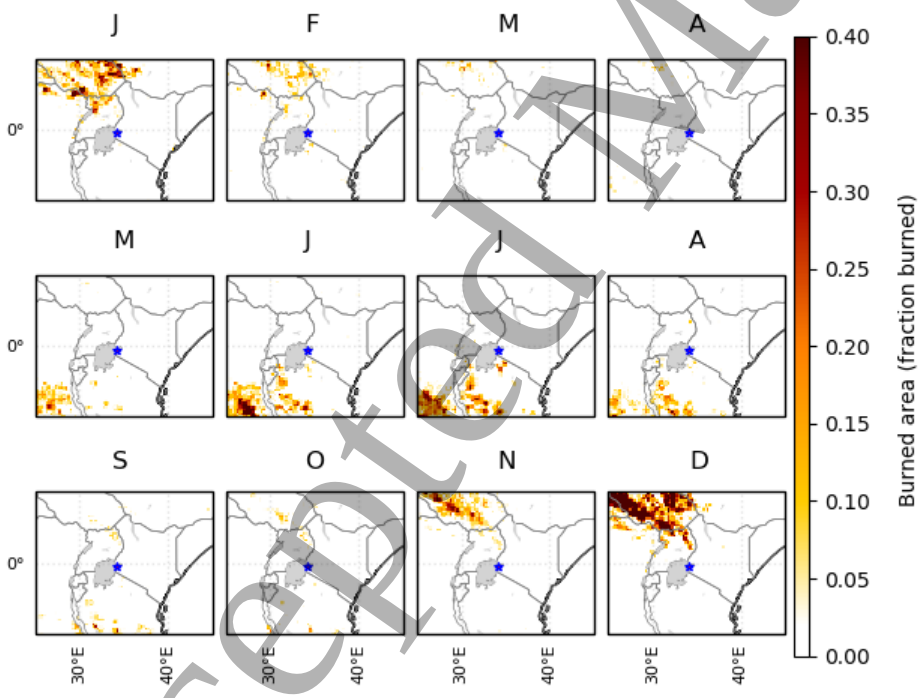

Figure 5 : Monthly mean burned area using GFED4s fire emissions from January 2017 to May 2019. The site of Mbita is marked as a blue star.

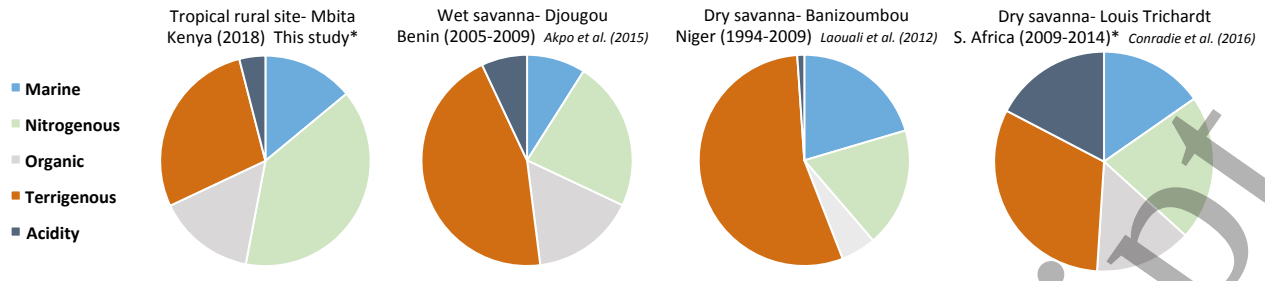

Figure 6: Estimation of the marine, nitrogenous, organic, acidity and terrigenous contributions to the rain chemical content measured at the tropical rural site of Mbita (Kenya) (*terrigenous in this study represents a mixture terrigenous/biomass burning as for Louis Trichardt) compared to two West and Central African sites: a wet savanna in Benin (Djougou) and a dry savanna in Niger (Banizoumbou), and to a south African remote dry savanna (Louis Trichardt).

The annual marine contribution to the total ionic precipitation content was calculated with Sea Salt Fraction VWM concentrations as: ($[Na^+]$+ SSF $[K^+]$+ SSF $[Ca^{2+}]$+ SSF $[Mg^{2+}]$+ $[Cl^-]$+ SSF $[SO_4^{2-}]$) / Total VWM ionic concentrations. The terrigenous/biomass burning contribution to the total ionic precipitation content was calculated with Non-Sea Salt Fraction (nSSF) VWM concentrations as: (nSSF $[K^+]$+ nSSF $[Ca^{2+}]$+ nSSF$[Mg^{2+}]$+ nSSF $[SO_4^{2-}]$) / Total VWM ionic concentrations. For 2018, the marine and terrigenous/biomass burning contribution to the rain chemical composition at Mbita was 14% and 28% respectively (Fig. 6).This contribution is comparable to the South African dry savanna (Conradie *et al* 2016) influenced by combustion and crustal sources (31%) but lower than in the Sahelian dry savanna (Laouali *et al* 2012) and the Benin wet savanna (Akpo *et al* 2015) where it contributes to 45 to 51% (Fig. 6).

