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

Adama Bakayoko¹, Corinne Galy-Lacaux², Véronique Yoboué¹, Jonathan E Hickman³, Frank Roux², Eric Gardrat¹, Frédéric Julien¹ and Claire Delon²

¹ Laboratoire des Sciences de la Matière, de l’Environnement et de l’Energie Solaire (LASMES), Université Félix Houphouët Boigny, Abidjan, Côte d’Ivoire
² Laboratoire d’Aérologie (Laero), Université de Toulouse, CNRS, UPS, Toulouse, France
³ NASA Goddard Institute for Space Studies, New York, NY, United States of America
⁴ Laboratoire Ecologie Fonctionnelle et Environnement, Université de Toulouse, CNRS, INP-ENSAT, Toulouse, France

E-mail: bakayokoadma@gmail.com and corinne.galy-lacaux@aero.obs-mip.fr

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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 2 yr 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 (WD) inputs in the unique natural resource represented by Lake Victoria and its catchment. Four main contributions to the chemical composition of precipitation, were identified: (a) a 28% terrigenous contribution related to crustal and biomass sources, (b) a 14% marine contribution related to Indian ocean air masses intrusion, (c) a 15% organic contribution due to volatile organic carbon emissions from biomass burning and vegetation and (d) 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 WD 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.

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 (WD) 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 WD, 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 km² and a total basin area of 195 000 km². Precipitation is the largest contributor of water (80%) to the lake (Tamatamah et al. 2005, Sutcliffe and Petersen 2007), leading some authors to describe the lake as ‘atmosphere controlled’ (Tate et al. 2004, Kizza et al. 2009). 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 (Nyiliita 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, Zhou et al. 2014, Masso et al. 2017, Nyiliita et al. 2020), where the authors also point out that N WD is missing in the budget estimation, due to the lack of available measurements. Indeed, very few studies on precipitation chemistry and WD 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, Vuai et al. 2013, Gao et al. 2018), highlighting the necessity to accurately quantify this input. Assessing WD 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 WD 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 International Nitrogen Management System project, and of the French program Cycle de l’Azote entre la Surface et l’Atmosphère en afriQUE. This work is based on a 2 y monitoring period (May 2017–April 2019) of WD 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 (a) a unique quantification of important biogeochemically species WD and (b) a comprehensive atmospheric WD 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 (figure 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 (figure 1(a)). 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 1131 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 (figure 1(b)). 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 (Makokha and Angeyo 2013, Boiyo et al. 2017b, 2018b). In addition to local sources, the long-range transport of aerosols towards the site could significantly influence the aerosol load (Gatebe et al. 2001, Boiyo et al. 2017a). 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 were described in several studies (Bootsma and Hecky 1993, Kayombo and Jorgensen 2006, Zhou et al. 2014, Masso et al. 2017, Nyiliita et al. 2020), where the authors also point out that N WD is missing in the budget estimation, due to the lack of available measurements. Indeed, very few studies on precipitation chemistry and WD 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, Vuai et al. 2013, Gao et al. 2018), highlighting the necessity to accurately quantify this input. Assessing WD 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 WD 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).

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represents more than 100 cattle per square kilometer (FAO 2005).

2.2. Climatology

A long term database of key meteorological parameters is available at ICIPE center for the period 1985–2019. It includes ambient 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 (figure 2(a)) (for more details, see supplementary material (Sup. Mat.) available online at stacks.iop.org/ERL/16/045013/mmedia 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 (figure 2(a)) 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 (figure 2(a)). 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 (Ogwang et al. 2015, Yang et al. 2015, Boiyo et al. 2017b).

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.): (a) a period in deficit from 1985 to 1992, (b) a period in excess from 1999 to 2013 and (c) 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) (figure 2(b)). The driest year with an index of −1.7 is observed in 1990 with 655.7 mm. Details may be found in the Sup. Mat.

2.3. Sample collection and analytical procedures

Rainwater samples at Mbita were collected using an automatic precipitation collector designed for the 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 was 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 precip-
itation (Pc). The annual and quarterly percent cover-
age length (%PCL) is also indicated in table 1.

