Research article

Trends in rainwater ions (Na^+, K^+, Cl^-) under the influence of the West African Monsoon in the wet savanna of Lamto, Côte d'Ivoire

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Abstract

Atmospheric chemical composition is strongly influenced by natural and anthropogenic emission sources, which may vary with season. In this study, we investigated the trends in the impact of the West African Monsoon (WAM) on the variability of sodium (Na^+), potassium (K^+), and chloride (Cl^-) ions in rainfall over the humid savanna of Lamto in Côte d'Ivoire. Over the 22 years (1994-2015) of the study period, a combination of in-situ rainfall measurements and satellite-based Climate Hazards Group InfraRed Precipitation with Station (CHIRPS) precipitation data was used to understand the rainfall patterns associated with the WAM on the concentrations of these ions. Non-parametric statistical analyses were also performed. Results revealed a significant increasing trend in all three ion concentrations at the event scale, suggesting that in addition to the natural sources such as the Atlantic Ocean, the Sahel and Saharan desert, other anthropogenic factors such as road construction and urbanisation may influence presence of these ions in the rainwater. Moreover, the strong positive correlations between Na^+ and Cl^- suggest a common marine source transported by the WAM flow still pre-dominates in Lamto. However, the increasing trend in K^+, which is a good tracer of biomass combustion, highlights that Lamto is also influenced by local surrounding anthropogenic sources such as biomass burning. An improved assessment of the impact of such local sources will be needed for efficient mitigation strategies in the area.

Keywords

Lamto, atmospheric chemical composition, rainfall, trend

Introduction

Global atmospheric chemical composition is continuously influenced by both natural processes (e.g. volcanic emissions, desert dust, lightning, or particles emitted by solar activity) and anthropogenic activities (e.g. traffic, industries, biomass burning, domestic fires). These processes emit various gaseous and particulate pollutants. These different atmospheric compounds (gases and particles) have an impact on climate, ecosystems and human health. The global climate is strongly influenced by existing complex interactions between atmospheric chemical compounds through emissions, processing mechanisms and atmospheric lifetimes (Pöschl and Shiraiwa, 2015). Atmospheric aerosols have direct and indirect impacts on climate (Giorgi et al., 2003; Levy et al., 2013), which can lead to changes in meteorology such as precipitation and temperature (Konare et al., 2008; N'Datchoh et al., 2018). In addition, depending on the prevailing climate and meteorological conditions at their sources, the emitted chemical compound may undergo several physico-chemical processes (Ning and Sioutas, 2010). Also, various gaseous compounds such as volatile organic compounds (VOCs), sulphur dioxide (SO_2), nitrogen oxides (NO_x) and many others emitted into the atmosphere undergo chemical transformations, mainly oxidation reactions, leading to the formation of secondary pollutants (e.g. ozone, radical species ($HO_x^{\cdot}$, $RO_x^{\cdot}$)), which include more stable oxidized species (Sillman, 1999; Camredon et al., 2007; Adon, 2010; Ossohou et al., 2020).

Moreover, the physico-chemical composition of the atmosphere for a given location can be influenced by local and remote sources. In the West Africa region, rapid socio-economic changes occur with various impacts on the environment, landscapes and human health (Ofoezie et al., 2022). These rapid changes lead to significant increases in anthropogenic emissions (Liousse et al., 2014; Keita et al., 2021; Doumbia et al., 2021), in addition to the existing natural emissions sources. These emissions greatly impact the chemical composition of the atmosphere at local and regional scales. In West Africa, previous work reported the existence of local (coastal areas) and long-range (central and southern Africa) transport of pollutants (Knippertz et al., 2015a; Brito et al., 2018; Flamant et al., 2018; Dajuma et al., 2020) in many remote places during the West African Monsoon (WAM). The WAM is a complex climate system that dominates the weather patterns and rainfall occurrence over West Africa region from roughly March to September by transporting moisture from the Atlantic Ocean to the land (Sultan et al., 2005; Omotosho and Abiodum, 2007; Fink et al., 2017). The WAM is governed by the annual shift of the intertropical discontinuity (ITD) following a south-north gradient, which lies around 8°N during January (dry season) and 20°N in around July (monsoon period). The ITD separates the south-westerly winds that bring moisture from the Atlantic Ocean onto the continent from the north-easterly Harmattan winds that transport dust aerosols from the Sahel and Sahara regions (Engelstaedter et al., 2006; Touré et al., 2012) as well as aerosols from biomass burning in the Savannah regions from November to February (N'Datchoh et al., 2015). Therefore, the WAM plays a crucial role in the region's agriculture, food security, and ecosystems and also in the region's atmospheric chemical composition.

Dry and wet deposition remain the main physical processes responsible for the removal of gases and aerosols from the atmosphere (Ryu and Min, 2022). Whereas dry deposition generally occurs at the surface of vegetation or soil, wet deposition consists of the absorption or interception of gases and aerosols by precipitation. The predominance of dry or wet deposition depends strongly on the geographical location of the site, the intensity of precipitation, as well as the physical aspect (size, hygroscopicity) and chemical properties of the atmospheric species (Deboudt et al., 2008). The International Network to study Deposition and Atmospheric chemistry in Africa (INDAAF) was established in 1995 with the support of the International Global Atmospheric Chemistry (IGAC) and focuses on the long-term monitoring of atmospheric chemical compounds and deposition fluxes, especially related to atmospheric sulphur and nitrogen, to investigate and better understand chemical processes in the African ecosystems (Ossohou et al., 2019; Adon et al., 2010; Galy-Lacaux et al., 2009; Galy-Lacaux and Delon, 2014). This INDAAF network contributed to the better understanding of the dynamics of tropical atmospheric chemical composition changes induced by human activities (Viers et al., 2001; Ossohou et al., 2020) over the western and central regions of Africa. Using rainfall chemistry observation of the INDAAF network, Galy-Lacaux et al. (2009) investigated the composition of rainfall samples from June 1994 to September 2005 in the Banizoumbou (Niger, dry savanna site). They highlighted that the rainwater chemistry was controlled by soil/dust emissions associated with terrigenous elements represented by SO_4^{2-}, Ca^{2+}, carbonates, K^+ and Mg^{2+}. Moreover, over Zoetele, an INDAAF site located in the southern Cameroon, between 1996 and 2000 the rainwater chemistry composition was reported to be acid with predominantly nitrogenous compounds (NH_4^+ and NO_3^-), Ca^{2+} and SO_4^{2-} (Sigha-Nkamdjou et al., 2003). They also reported very low Na^+ and Cl^- concentrations, which accounted for the marine source. When characterizing the proportion of water-soluble ions (organic and inorganic) in the particle matter ($PM_{2.5}$ and PM_{10}) at the INDAAF sites in Banizoumbou (Niger) and Djougou (Benin, wet savanna site) from 2005 to 2007 and 2005 to 2009, respectively, Moussa et al. (2018) reported that concentrations of Ca^{2+}, NO_3^- and SO_4^{2-} were higher during the dry season than the wet season in Banizoumbou. The most dominant ions in Djougou were Ca^{2+}, NO_3^- and SO_4^{2-} with a predominance of their concentration during the dry season compared to the wet season. They also highlighted that the NH_4^+ concentrations were not negligible during the wet season at both sites. Their results highlighted a seasonal difference in the PM loading in the atmosphere, with higher concentrations in the dry savanna than in the wet savanna (Moussa et al., 2018). At the wet savanna ecosystem' site of Lamto (Côte d'Ivoire), the analysis of inorganic (Ca^{2+}, Mg^{2+}, Na^+, K^+, NH_4^+, Cl^-, SO_4^{2-}, NO_3^-) and organic ($HCOO^-$, CH_3COO^-) ions in rainwater between 1995 and 2002 underlined that the precipitation chemistry was influenced by four main sources: natural biogenic emissions from savanna soils (NO_x and NH_3), biomass burning (savanna and domestic fires), terrigenous particles emissions from dry savanna soils, and marine compounds embedded in the WAM (Yoboué et al., 2005; Touré et al., 2016; Kassamba-Diaby et al., 2023).