WD fluxes are modulated by the monthly precipitation amounts and maximum monthly WD were recorded in MAM in the core of the 2018 wet season (Fig. 3b). Marine species maximum monthly WD were measured from June to October 2017 with a combination of high VWM and high unexpected rain depth at this period. Biomass burning and terrigenous influence in DJF is more important in 2019 than in 2018 due to larger rain amounts. The combination of large concentrations and high rain in August 2018 leads to large WD for all compounds (Fig. 3).

Acidity

The main potential acidifying components of rains are SO_4^{2-} and NO_3^- for mineral acidity and $HCOO^-$, CH_3COO^-, $C_2O_4^{2-}$ and $C_2H_5COO^-$ for organic acidity (Table 3). These concentrations reflect potential contributions to the acidity in Mbita rains of NO_3^- (27.72%), $HCOO^-$ (26.56%), SO_4^{2-} (21.66%), CH_3COO^- (17.83%), $C_2O_4^{2-}$ (5.48%) and $C_2H_5COO^-$ (0.77%), and a negligible contribution of

$C_2H_5COO^-$. Organic and mineral acids represent 50.63% and 49.37% of the potential acidity, respectively (Table 3). Acetic and formic acids are generally derived from vegetation, biomass burning and bio fuel, fossil fuel, agricultural emissions and soils (Paulot *et al* 2011). Wet deposition is one of the major sinks of organic acids. Monthly VWM concentrations of organic species in rain exhibit peaks in JF and JAS. Mbita rain presents a total VWM organic acids concentration of 16.22 μeq L^{-1} representing 15% of the total ionic content in rain. This result is comparable to values found in the wet savanna of Djougou in Benin (Akpo *et al* 2015) and in the South African dry savanna (14-16%, Conradie *et al* 2016), but higher than in Sahelian dry savanna of Niger (5%, Laouali *et al* 2012) (Fig. 6).

The pH of the rainwater at Mbita for the year 2018 varies between 4.5 and 7.5 with an annual average value of 5.8 ± 0.6 (Fig. 7). As a comparison, Visser (1961) observed a median pH of 7.9 near Kampala (Lake Victoria, Kenya side), Rodhe *et al* (1981) reported a median of 6.1 for nine East African sites. The pH distribution shows that 50.6% of the analyzed rains are alkaline, 35.3% acid and 14.1% neutral compared to a pH of 5.6 considered as neutral for rain (Fig. 7). Yoboué *et al* (2005) observed a positive gradient for rain acidity along the West African transect dry savanna - wet savanna - equatorial forest with average pH values of 5.67, 5.16 and 4.92 respectively. Mean pH in Mbita presents a slightly alkaline character and is close to pH in rural dry savanna areas. At the 2018 annual scale, VWM concentrations of H^+ calculated from the mean pH is 4.21 μeqL^{-1}.