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 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 stud-
ied period according to the parameters calculated in

2.4. Analytical methods and quality procedure
The chemical analyses of the samples were performed
at the Chemical Laboratory of the Laero, in Toulouse

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 and (c) standardized
precipitation index (SPI) over the period 1985–2019.
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$_3$COO$^−$ > HCO$_3^−$ > C$_2$O$_4^{2−}$ > C$_3$H$_2$COO$^−$ > NO$_2^−$.

3.1.1. 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 (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 (SSFs, 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 (figure 3(a)), 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 (figures 4(a) and (b)) 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 d for the air observed at 800 and 750 hPa at Mbita. The same course took 5.5 d for the air observed at 800 and 750 hPa at Mbita. The same course took 5.5 d for the air observed at 800 and 750 hPa at Mbita. The same course took 5.5 d for the air observed at 800 and 750 hPa at Mbita.
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.
monthly VWM concentrations from 65 to 96 µeq l⁻¹ (figure 3(a)), 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 (1–50 µeq l⁻¹). The analysis of monthly aerosol optical depth (AOD) measurements by Boiyo et al (2018b, 2019) obtained at Mbita ICPE 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 Ca²⁺ contribution is the second most important after NH₄⁺ (table 2). Ca²⁺ is significantly correlated to K⁺, Mg²⁺ and SO₄²⁻ (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 Ca²⁺, Mg²⁺, K⁺, SO₄²⁻ are 98%, 50%, 97%, 88% respectively. Monthly VWM evolution of Ca²⁺, Mg²⁺, K⁺, SO₄²⁻ concentrations over the period 2017–2019 exhibit two peaks in JJA and in January (figure 3(a)). The analysis of back trajectories arriving at Mbita on 5 January 2018 0700 UTC (figures 4(c) and (d)) 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 (figure 4(d)). The origin of air at 850 hPa is much closer as it stays very close to the surface level. Ca²⁺ 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⁺, Mg²⁺, SO₄²⁻ and Ca²⁺ 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 (Kaufman 2005, Jish Prakash et al 2015). Boiyo et al (2018a, 2018b, 2019) 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 Mg²⁺, SO₄²⁻ and K⁺ (Avila et al 1997). Terrigeneous contributions were also found in other African ecosystems and regions of the world influenced by arid areas (Celle-Jeanton et al 2009, Galy-Lacaux et al...
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.

2009, Kulshrestha et al 2009, Desboeufs et al 2010, Laouali et al 2012).

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 (10–16 \(\mu\)eq l\(^{-1}\)), twice as large as the annual VWM (5.57 \(\mu\)eq 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) (figure 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. Figure 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 (Makokha and Angeyo 2013, Boiyo et al 2017b). 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 (figure 3).

The annual marine contribution to the total ionic precipitation content was calculated with Sea Salt Fraction VWM concentrations as: 

\[
\left(\frac{[\text{Na}^{+}] + \text{SSF}[\text{K}^{+}] + \text{SSF}[\text{Ca}^{2+}] + \text{SSF}[\text{Mg}^{2+}] + [\text{Cl}^{-}] + \text{SSF}[\text{SO}_4^{2-}]}{\text{total VWM ionic concentrations}}\right)
\]

The terrigenous/biomass burning contribution to the total ionic precipitation content was calculated with nSSF VWM concentrations as: 

\[
\left(\frac{\text{nSSF}[\text{K}^{+}] + \text{nSSF}[\text{Ca}^{2+}] + \text{nSSF}[\text{Mg}^{2+}] + \text{nSSF}[\text{SO}_4^{2-}]}{\text{total VWM ionic concentrations}}\right)
\]
respectively (figure 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%–51% (figure 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 (figure 3(b)). 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 DIF 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 (figure 3).

### 3.1.2. Acidity

The main potential acidifying components of rains are SO$_4^{2-}$ and NO$_3^-$ for mineral acidity and HCOO$^-$, CH$_3$COO$^-$, C$_2$O$_4^{2-}$ and C$_2$H$_5$COO$^-$ 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$_3$COO$^-$ (17.83%), C$_2$O$_4^{2-}$ (5.48%) and C$_2$H$_5$COO$^-$ (0.77%), and a negligible contribution of C$_3$H$_7$COO$^-$. 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). WD 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) (figure 6).