Recent studies on long-term trend of chemical components in rainwater in Lamto (Ossohou et al., 2019; 2020) do not include the water-soluble ions sodium (Na^+), potassium (K^+), and chloride (Cl^-) ions in their analysis. Considering that many external factors (e.g. distance from the coast, rainfall pattern, local and long-range transport, local emissions, seasonal variability) may influence the concentrations of these water-soluble ions, until now, their trends in concentration in the rainwater at Lamto have not yet been investigated. The water-soluble ions, Na^+ and Cl^-, are oceanic in origin and can be used to track changes in the WAM flow. The K^+ ion may highlight changes related to anthropogenic activities. As the long-term trends in rainfall, influenced by climate change, can indirectly impact these ion concentrations through the rainfall patterns as well as the relative importance of different sources and atmospheric processes, it appears important to document the recent trends of these ions in the collected Lamto rainwater. Therefore, this work aims to investigate trends in the impact of WAM contribution to Na^+, Cl^- and K^+ variability over Lamto.

Methodology

Study area: Lamto station

The climate in Côte d'Ivoire is influenced by the dynamics of the West African Monsoon (WAM), which leads to the occurrence of distinct dry and rainy seasons (Kouadio et al., 2015). Côte d'Ivoire has three climatic zones along a south-north gradient (Kouadio et al., 2007, 2011, 2015; N'Datchoh et al., 2022). The southern and central climatic zones close to the Guinean coast are characterised by two wet seasons. The Lamto station is in a reserve (06°13' N, 05°02' W) located in the central part of the country within the V-Baoulé region (about 120 km north of Abidjan, the economic capital). The reserve lies in the transition zone between the savanna and tropical forest, and covers an area of about 2,617 hectares (Figure 1). The Lamto station serves as an observation site for seismology, geophysics, climatology and ecology. This site serves as a reference for a wet savanna ecosystem in the International Network to study Deposition and Atmospheric chemistry in AFrica (INDAAF) network.

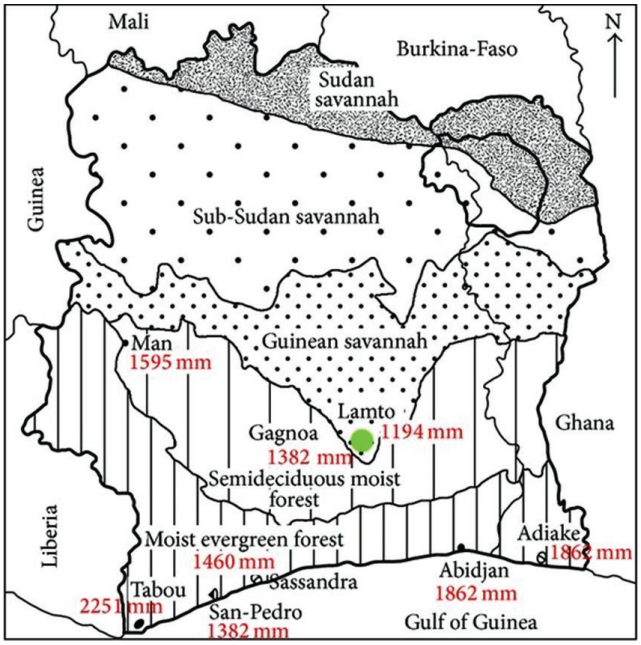

Figure 1: *Location of the Lamto station represented by the green dot (Diawara et al., 2014)*

Different Guinean savanna facies can be distinguished at Lamto, including wooded savannas, shrub savannas, grassy savannas, and gallery forests. The open and moderately open shrubby facies occupy three-quarters of the Andropogoneae shrub savanna zone, which comprises nearly half of the savannas in the Lamto reserve (Le Roux et al., 1995, 1997). The herbaceous stratum is composed of perennial grasses (C4 plants) reaching nearly 2 m in height at flowering. The passage of wildfires, generally in mid-January, destroys the epigenetic phytomass. The regrowth of grasses and the budding of shrubs take place in the following month.

Lamto station has a warm and wet transitional climate under the influence of the WAM which determines its seasons. Lamto has a mean annual rainfall of 1,240.9 ± 133.5 mm (Ossohou et al., 2020). The climate in Lamto is divided into four main seasons: a long dry season from early November to February, a long wet season from March to July, a short dry season in August, and a short wet season from September to November (Srikanthasamy et al., 2021). Each year, the site is affected by the Harmattan in the middle of the dry season. Relative humidity in Lamto remains high for most of the year, with a maximum of 82.8% in June and a minimum of 65.6% in January. Air temperatures are relatively constant throughout the year (27°C on average). The Leaf Area Index (LAI) generally follows a similar pattern to rainfall, with high values from April to June and from August to November (Ossohou et al., 2020). The composition of the atmosphere at Lamto is mainly influenced by biogenic emissions from the wet savanna and other emissions at the local scale (such as seasonal biomass burning, domestic fires, surrounding anthropogenic sources) and regional scale (such as seasonal Sahel and Saharan dust transported by the Harmattan flows, and the seasonal long-range transport of pollutants from the Atlantic Ocean, central and southern hemisphere of the continent by the WAM flows). Local disturbances in atmospheric composition are mostly of anthropogenic origin, originating from coffee and cocoa plantations, savanna fires and domestic fires from small surrounding villages (Yoboué et al., 2005).

CHIRPS reanalyses

CHIRPS (Climate Hazards Group InfraRed Precipitation with Station data) comprises a near-global daily rainfall satellite dataset of over 30 years. CHIRPS measurements, spanning the region from 50°S to 50°N and all longitudes from 1981 to the present, integrate satellite imagery at 0.05° resolution with in situ station data to create gridded precipitation time series for trend analysis and seasonal drought monitoring. This dataset has been used in numerous studies for effective hydrological forecasting and trend analysis (Funk et al., 2015; Diba et al., 2022; Ndeye et al., 2021), as well as for the validation of Cordex-Africa regional climate model outputs (Mahamadou et al., 2018). Working in a region where ground-based observations are still sparse and considering the recent rapid spread of low-cost sensors, we attempted to evaluate Lamto rainfall observed data with products such as CHIRPS data. This gives an idea of how these reanalysis data, which have been used in regional and country scale studies, can be used for air quality studies when in-situ measurements are not available. To achieve this, daily CHIRPS data between 1994 and 2015 were compared to rainfall measurements collected at the Lamto station.

INDAAF rainfall collector

Since 1994, rainwater has been sampled at stations in the INDAAF network to determine the chemical composition (mineral and water-soluble organic acids) of each rainfall event. For the period 1995–2002, Yoboué et al. (2005) conducted the first study on the chemical composition of rainfall at the Lamto station. This study includes the study period of Yoboué et al. (2005), as well as 1994 and the period from 2003 to 2015, which is 22 years of data on the chemical content of rainfall at Lamto. The Lamto station is equipped with a semi-automatic rain collector

specially designed for the INDAAF network. A local operator collects water from each rain event in a Greiner tube (50 ml). The rain samples are stored immediately after collection in a freezer at -18°C. The instrumentation and collection protocol follow the recommendations of the World Meteorological Organization (WMO) (report No. 160). International intercomparison studies are carried out twice a year to ensure the quality of the analysis. The Laboratoire d'Aérologie (LAERO) has been involved in the WMO-GAW (Global Atmosphere Watch) Quality Assurance Quality Control intercomparison programme since 1996 under reference 700106. Based on these tests, the analytical accuracy of ion chromatography, the technique used at the LAERO to analyse the chemical content of rainfall, is estimated to be 5% or better for all ions, which is within the uncertainties of all values measured in this study. The experimental protocol for sampling rainfall with the INDAAF semi-automatic collector has been extensively described in previous studies (Galy-Lacaux and Modi, 1998; Sigha-Nkamdjou et al., 2003; Yoboué et al., 2005; Galy-Lacaux et al., 2009; Laouali et al., 2012; Akpo et al., 2015). Rain samples, analysed by ion chromatography at the LAERO, were used to determine the ion concentrations. In this study, we focused only on the chloride (Cl^-), potassium (K^+) and sodium (Na^+) ions from 1994 to 2015, which have received less attention in recent studies and are strongly related to the WAM dynamic and are a good proxy to monitor potential changes in its chemical composition. During this period, 1,787 rain samples were collected and analyzed at the Lamto station.