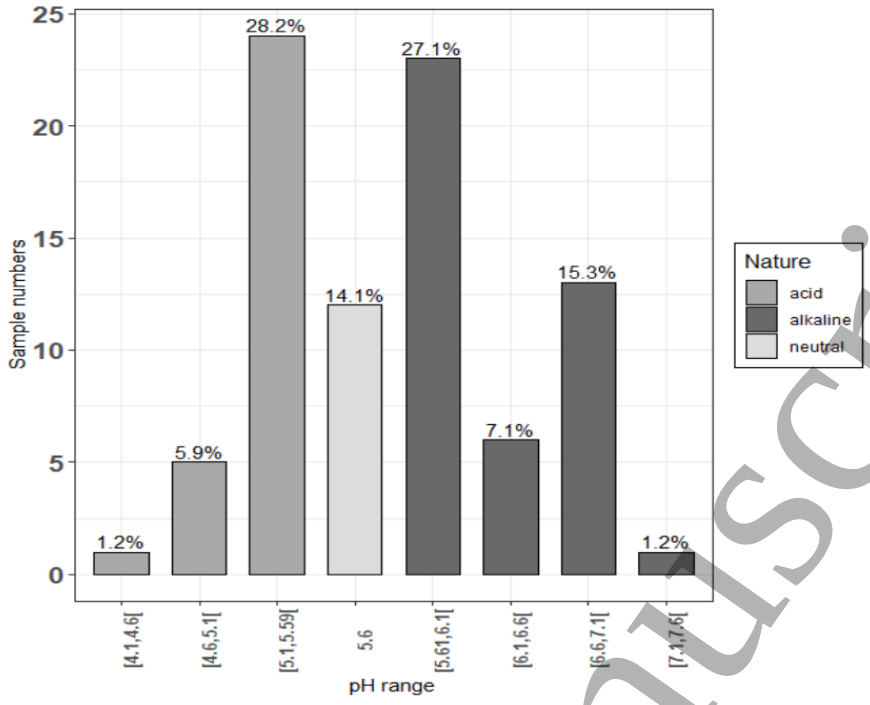

Figure 7: Frequency distribution of pH measured in Mbita rainwater in 2018.

The pH value alone is not sufficient to give information about acidity of rain water. Reactions between acidic and alkaline species in rainwater determine the final rainwater pH values. Using potential acidity (Sup. Mat. Eq. 1) and fractional acidity (Sup. Mat. Eq. 2), 87% of potential rain acidity (27.8 µeq L^{-1}) is found to be neutralized by alkaline ions (Table 3). Analyses of Neutralization Factors (Sup. Mat. Eq. 3) suggest that NH_4^+ and Ca^{2+} are responsible for most of the acid neutralization with NF values for NH_4^+, Ca^{2+} of 1.32 and 0.55 respectively. The large NH_4^+/ NO_3^- and NH_4^+/SO_4^{2-} ratios (4.14 and 5.19) suggest that nitrogenous species are completely neutralized in the atmosphere as a result of the formation of NH_4NO_3 and $(NH_4)_2SO_4$ aerosols (Duan *et al* 2003, Seinfeld 1986). This is confirmed by the Ammonium availability Index (AAI, Sup. Mat. Eq. 4) of 161 % in 2018, indicating that ammonium is sufficient to completely neutralize the sulfuric and nitric acids in Mbita rainwater (Behera and Sharma 2010). Ratios between cations and anions $(Ca^{2+}+Mg^{2+}+NH_4^+)/(NO_3^-+SO_4^{2-})$ = 3.48 and $(NO_3^-+SO_4^{2-})/(Ca^{2+}+Mg^{2+})$ = 0.87 reflect the alkaline nature of rainwater and the influence of Ca^{2+} and Mg^{2+} compounds described earlier. In general, acidity was not correlated with mineral acids, and weakly correlated with organic acids. These results confirm the presence of heterogeneous chemistry processes of neutralization between alkaline dust particles, gaseous nitric and sulfuric acid and organic ions. Galy-Lacaux *et al* (2001) showed that this capture could be completed and could

explain to a large extent the neutralization processes of rain's potential acidity. This result has been widely emphasized in other parts of the world where desert soil dust emissions influence rain chemistry, as for example in Spain or in the Mediterranean basin (Avila *et al* 1998, 1997, Herut *et al* 2000), and in Asia (Hu *et al* 2003, Kulshrestha *et al* 2005, 2003).

Table 3: Analysis of potential contributions of mineral and organic acids to total acidity in Mbita rain in 2018

Organic Acidity	**VWM (μeqL^{-1})**	**Mineral Acidity**	**VWM (μeqL^{-1})**
$HCOO^-$	8.51	**NO_3^-**	8.88
CH_3COO^-	5.71	**SO_4^{2-}**	6.94
$C_2H_5COO^-$	0.25		
$C_2O_4^{2-}$	1.75		
Total Organic (TO)	**16.22**	**Total Mineral (TM)**	**15.82**
Potential Acidity (pA) = TO+TM	**32.04**		
Calculated H^+	**4.21**		
pA-H^+	27.83		

Nitrogenous contribution

NH_4^+ is the dominant nitrogen ion in precipitation in Mbita at both monthly and annual timescales (Table 2). The predominance of NH_4^+ in rainwater has also been observed in dry savanna (19.1 to 25.2 μeq L^{-1}; (Laouali *et al* 2012)), including savannas influenced by industrial emissions (~29 μeq L^{-1}; (Conradie *et al* 2016)), as well as in wet savanna sites (14.3 to 16.8 μeq L^{-1}; (Yoboué *et al* (2005); Akpo *et al* (2015)), though concentrations in Mbita are higher than in these studies. Annual VWM NO_3^- concentrations at Mbita are comparable to those obtained in wet savannas (8.2 to 8.9 μeq L^{-1}, Yoboué *et al* 2005, Akpo *et al* 2015), but lower than those found in dry savannas (10.4 to 14.6 μeq L^{-1}, Laouali *et al* 2012, Conradie *et al* 2016).