![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 (Banizombou), and to a south African remote dry savanna (Louis Trichardt).](image-url)

| Table 3. Analysis of potential contributions of mineral and organic acids to total acidity in Mbita rain in 2018. |
|---|---|---|
| Organic acidity | VWM (µeq l$^{-1}$) | Mineral acidity | VWM (µeq l$^{-1}$) |
| HCOO$^-$ | 8.51 | NO$_3^-$ | 8.88 |
| CH$_3$COO$^-$ | 5.71 | SO$_4^{2-}$ | 6.94 |
| C$_2$H$_5$COO$^-$ | 0.25 | | |
| C$_2$O$_4^{2-}$ | 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 |

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 (figure 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 (figure 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 µeq l$^{-1}$.

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. equation (1)) and fractional acidity (Sup. Mat. equation (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. equation (3)) suggest that NH$_4^+$ and Ca$^{2+}$ are responsible for most of the acid neutralization with NF values
for $\text{NH}_4^+$, $\text{Ca}^{2+}$ of 1.32 and 0.55 respectively. The large $\text{NH}_4^+/\text{NO}_3^-$ and $\text{NH}_4^+/\text{SO}_4^{2-}$ ratios (4.14 and 5.19) suggest that nitrogenous species are completely neutralized in the atmosphere as a result of the formation of $\text{NH}_3\text{NO}_3$ and $(\text{NH}_3)_2\text{SO}_4$ aerosols (Seinfeld 1986, Duan et al 2003). This is confirmed by the ammonium availability index (Sup. Mat. equation (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 ($\text{Ca}^{2+} + \text{Mg}^{2+} + \text{NH}_4^+)/(\text{NO}_3^- + \text{SO}_4^{2-}) = 3.48$ and $(\text{NO}_3^- + \text{SO}_4^{2-})/(\text{Ca}^{2+} + \text{Mg}^{2+}) = 0.87$ reflect the alkaline nature of rainwater and the influence of $\text{Ca}^{2+}$ and $\text{Mg}^{2+}$ 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 1997, 1998, Herut et al 2000), and in Asia (Hu et al 2003, Kulshrestha 2003, 2005).

3.1.3. Nitrogenous contribution

$\text{NH}_4^+$ is the dominant nitrogen ion in precipitation in Mbita at both monthly and annual timescales (table 2). The predominance of $\text{NH}_4^+$ in rainwater has also been observed in dry savanna (19.1–25.2 $\mu$eq l$^{-1}$; Laouali et al (2012)), including savannas influenced by industrial emissions ($\sim$29 $\mu$eq l$^{-1}$; Conradie et al (2016)), as well as in wet savanna sites (14.3–16.8 $\mu$eq 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 (8.2–8.9 $\mu$eq l$^{-1}$, Yoboué et al (2005), Akpo et al (2015)), but lower than those found in dry savannas (10.4–14.6 $\mu$eq l$^{-1}$, Laouali et al (2012), Conradie et al (2016)).

The mean and standard deviation (±SD) of dissolved inorganic nitrogen (DIN) concentrations ($\text{NH}_4^+ + \text{NO}_3^-, \text{NO}_2^-$) 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 concentrations (8.7 mg l$^{-1}$) at Kampala (Uganda, Lake Victoria shore, (Visser 1961), 0.09–0.13 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 $\text{NH}_4^+$, $\text{NO}_3^-$ and $\text{NO}_2^-$ 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% (figure 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 (Schlesinger and Hartley 1992, Galy-Lacaux and Modi 1998, Delon et al 2012). Biomass burning and domestic combustions are other principal sources of NH₃ (Delmas et al 1995, Brocard et al 1996, Galy-Lacaux and Modi 1998, Lauwali et al 2012). In Mbita, monthly NH₄⁺ and NO₃⁻ VWM concentrations in rain exhibit maximum values in JJA and in January from 2017 to 2019 (figure 3(a)). 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) (figure 5). Moreover, monthly mean IASI NH₃ and OMI NO₃ Vertical Density Columns from 2017 to May 2019 show the same feature (figure 8). Results clearly indicate that NO₂ are enhanced in DJ and JJA in relation with the biomass burning sources occurrence in the northern and southern hemisphere. NH₃ vertical column density 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 NH₄⁺/NO₃⁻ ratio for N deposition is usually less than 1 in areas with advanced industrialization while areas with intensive agriculture are characteristic of high NH₄⁺/NO₃⁻ ratios (>1) (Fahey et al 1999, Zhao et al 2009). In our study, the NH₄⁺/NO₃⁻ ratio of 4.13 indicates that NH₃ emissions are certainly also related to mixed crops agriculture in Mbita region as well as livestock presence.