Statistical analysis

This work uses the non-parametric Mann-Kendall (MK) and Seasonal Mann-Kendall (SMK) statistical trend tests (Mann, 1945; Kendall, 1979; Kendall and Kriebel, 1982). These tests were applied to ion concentrations of Cl^-, K^+ and Na^+ in rainwater samples collected as well as at the rainfall amount at Lamto to determine long-term trends. The MK test is a statistical method that operates under three key assumptions. Firstly, it presumes that any given value can be consistently categorised as less than, greater than, or equal to another value. Secondly, the test assumes that all data points are independent of one another. Finally, it requires that the data distribution remains unchanged, whether in its original form or after undergoing a transformation. These assumptions are essential for the proper application and interpretation of the MK test results, ensuring the validity of the conclusions drawn from the analysis. The SMK follows the same general principle as that of the MK test. The SMK test statistic is calculated by applying the MK test to each month separately and aggregating the monthly results. Over the entire time series, only observations from the same month are compared (e.g., January to January), then the results are combined. The test does not make comparisons across different months, ensuring that the analysis remains within the defined seasonal boundaries. The MK test was used to investigate trends in seasonal and annual data, while the SMK test was applied to monthly data. The SMK and MK tests were computed at the monthly, seasonal and annual scales as described in Ossohou et al. (2023). The magnitude of the trends in both MK and SMK were estimated using Sen's Slope (SS) (Sen, 1968) and the Seasonal Kendall Slope Estimator (SSE), respectively. The SS and SSE are valuable tools for detecting and quantifying trends in time series data, particularly in environmental studies where data often deviates from normality and can be influenced by seasonality (Aditya et al., 2021). Moreover, these statistical methods for detecting and quantifying trends are robust, little affected by errors within the data, and not sensitive to outliers (Sicard et al., 2011). In this work, the significance level was set at 95% with p-value < 0.05 and all the analyses were performed with the Python software.

Results and discussion

Comparison of CHIRPS data with observations from Lamto station

A comparison of the monthly and average annual rainfall from CHIRPS and in-situ measurements is presented in Figures 2 and 3, respectively. Rainfall measured at Lamto shows a minimum annual rainfall of 726 mm (2013) and a maximum of 1,549 mm (2007), with a mean of 1,198 ± 201 mm. It shows a low variability, with a coefficient of variation of 16.8% over the 22-year period. At the same timescale, CHIRPS data records a higher minimum rainfall of 1058 mm (1997) and a maximum of 1710 mm (2010), resulting in a mean of 1364 ± 166 mm. The coefficient of variation of 12.2% indicates less variability and suggests more stable rainfall estimates. Monthly rainfall determined with CHIRPS is, in general, higher than rainfall measured at Lamto (Figure 2). Overall, both time series have a similar pattern. The bimodal character of the annual rainfall cycle in Lamto is well highlighted in the two datasets (Figure 3). CHIRPS tends to overestimate rainfall amounts during the main rainfall season (May and June) associated with ITD movement as well as during the WAM retreat from land (September and October). This overestimated rainfall amount in the CHIRPS data seems to dissipate during the main dry seasons (from December to March). CHIRPS rainfall is becoming much closer to in-situ recorded observations. In West Africa, several studies (Quagraine et al., 2020; Atiah et al., 2020; Amekudzi et al., 2016) highlight the good performance of satellite rain-gauge bases data and reanalysis data to study WAM, as the region has a very limited spatial coverage for rainfall in-situ measurement sites. Despite their wide use, these products need to be validated when in-situ data are available because their calibrations are necessary to reduce their inherent biases and uncertainties (Macharia et al., 2022; Mekonnen et al., 2023).

Pearson correlation between CHIRPS and in-situ data (Figure 4) is estimated to be 0.75 with p-value < 0.01, suggesting a good correlation between the two datasets over Lamto. The current finding between CHIRPS and in-situ observations in Lamto agrees with previous work conducted by Sacré et al. (2020) using the rain-gauge data from the BAse de DOnnées PLUviométriques (BADADOPLUS) database for five cities (Dakar (Senegal), Ouagadougou (Burkina Faso), Niamey (Niger), Adiaké (Côte d'Ivoire) and Cotonou (Benin)) and CHIRPS during 1998 to

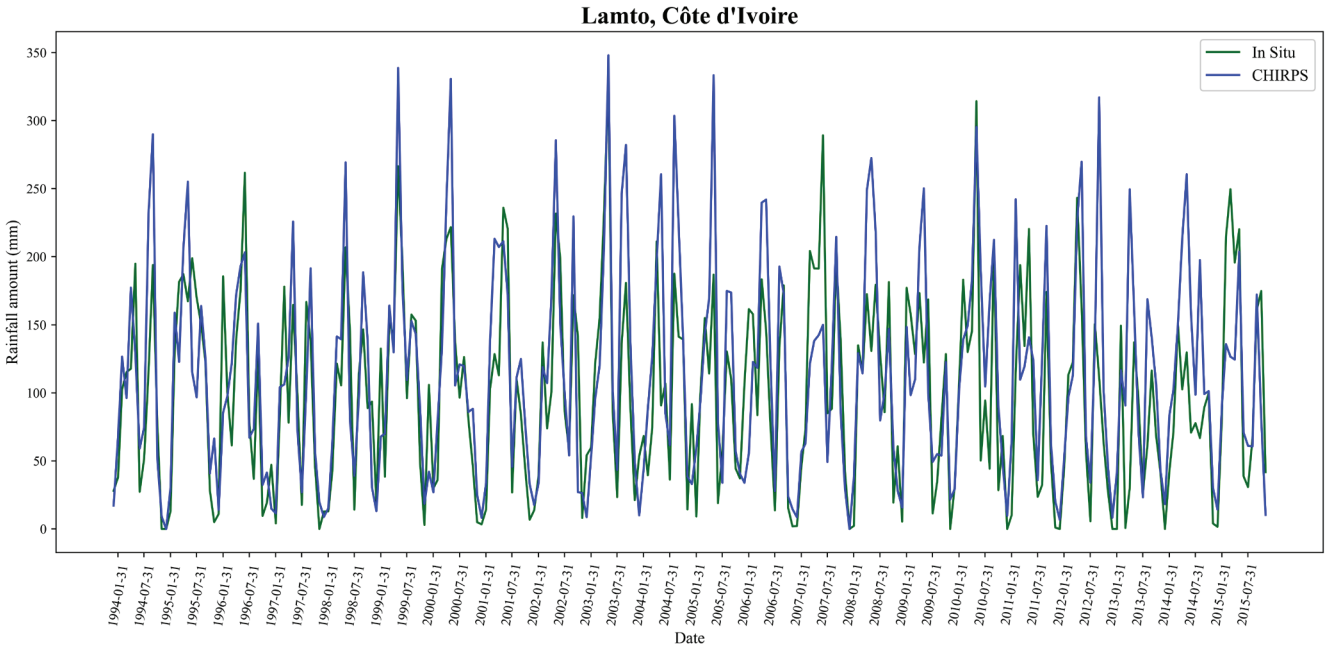

Figure 2: *Monthly variations of rainfall amount from in situ and CHIRPS measurements at Lamto, Côte d'Ivoire (1994-2015)*