The mean and standard deviation (±SD) of DIN concentrations (NH_4^+ + NO_3^- + NO_2^-) in Mbita is 1.41±1.6 mgN.L^{-1}. In comparison to older studies in the same region, our results show higher values. Indeed, data from the literature give DIN concentration of 0.87 mg.L^{-1} at Kampala (Uganda, Lake Victoria shore, (Visser 1961), 0.09 to 0.13 mg.L^{-1} on the shore of Lake Malawi (Bootsma and Hecky

1999), from 0.012 to 4.2 mgN L^{-1} at Lake Victoria sites in Tanzania (Vuai *et al* 2013), and 0.92 mgN L^{-1} on Lake Tanganyika (Gao *et al* 2018).

The sum of NH_4^+, NO_3^- and NO_2^- VWM concentrations represents 39% of the total chemical content of rain, in contrast to other remote African sites where the values ranged from 17 to 23% (Fig. 6).

Ammonium content in precipitation results from the inclusion of gaseous ammonia and particles. Nitrate content results from NO_2 and HNO_3 concentrations in the atmosphere, further scavenged by clouds. Major sources of NH_3 include bacterial decomposition of urea in animal excreta and emission by natural or fertilized soils (Delon *et al* 2012, Galy-Lacaux and Modi 1998, Schlesinger and Hartley 1992). Biomass burning and domestic combustions are other principal sources of NH_3 (Brocard *et al* 1996, Galy-Lacaux and Modi 1998, Laouali *et al* 2012, Delmas *et al* 1995). In Mbita, monthly NH_4^+ and NO_3^- VWM concentrations in rain exhibit maximum values in JJA and in January from 2017 to 2019 (Fig. 3a). As mentioned above, Mbita is largely influenced by biomass burning sources during these two periods as well as additional local activities (land clearing and biomass burning) in DJF (Boiyo *et al* 2017b) (Fig. 5). Moreover, monthly mean IASI NH_3 and OMI NO_2 Vertical Density Columns from 2017 to May 2019 show the same feature (Fig. 8). Results clearly indicate that NO_2 are enhanced in DJ and JJA in relation with the biomass burning sources occurrence in the northern and southern hemisphere. NH_3 VCD maximum occurs in February and is certainly related to both biomass burning and local land clearing by farmers' agricultural burning to prepare their fields. It has been reported that NH_4^+/NO_3^- ratio for N deposition is usually less than 1 in areas with advanced industrialization while areas with intensive agriculture are characteristic of high NH_4^+/NO_3^- ratios (>1) (Fahey *et al* 1999, Zhao *et al* 2009). In our study, the NH_4^+/NO_3^- ratio of 4.13 indicates that NH_3 emissions are certainly also related to mixed crops agriculture in Mbita region as well as livestock presence.