3.2. Nitrogen wet deposition budget and dissolved organic nitrogen contribution

Monthly VWM concentrations of DIN and dissolved organic nitrogen (DON) present larger values during the dry seasons (figure 9(a)), especially in JJA 2017, January 2018, JIASO 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 (Zhang et al 2002, Pöschl 2005, Matos et al 2016). 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 (Spitzy 1990, Mace 2003a, 2003b), 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
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.
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-\(\text{NH}_4^+\) | N-\(\text{NO}_3^-\) | N-\(\text{NO}_2^-\) | 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    |        |        | Vuái et al (2013)            |
| Tanzania side               | 1050    | Peri urban          |                      |                      |                      | 6.8    |        |        |                              |
| Tanzania side               | 1050    | Urban               |                      |                      |                      | 6.4    |        |        |                              |
| Lake Victoria               |         |                     |                      |                      |                      |        |        |        |                              |
| Mbíta (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      |                     |                      |                      |                      |        |        |        | Namugize (2015)              |
| La Selva (Costa Rica)       | 3500–4500 | Agricultural land | 0.27                 | 3.74                 |                      | 4.01   |        |        | Eklund et al (1997)          |
| West and Central Africa     |         |                     |                      |                      |                      |        |        |        |                              |
| Sahelian dry savannas       | 449–744 |                     |                      |                      |                      |        |        |        | Galy-Lacaux and Delon (2014) |
| Wet savannas                | 1261–1274 | Rural sites         |                      |                      |                      | 3.5–5.3 |        |        |                              |
| Forest                      | 1557    |                     |                      |                      |                      | 3.6    |        |        |                              |

(Continued)
| Site                          | Pi (mm) | Characteristics | N-$\text{NH}_4^+$ | N-$\text{NO}_3^-$ | N-$\text{NO}_2^-$ | DIN   | DON | TDN | References              |
|------------------------------|---------|-----------------|-------------------|-------------------|-------------------|-------|-----|-----|--------------------------|
| South Africa                 |         |                 |                   |                   |                   |       |     |     |                          |
| Amersfoort                   | 729.75  |                 |                   |                   |                   | 6.32  |     |     | Conradie et al (2016)    |
| Vaal Triangle                | 956.43  |                 |                   |                   |                   | 6.97  |     |     |                          |
| Louis Trichardt              | 728.2   | Semi-arid       |                   |                   |                   | 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              |                   |                   |                   |       |     |     | Jordan et al (1980)      |
| Congo forest (RDC)           |         | Tropical forest |                   |                   |                   | 5.46  | 12.74| 18.20| Bauters et al (2018)     |
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 2011, 2012, Matos et al 2016, Song et al 2017).

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 (figure 9(b)). 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$$^+$-N WD fluxes, except during the special period around July 2017 where DON contribution exceeds DIN contribution, due to the high unexpected marine influence. Maximum DIN WD fluxes (≥1 kgN ha$^{-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$^{-1}$ (tables 2 and 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 (Langenberg et al 2003, Gao et al 2018), 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; Ekland 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).

Net 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 T N yr$^{-1}$, 20, 000 T N yr$^{-1}$ and 44, 032 T N yr$^{-1}$ respectively. Taking into account the total N WD flux from this study, including DON estimation, Lake Victoria receives 58, 760 T N yr$^{-1}$. According to Zhou et al (2014), Nile river and fishery N export from Lake Victoria represent 40, 000 T N yr$^{-1}$ and 4, 000 T N yr$^{-1}$ respectively, leading to 14, 400 T N yr$^{-1}$ staying in the lake and contributing to eutrophication.

4. 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. WD 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 WD 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 combustion 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 WD 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 t N 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.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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