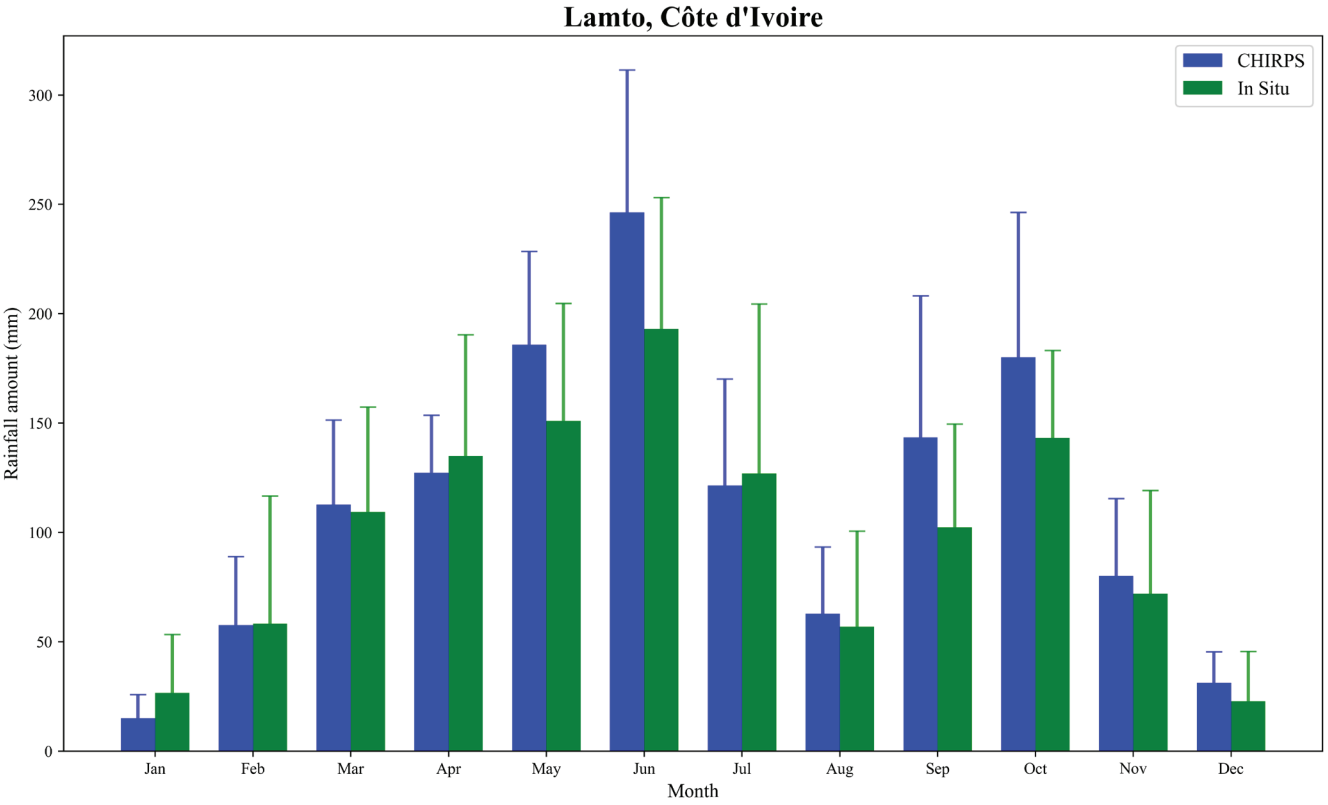

Figure 3: *Mean annual cycle of monthly rainfall amount from in situ and CHIRPS measurements at Lamto station, Côte d'Ivoire (1994-2015)*

2010. They underline that CHIRPS data were highly correlated (correlation coefficient higher than 0.8). Using a longer period (1994-2020), we found that CHIRPS data reproduce patterns of observed rainfall in Lamto with a significantly good correlation. Therefore, in cases where there are limited in-situ observations of rainfall, satellite rainfall such as CHIRPS can be valuably, but carefully, used for air quality and modelling studies despite some biases.

Rainfall chemistry in Lamto

In Figure 5, we present variations in sodium (Na^+), potassium (K^+) and chloride (Cl^-) ions measured for each rainfall event in the

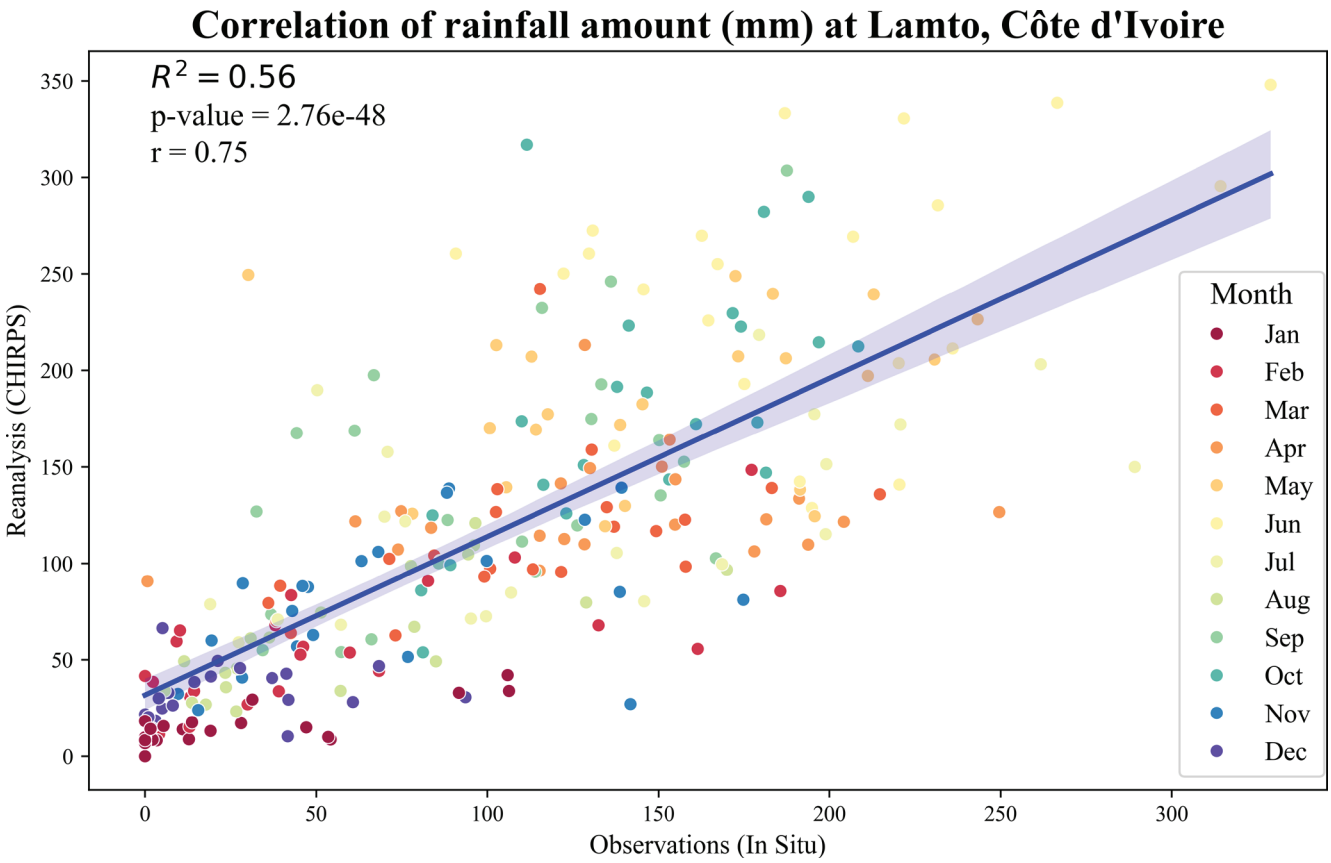

Figure 4: *Correlation month by month between in situ and CHIRPS measurements at Lamto, Côte d'Ivoire.*