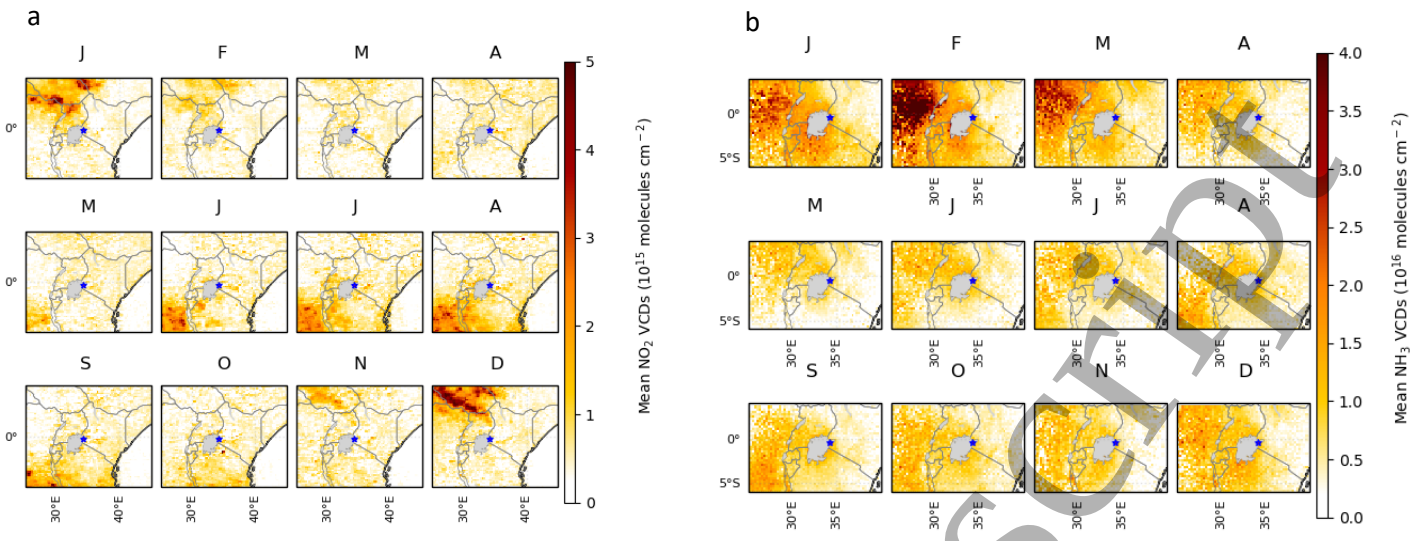

Figure 8: (a) Monthly mean OMI tropospheric NO_2 from January 2017 to May 2019 and (b) IASI tropospheric NH_3 Vertical Column Densities (VCDs) in 10^{15} molecules cm^{-2} from January 2018 to May 2019. The site of Mbita is marked as a blue star.

3.2 Nitrogen wet deposition budget and Dissolved Organic Nitrogen contribution

Monthly VWM concentrations of DIN and DON present larger values during the dry seasons (Fig. 9a), especially in JJA 2017, January 2018, JJASO 2018 and JF 2019. DIN concentrations were discussed in the previous section, showing that Mbita is influenced by biomass burning sources from both hemispheres during the dry season, as well as agriculture and livestock sources year round. Concentration of DON in rainwater results from the presence of aerosols in the atmosphere, further dissolved in rain. The total organic nitrogen content of these aerosols is composed of protein compounds (Matos *et al* 2016, Pöschl 2005, Zhang *et al* 2002). Atmospheric organic matter is increasingly considered to be an indispensable part of the global nitrogen cycle (Wedyan and Preston 2008, Zamora *et al* 2011). In particular, the water-soluble fraction of aerosol organic matter has direct links to bioavailable nutrients. Protein compounds are characterized by their solubility in water (Matos *et al* 2016), suggesting that water-soluble protein compounds are extremely important in the nutrient cycle of atmospheric and biospheric nitrogen.

DON concentrations contribute from 7 to 60% (average 29±15%) of TDN concentrations, with the highest value (60%) in August 2017. Air masses were found to be particularly influenced by marine sources from the Indian Ocean at that period, and this influence is retrieved in DON concentrations. Indeed, oceans are a source of organic nitrogen (Calderón *et al* 2006), through aerosol sea spray containing amino acids (Neff *et al* 2002), and from metabolic processes in marine animals and bacteria releasing amines in the gas phase (Yang *et al* 1994). The important contribution of DON WD fluxes (nearly 31%) throughout the period of study may be explained by biomass burning, livestock sources and dust resuspension. Agricultural sources of organic nitrogen include urea applied as fertilizer (Cornell *et al* 1998, Mace 2003a), and aliphatic amines from animal husbandry operations (Schade and Crutzen, 1995). Biomass combustion, particularly active in DJF and JJA, releases amino acids (Mace 2003b, 2003a, Spitzy 1990), as well as other substances such as humic acid-like compounds that can be photolyzed to release free amino compounds (Chan *et al* 2005, Matsumoto and Uematsu 2005). A lot of studies showed that biomass combustion, continental biogenic discharges (including viruses, protozoa, algae, fungi, bacteria, pollen, spores, human and animal epithelial cells, and insect and plant fragments), and agricultural activities are the main sources of aerosolized protein materials (Cape *et al* 2012, 2011, Matos *et al* 2016, Song *et al* 2017).