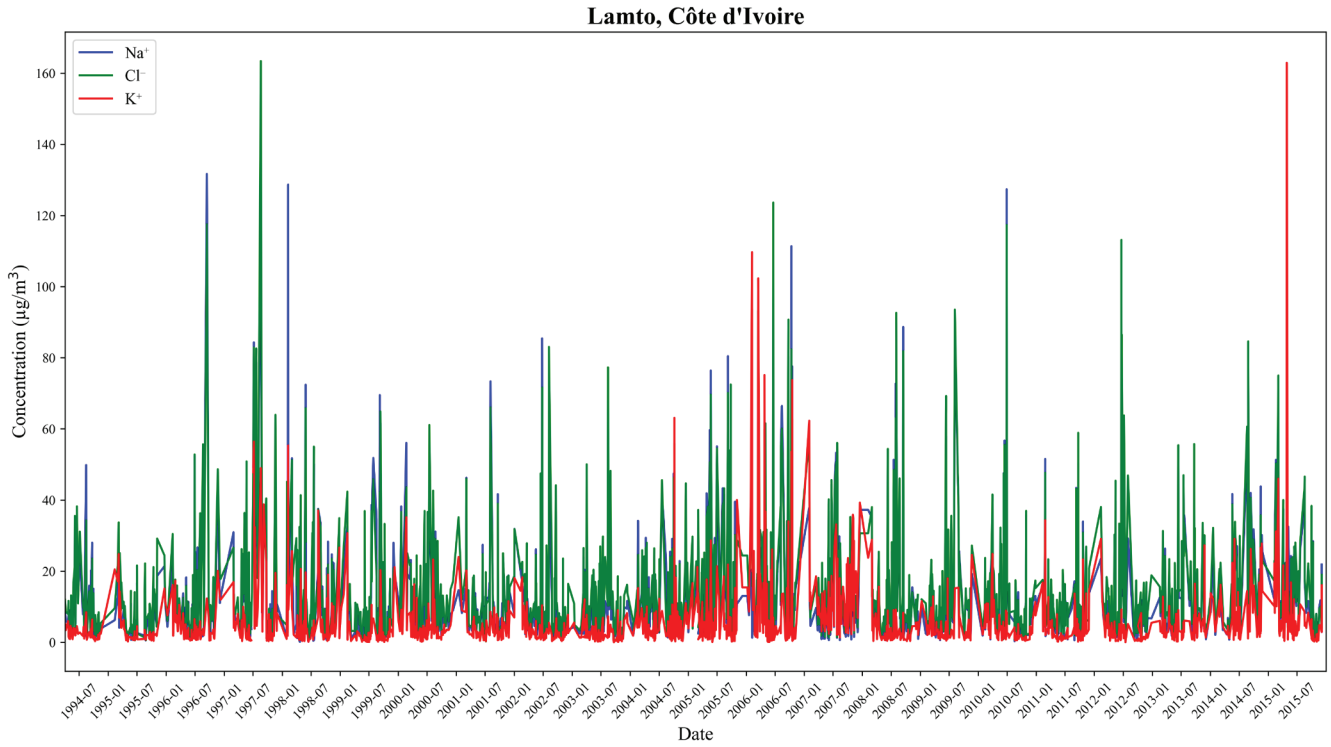

Figure 5: *Time series of sodium (Na^+), potassium (K^+) and chloride (Cl^-) ions concentration contained in rainfall events at Lamto between 1994 and 2015.*

wet savanna of Lamto from 1994 to 2015. Note that rain sample depths collected during each rainfall event are generally greater than the actual rainfall amounts.

The statistical analysis for Na^+, K^+ and Cl^- concentrations contained in the rainwater collected at Lamto between 1994 and 2015 is summarised in Table 1. The high variability (wide ranges

and high peaks) in Na^+, K^+ and Cl^- concentrations underlines the influence of specific events (local and/or remote) that affect the chemical composition of Lamto. The influence of these events on Na^+, K^+ and Cl^- concentrations implies the presence of external drivers in addition to the known natural drivers.

Table 1: *Statistics of Na^+, K^+ and Cl^- in rainwater at Lamto from 1994 to 2015*

	Na^+	K^+	Cl^-
Total number of data	1,783	1,783	1,783
Percentage of missing values	0.17	0.17	0.17
Mean concentration ± Mean standard deviation (µg/m³)	11.74±14.79	5.27±8.70	13.42±15.11
Minimum (µg/m³)	0.13	0.03	0.21
1st quartile (µg/m³)	3.26	1.25	4.29
Median (µg/m³)	6.92	2.71	8.60
3rd quartile (µg/m³)	13.92	5.90	16.89
Maximum (µg/m³)	139.28	162.97	163.44

We use the concentrations in Figure 5 to plot mean monthly variations of atmospheric concentrations of Na^+, K^+, and Cl^- ions, along with the rainfall climatology (in-situ and CHIRPS), in the wet savanna of Lamto (Figure 6).

To understand the high variability in these ion concentrations and its relationship with WAM, it appears important to investigate the annual and seasonal patterns. The climatology (Figure 5) and mean annual cycle (Figure 6) of these three ions were calculated and compared to the monthly and average rainfall. At the seasonal scale, Na^+, Cl^- and K^+ concentrations exhibit a bimodal pattern similar to rainfall, although their peaks do not occur within the same seasons. The ion concentrations peak during the dry seasons (February and August), while rainfall peaks in June and October.

Table 2: *Trend statistics (z, τ and SS) from the MK test coupled to SS, applied to rainfall amounts (Ht), Na^+, K^+ and Cl^-. The trends were evaluated over the 1994-2015 period.*

Time scale	Statistical and trend indicators	Ht	Na^+	K^+	Cl^-
Event	z	1.15	2.57	4.06	2.95
	τ	0.007	0.041	0.064	0.047
	SS	0.00000 ns	0.00068*	0.00042*	0.00093*
Annual	z	-0.45	0.28	0.00	0.39
	τ	-0.074	0.048	-0.004	0.065
	SS	-5.38333 ns	0.01093 ns	-0.00308 ns	0.09480 ns

**significant at the 95% confidence level ($p < 0.05$); ns: non-significant ($p > 0.05$).*

The Na^+, K^+, and Cl^- concentrations' climatology in Lamto is characterised by an increase during January and February. Na^+ concentration increases from approximately 10.76 ± 4.90 µg/m³ in January to a peak of around 16.18 ± 8.54 µg/m³ in February. K^+ follows a similar pattern, rising from about 10.01 ± 4.92 µg/m³ in January to 13.23 ± 6.19 µg/m³ in February. Cl^- shows the highest concentration among the three ions, increasing from around 15.89 ± 8.00 µg/m³ in January to 16.68 ± 6.18 µg/m³ in February. During this period, biomass burning activities occur in the region (N'Datchoh et al., 2015) and monthly rainfall remains relatively low (41 and 58 mm). The Lamto savanna is set on fire to preserve its ecosystem. These combustion sources result

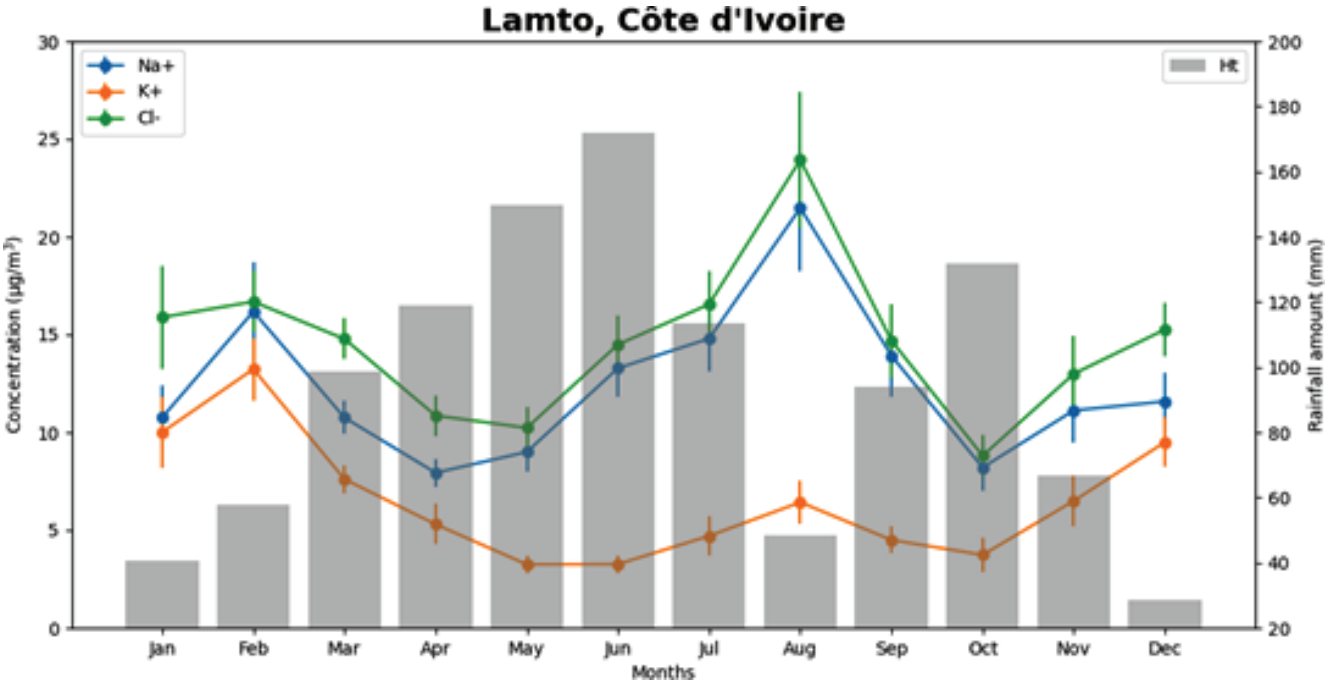

Figure 6: *Mean monthly variations of Na+, K+ and Cl− over the period 1994-2015 at Lamto, Côte d'Ivoire*

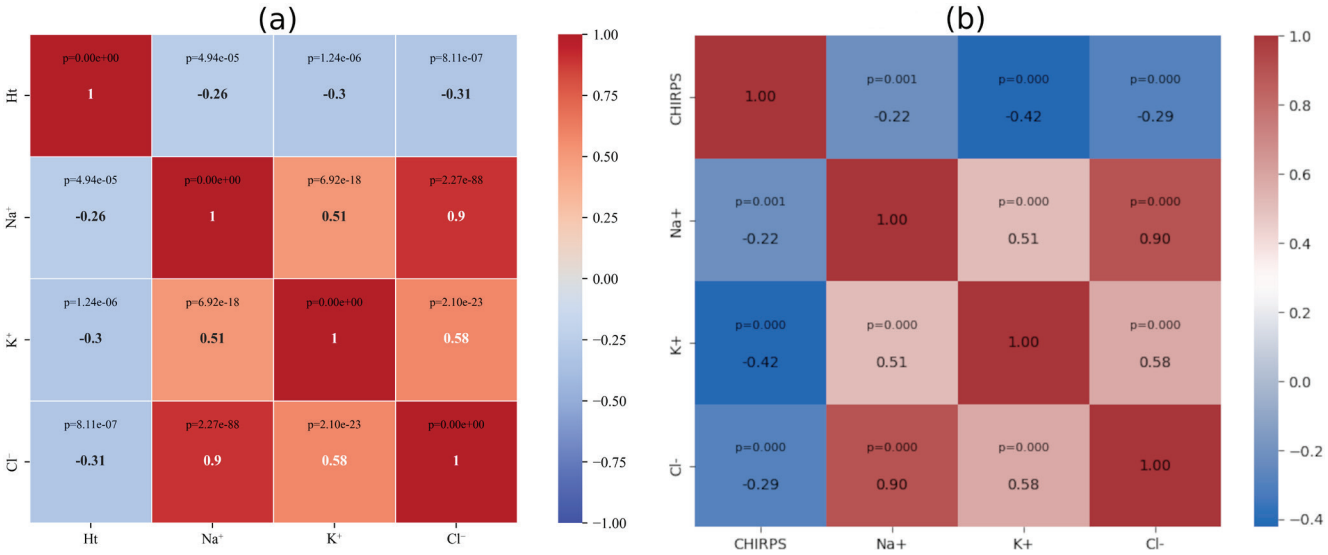

Figure 7: *Heatmaps of Pearson correlation coefficients between Na^+, Cl^-, and K^+ ion concentrations in rainwater and (a) rainfall amount collected and (b) monthly CHIRPS rainfall at the Lamto station, Côte d'Ivoire*

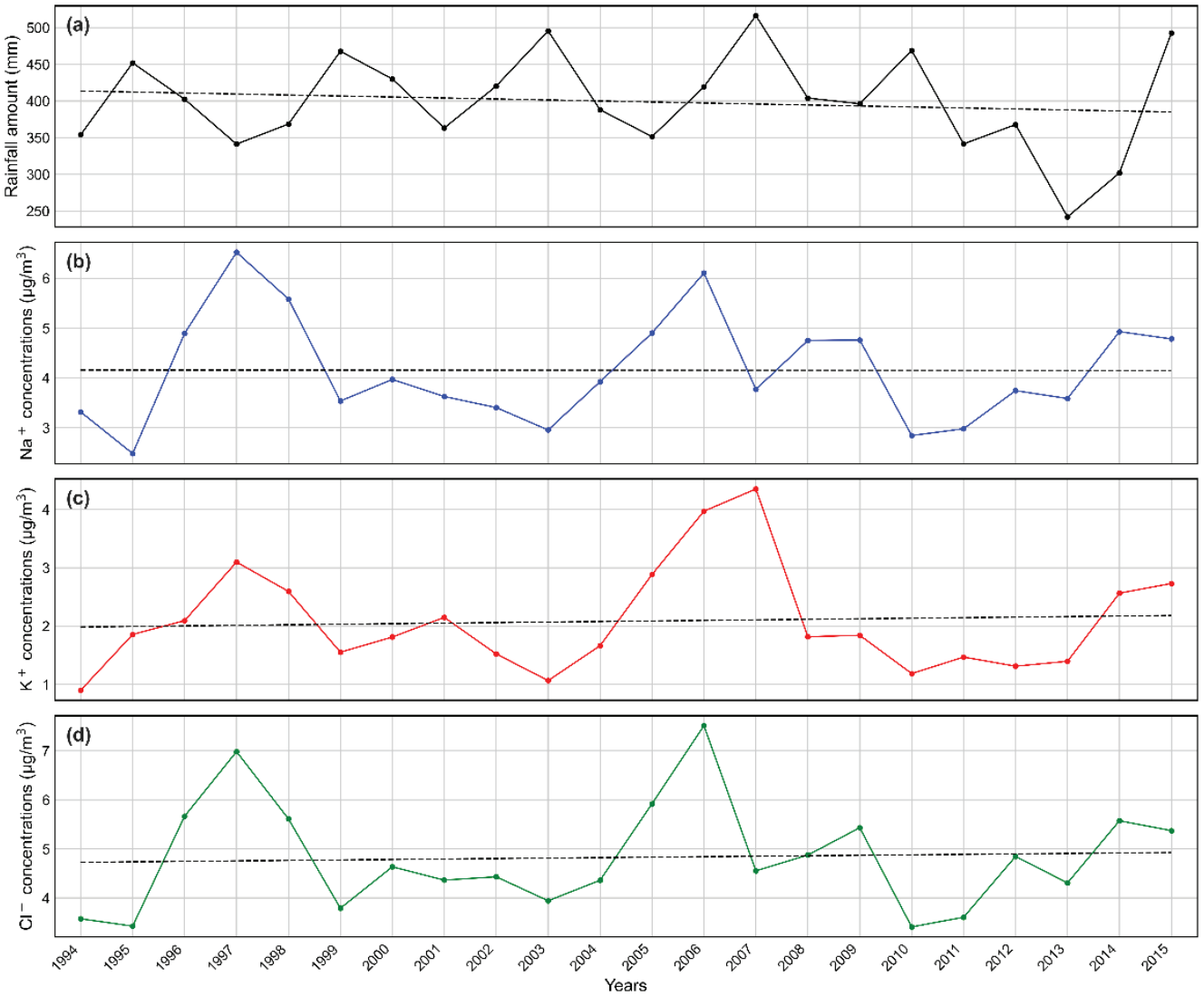

Figure 8: *Interannual (a) rainfall amount, (b) Na^+, (c) K^+, (d) Cl^- concentrations in rainwater, and linear trends from 1994 to 2015 at Lamto, Côte d'Ivoire.*