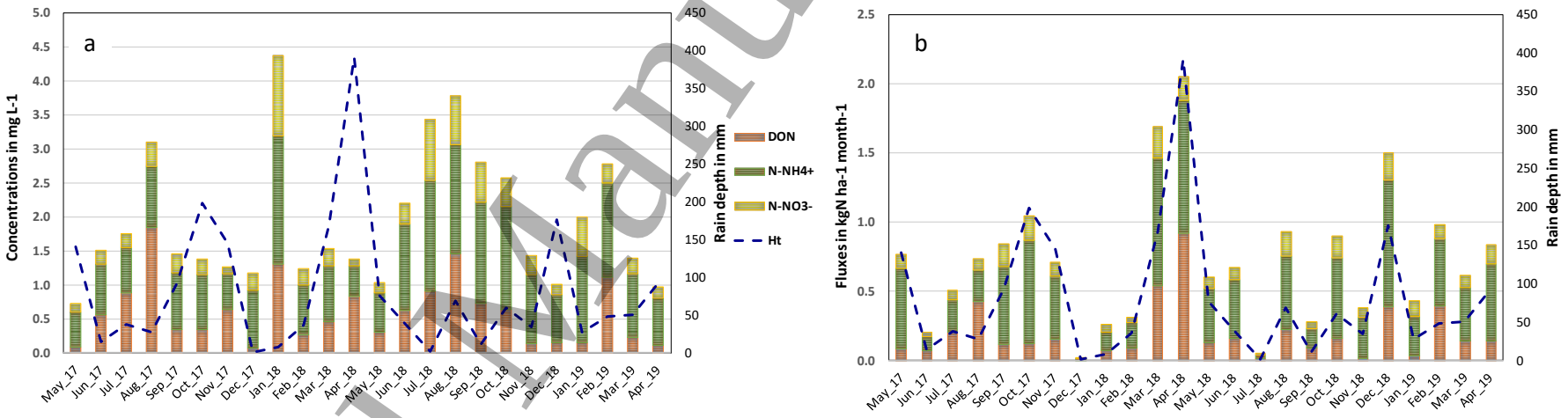

Figure 9: (a) Monthly concentrations in mg L^{-1} of Dissolved Organic Nitrogen (DON) and Dissolved Inorganic Nitrogen (DIN) in the forms of $N\text{-}NO_3^-$ and $N\text{-}NH_4^+$ (b) Wet deposition Nitrogen budget at Mbita over the period 2017-2019. Dotted blue line is monthly rain depth (Ht, mm) at Mbita over the period 2017-2019.

The mean and standard deviation (range) of WD fluxes for NH_4^+-N, NO_3^--N, and DON, are 0.42±0.28 (0.01-0.96), 0.10±0.06 (0.003-0.23), and 0.19±0.21 (0.001-0.92) kgN ha^{-1} month^{-1} respectively. On a monthly time, scale, maximum DIN and DON deposition fluxes are observed during wet seasons (Fig. 9b). DON contribution varies from 4 to 58% of TDN, NH_4^+-N contribution from 30 to 78%, and NO_3^--N from 8 to 27%. DIN contribution at the monthly scale remains larger during the whole period, with a large predominance of NH_4^+-N wet deposition fluxes, except during the special period around July 2017 where DON contribution exceeds DIN contribution, due to the high unexpected marine influence. Maximum DIN WD fluxes (> 1 kgN ha^{-1} month^{-1}) are observed at the beginning of the long rain season in March and April 2018, and in December 2018, because of strong rains and large DIN concentrations. Gao *et al* (2018) also found large WD DIN fluxes (2.37 kgN ha^{-1} month^{-1}) into Lake Tanganyika at the beginning of the rain season, attributed to soil preparation by farmers (stubble burning) before short and intense rains.

The annual WD fluxes for NH_4^+-N, NO_3^--N, and DON are 5.06, 1.25 and 2.32 kgN ha^{-1} yr^{-1} (Table2, Table 4). Annual DIN WD represents 6.31 kgN ha^{-1} yr^{-1}, with long (MAM) and short (ON) rain seasons accounting for 36 and 21% respectively, while rainfall amounts for the same seasons represent 47 and 22% respectively.

Annual DIN WD fluxes in Mbita are comparable to WD measured in other sites of Lake Victoria by Vuai *et al* (2013), and in other East African great lakes, such as Lake Tanganyika (Gao *et al* 2018, Langenberg *et al* 2003), and Lake Kivu ((Namugize 2015), Table 4). The predominance of N-NH_4^+ in DIN WD fluxes reported by Gao *et al* (2018) is in accordance with our results. Similar DIN WD fluxes were also measured in West and Central Africa (Galy-Lacaux and Delon 2014) and in industrialized south African dry savannas (Conradie *et al* 2016). In other tropical regions DIN WD fluxes show comparable values (Amazoniana (1972) in Brazil; Eklund *et al* (1997) in Costa Rica, Table 4). On the other hand, DIN WD fluxes from our study are two times lower than those estimated from annual rainfall and VWM given in Visser (1961) in Lake Victoria, and larger than those estimated from Bootsma *et al* (1996) and Bootsma and Hecky (1993) in Lake Malawi. These differences may be attributed to collection protocols and analysis techniques (colorimetric or spectroscopic) of the samples as well as the time scales which differ from one study to another. Our study was carried out over a much longer sampling period (2 years), continuously (at the rain event), thus giving a more representative average characterization of the total N WD flux than previously found in older studies. The sampling method of the cited authors was either in bulk, weekly or monthly, sometimes even