in the release of compounds such as KCl and gases into the atmosphere. The irregular rains of the dry season leach these compounds, resulting in peak concentrations of K^+ and Cl ions in rainwater during February (Naidin et al., 2024; Sun et al., 2019).

From February to May (April for Na^+), Cl^- and K^+ concentrations decrease to approximately 10.23 ± 3.82 µg/m³ and 3.25 ± 1.59 µg/m³, respectively. Na^+ also shows a decline to around 7.93 ± 2.54 µg/m³ by April. This general decrease in ion concentrations from March onwards coincides with rainfall increasing as the WAM settles over the West African region. This suggests that the rainy season's onset likely contributes to local atmosphere washout by removing these ions through wet deposition processes.

From May to August (for Cl^- and K^+ and April to August for April for Na^+), there is a sharp increase in ion concentrations. Na^+ concentration rises from about 8 µg/m³ in April to 21.48 ± 11.42 µg/m³ in August. While Cl^- and K^+ also recorded an increasing concentration from to May to August (Cl^-: 10 µg/m³ to 23.93 ± 12.07 µg/m³; and K^+: 3 µg/m³ to 6.43 ± 3.64 µg/m³). This period corresponds to the peak of the main rainy season in June (about 172 mm). The second peak in ion concentrations occurs during the little dry season (August) when the monsoon jumps to its northward position and southerly winds prevail over the entire region (Sultan and Janicot, 2003). This continuous increase in ion concentrations despite high rainfall suggests a possible influx of these ions from other sources, such as atmospheric transport or local emissions, which outweighs the removal by wet deposition.

From August to October, as the WAM retreats from the region with the ITCZ moving its position southward, the concentrations of Na^+ (21 µg/m³ to 8.20 ± 3.38 µg/m³), K^+ (24 µg/m³ to 8.81 ± 2.97 µg/m³), and Cl^- (6 µg/m³ to 3.73 ± 2.68 µg/m³) begin to decline. During this period, rainfall increases from 48 mm in August to 132 mm in October. The decline in ion concentrations can be attributed to reduced local emissions and possibly enhanced wet deposition due to increasing rainfall amount.

Finally, from October to December as the main dry season settles with prevalent northeastward wind and rainfall reducing (from 132 mm to 29 mm), the concentrations of Na^+ (8 µg/m³ in October to 11.58± 4.35 µg/m³), K^+ (9 µg/m³ in October to 15.26± 3.95 µg/m³), and Cl^- (4 µg/m³ in October to 9.50± 3.92 µg/m³) show a slight increase. This slight increase in ion concentrations with the occurring of the dry season could be explained by a combination of factors, such as wet deposition efficiency decreasing with rainfall and potential increases in local emission sources.

In Africa, the dry season is often associated with anthropogenic emissions, such as biomass burning in the savanna biome which increases over the West Africa region from November to February (N'Datchoh et al., 2015). Biomass fires release particles containing K^+ and Cl^-. In addition, Lamto and the surrounding areas undergo biomass burning activities from October to December for land preparation for cropping and grazing (Ossohou et al., 2019; Soro et al., 2021). Another characteristic of the dry season in this part of Africa is the prevalent northeasterly wind associated with the Harmattan which results in the long-range transport of Sahelian and Saharan dust (Touré et al., 2012) containing high quantities of K^+ ions (Formenti et al., 2011). Moreover, local dust emissions from unpaved roads surrounding the station, are enhanced during the dry season and may also contribute to enriching these ions in the atmosphere. Thus, particles from a variety of potential sources are washed out by precipitation when it occurs, resulting in the high ion concentrations in the rainfall collected during this period.

In conclusion, the temporal variability in ion concentrations in Lamto is closely linked to seasonal changes in rainfall. High ion concentrations during the dry season suggest limited removal by wet deposition, while decreases in ion concentrations during the rainy season indicate effective removal processes. The interaction between local emissions, atmospheric transport, deposition mechanisms and the WAM appears to govern the observed patterns in ion concentrations throughout the year.

A heatmap (Figure 7) shows the correlation coefficients and the corresponding p-values between the collected rainfall amount (Ht) and the concentrations of Na^+, Cl^-, and K^+ in rainwater (Figure 7a), and the CHIRPS monthly rainfall and the concentrations of these water-soluble ions (Figure 7b) at the Lamto station for all rainfall events from 1994 to 2015. P-values indicate the statistical significance of the correlations. There are statistically significant negative correlations between rainfall amount and the concentrations of Na^+, K^+ and Cl^-. Negative correlation value suggests that as the rainfall amount increases, Na^+, K^+ and Cl^- concentrations tend to decrease. These negative correlations between the collected rainfall amount and ion concentrations could suggest that dilution occurs during higher rainfall events, reducing the concentrations of Na^+, K^+, and Cl^- in the rainwater. The correlation between the CHIRPS rainfall and the concentration of the water-soluble ions of interest has similar patterns. Their concentrations tend to decrease as the WAM precipitation increases, supporting that dilution may occur.

Significant positive correlation (0.51 with a p-value < 0.01) between Na^+ and K^+ ion concentrations are found, indicating that these ions tend to increase together. A stronger significant correlation (0.9 with p-value < 0.01) between Na^+ and Cl^- is observed, indicating that these ion concentrations mostly vary together. There is also a positive correlation between K^+ and Cl^-, with a correlation coefficient of 0.58 (p-value < 0.01), indicating that these ions also tend to vary together, albeit to a lesser extent than sodium and chloride. The strong positive correlation between Na^+ and Cl^- suggests that both ions may come from common sources (sea spray) and/or be affected by similar physicochemical processes. According to Seinfeld and Pandis (1998), most of the atmospheric Na^+ is derived from sea salt and mineral dust, which account for 36% and 42% of global aerosol emissions, respectively. As the Lamto station is located in the tropical region of Africa and under the influence of the WAM (southwesterly wind bringing moisture from the

Atlantic Ocean to the continent during boreal summer) and the Harmattan trade winds (northeasterly wind transporting dust from the Sahel and Sahara), these sources may be one of the important contributors to Na^+ and Cl^- presence in the Lamto atmosphere.

Trends in rainfall chemistry

The MK test coupled to the SS was applied to the daily rainfall amounts recorded at the Lamto meteorological station over the 1994–2015 period. These daily rainfall data were summed to derive monthly and annual rainfall totals. The MK coupled to the SS approach was also applied to the annual rainfall totals. For chemical analyses, only the rainfall volumes effectively collected by the wet-only sampler described in the INDAAF rainfall collector subsection were considered, as they represent the actual water samples from which subsamples were taken for Na^+, K^+, and Cl^- determinations. The same statistical tests were then applied to the event-based and annual ion concentration datasets derived from these collected rain samples. To account for the seasonality in the data, we have applied the SMK test coupled to the SSE for monthly cumulative rainfall over the period 1994-2015. This statistical method was also applied to monthly mean concentrations of Na^+, K^+ and Cl^- ions over the same period, as described in monthly trends subsection. For this study, the significance threshold was set at 95% ($p < 0.05$).