seasonally and for shorter periods than the ones reported in this work. Local climatic conditions and air mass movements could also explain this difference. Furthermore, few DON estimations are available in the literature for tropical ecosystems, and are often inferred from bulk deposition, as given by Van Langenhove *et al* (2020) for a tropical forest in French Guiana with 8.75 kgN ha^{-1} yr^{-1} (Table 4), or in Bauters *et al* (2018) reporting DIN and DON contributions in the Congo forest (5.5 and 12.7 kgN ha^{-1} yr^{-1} respectively).

Wet N deposition (DIN+DON) in Mbita gives a WD flux of 8.54 kgN ha^{-1} yr^{-1}. From Kayombo and Jorgensen (2006) we estimated a 38% contribution of dry deposition to the atmospheric (wet+dry) deposition in Lake Victoria. With this value, a total atmospheric deposition (wet+dry) of 15 kgN ha^{-1} yr^{-1} may be estimated from our results, to be compared to 12.43 kgN $ha^{-1}yr^{-1}$ in Lake Malawi and to values ranging from 7.5 to 19 kgN $ha^{-1}yr^{-1}$ reported by Scheren *et al* (2000) for remote non marine tropical watersheds. These values exceed the 10 kgN ha^{-1} yr^{-1} threshold given by Bobbink *et al* (2010) above which African ecosystems will be subject to stress due to N deposition excess. According to the global distribution map of mean critical loads for eutrophication (Bouwman *et al* 2002), the Lake Victoria ecosystem is exposed to eutrophication.

From DIN WD fluxes reported in Table 4 and with the surface of African great lakes, we estimated the total DIN input in lakes Kivu (2, 400 km^2), Tanganyika (32, 600 km^2) and Victoria (68, 800 km^2), with 2, 176 tN yr^{-1}, 20, 000 tN yr^{-1} and 44, 032 tN yr^{-1} respectively. Taking into account the total N WD flux from this study, including DON estimation, Lake Victoria receives 58, 760 tN yr^{-1}. According to Zhou *et al* (2014), Nile river and fishery N export from Lake Victoria represent 40, 000 tN yr^{-1} and 4, 000 tN yr^{-1} respectively, leading to 14, 400 tN yr^{-1} staying in the lake and contributing to eutrophication.

Table 4: Dissolved Inorganic Nitrogen (DIN), Dissolved Organic Nitrogen (DON) and Total Dissolved Nitrogen (TDN) in kgN ha^{-1} yr^{-1} in tropical ecosystems and African great lakes.

Site	Pi (mm)	Characteristics	N-NH_4^+	N-NO_3^-	N-NO_2^-	DIN	DON	TDN	References
Lake Victoria									
Kampala (Uganda)	1300	ND	6.36	5		11.36			(Visser 1961)
North shore (Kenya)	ND	ND				11.96			(Bootsma and Hecky 1993)
Itumbiri (Tanzania)	1000	Rural				7.7			(Vuai *et al* 2013)
Tanzania side	1050	Peri Urban				6.8			
Tanzania side	1050	Urban				6.4			
Lake Victoria									
Mbita (Kenya)	1070	Tropical Rural	5.14	1.25	0.01	6.4	2.32	8.54	**This study**
Lake Tanganyika									
Burundi-RDC-Tanzania-Zambia	819	Tropical ecosystem	4.18	1.67		5.86			(Gao *et al* 2018)
Burundi-Tanzania-Zambia	1200	Tropical-Agriculture				6.72			(Langenberg *et al* 2003)
-	ND	ND				12.05			(Bootsma and Hecky 1993)
Lake Malawi									
Monkey Bay (Malawi)	ND	ND				0.85-1.56			(Bootsma and Hecky 1993)
Senga Bay (Malawi)	ND	ND				1.5			(Bootsma and Hecky 1999)
Lake Kivu (RDC)									
Catchment	ND	Agricultural land	0.27	3.74		4.01			(Namugize 2015)
La Selva (Costa Rica)	3500-4500	Tropical forest	2.6 - 5	1.8 - 3.4		4.4-8.4			(Eklund *et al* 1997)
West and Central Africa									
Sahelian dry savannas	449-744	Rural sites				2-3.4			(Galy-Lacaux and Delon 2014)
Wet savannas	1261-1274					3.5-5.3			
Forest	1557					3.6			
South Africa									
Amersfoort	729.75	Semi-arid				6.32			(Conradie *et al* 2016)
Vaal Triangle	956.43					6.97			
Louis Trichardt	728.2					1.87			
Skukuza	583.23					2.12			
Other tropical regions									
Paracou (French Guiana)	3100	Tropical forest	0.677*	1.092*			8.75*		Van Langenhove *et al. (*2020)
Lake Valencia (Venezuela)	ND	ND	2.43	1.28	0.05	3.76	1.33*		(Lewis 1981)
Amazon basin(Brazil)	ND	ND	3.15	2.52		5.67			(Amazoniana 1972)
Amazon basin(Venezuela)	ND	ND	21.4						(Jordan, *et al* 1979)
Congo forest (RDC)		Tropical forest				5.46	12.74	18.20	(Bauters *et al* 2018)

Conclusion

This study presents for the first time a complete characterization of the precipitation chemistry in a tropical rural site in Kenya on the shores of the Lake Victoria, based on the monitoring of quality controlled in situ measurements from May 2017 to April 2019. Wet deposition fluxes of major ionic species considered as important nutrient inputs to ecosystems were characterized and quantified at the annual and monthly scales. We emphasize that wet deposition is influenced by the variation in emission source strength, precipitation rate and the origin of air masses. In the Lake Victoria basin, four main contributions to the chemical composition of precipitation were related to potential atmospheric sources of gases and particles of importance. Marine species, with a contribution of 14%, were clearly related to marine air masses coming from the Indian Ocean. The organic contribution in precipitation is estimated for the first time on the shores of the lake Victoria and represents about 15%, due to VOC from biomass burning combustions from both the southern and northern hemisphere, and by BVOC from vegetation (forests in the vicinity of the site). Terrigenous species were attributed to the crustal and biomass burning sources contributing 28% of the total rain chemical content, due to biomass burning sources in both hemispheres and dust emissions from arid areas in the North and East of Kenya. The most important result is the nitrogen species' largest contribution to the total chemical content of the rains (39%), with the predominance of NH_4^+ ion in precipitation due to local sources such as livestock and fertilizers, as well as regional and continental biomass burning and nearby agricultural fires. This means that nitrogen deposition to Lake Victoria is strongly influenced by anthropogenic sources.

This study gives unprecedented insights in the seasonality of N wet deposition inputs. DON contribution to the high N load in rains is 26% of the total WD flux, highlighting the crucial need to quantify organic N in rains in tropical ecosystems. With a total N atmospheric input to Lake Victoria of 103, 200 tN yr^{-1} estimated thanks to our results and literature results, and according to critical loads for N eutrophication, we assume that the Lake Victoria ecosystem is exposed to eutrophication.

In a near future, this study on wet N deposition on the Lake Victoria basin will be complemented by a quantification of N dry deposition fluxes. We highly recommend extensive and regular monitoring of atmospheric deposition including wet and dry processes over seasonal representative temporal periods in Kenya, Tanzania and Uganda both in-shore and off-shore to provide guidance and improve the efficiency of nitrogen use in agricultural areas, and reduce losses to the Lake Victoria environment. This could be done through the organization of awareness-raising campaigns toward end users. In order to reduce nitrogen pollution of the lake, it will be necessary in the future to improve agricultural

techniques and management of human and animal waste. Finally, ongoing and future studies on N deposition are very important and should be communicated to policy makers to manage the unique natural resource represented by the Lake Victoria and its catchment and to mitigate nutrient budgets in the great African lakes ecosystems.

Acknowledgements: This study was supported by the United Nations Environmental Program (GEF-UNEP) International Nitrogen Management System (INMS) project, and by the French program Cycle de l'Azote entre la Surface et l'Atmosphere en afriQUE (CASAQUE) funded by Les Enveloppes Fluides et Environnement (LEFE) / Ecosphère Continentale et Côtière (EC2CO) of the Institut National des Sciences de l'Univers- Centre National de la Recherche Scientifique (INSU-CNRS). This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 871944. We acknowledge Peter Nyongesa and Francis Ncodimba from ICIPE Center for hosting this project and facilitating logistics. We are particularly grateful to the technician field, Sylas Okumu, for his work. IASI is a joint mission of EUMETSAT and the Centre National d'Etudes Spatiales (CNES, France). The version 3 of the IASI-NH_3 dataset was created by the SQUARES group at ULB (Université libre de Bruxelles (ULB), Spectroscopy, Quantum Chemistry and Atmospheric Remote Sensing (SQUARES), Brussels, Belgium). We would like to thank Martin Van Damme, Lieven Clarisse, and Pierre Francois Coheur for the NH_3 dataset.

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