Event-related and annual trends

Interannual variations in rainfall amounts, as well as those in Na^+, K^+, and Cl^- ion concentrations in rainfall, with trend lines, are shown in Figure 8. The MK test applied to rainfall amount per event indicates no significant trend ($p > 0.05$). The corresponding z, τ, and SS values (Table 2) consistently point to a very weak and statistically non-significant variation, confirming the absence of any meaningful trend in event-based rainfall amounts at Lamto. This result suggests that, over the observation period, there has been no noticeable increase or decrease in rainfall collected per event.

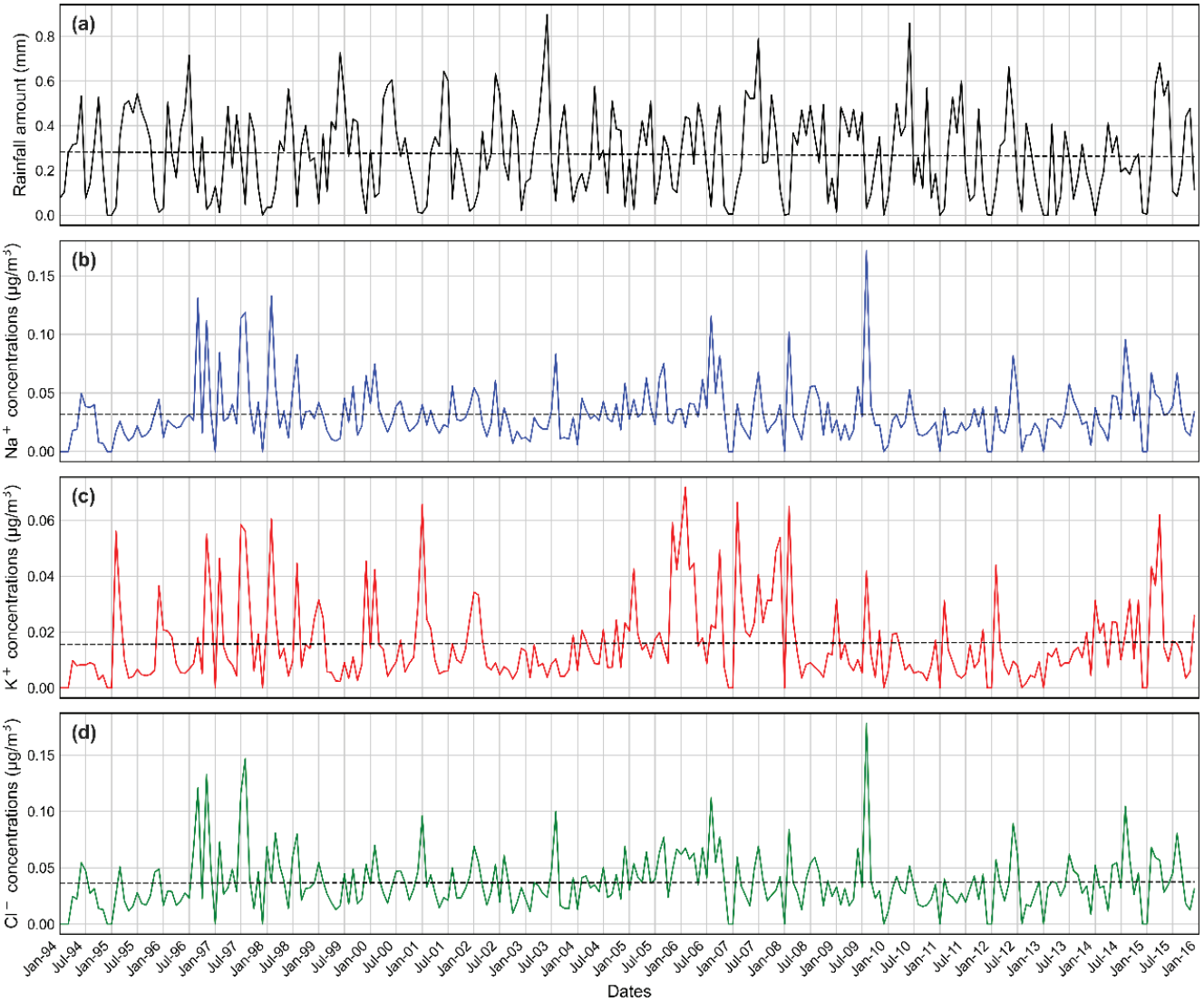

Figure 9: *Monthly timeseries of (a) rainfall amount, (b) Na^+, (c) K^+, (d) Cl^- concentrations in rainwater, and linear trends from 1994 to 2015 at Lamto (Côte d'Ivoire).*

In contrast, the event-based analysis of Na^+, K^+, and Cl^- ions reveals significant and consistent increasing trends for all three species (Table 2). The MK test indicates that these trends are statistically significant, most pronounced for K^+ ($p < 0.001$), followed by Cl^- ($p < 0.01$) and Na^+ ($p < 0.05$). The positive τ values and non-zero Sen's slope estimates confirm the gradual increase in their concentrations in rainwater over the 22-year study period, suggesting progressive changes in the atmospheric composition at Lamto station.

Na^+ and Cl^- are often associated with sea salt aerosols, suggesting increased marine aerosol transport to the region, while K^+ is often linked to biomass burning and agricultural activities. The environment of Lamto, a scientific reserve in Côte d'Ivoire, has changed significantly over the decades. These changes are linked to human activities and land modification. Several factors, such as infrastructure construction, agricultural expansion and industrial activities, have accelerated these transformations. In 2013, the highway connecting Abidjan to Yamoussoukro was completed. This infrastructure has facilitated access to the region. It has also intensified human activities, in particular transport and trade. These activities have increased emissions of atmospheric pollutants. Since the 2000s, the introduction of industrial crops such as rubber and oil palm has significantly altered the land around Lamto. These crops provide a strong agricultural demand. They require the use of fertilizers and pesticides. These chemicals are carried by runoff water and increase the concentration of ions, such as potassium (K^+), in rainfall. In addition, village crops have replaced natural forests and savannas, reducing biodiversity and disrupting local biogeochemical cycles (Kouassi et al., 2022). In the early 2010s, factories were set up near Lamto. Although some have since closed, their activities have had a lasting impact on the environment. Industrial emissions, such as fine particles and sulphur compounds, enrich precipitation with Na^+ and Cl^- ions. This pollution illustrates the links between local activities and atmospheric conditions.

Climate change is amplifying these phenomena. Episodes such as El Niño influence precipitation patterns and increase the transport of pollutants from cities to rural areas. These phenomena, combined with rising temperatures, have intensified environmental pressures on the Lamto region over the past two decades. These trends also suggest that, in addition to natural sources (remote: marine aerosols, Sahel and Saharan dust; local: dust, biomass burning) related to the season, other anthropogenic factors may also contribute to this increasing trend. This shift in atmospheric chemistry could have environmental implications, affecting soil and water quality through increased chemical deposition, and highlights the need for ongoing monitoring and potential mitigation efforts.

Across all variables at the annual scale – rainfall amount and concentrations of sodium, potassium, and chloride ions – the MK test exhibits no statistically significant trends over the 22-year period. The p-values for all tests exceed the common significance threshold of 0.05, with values ranging from 0.26 for rainfall amounts to as high as 1.0 for K^+, suggesting that any observed changes are likely due to random variations rather than systematic trends.

Monthly trends

Monthly variations in rainfall amounts and average Na^+, K^+, and Cl^- ion concentrations are shown in Figure 9. The SMK test applied to monthly rainfall amount (Ht) indicates no significant trend ($p > 0.05$). The test statistic ($z = -0.26$) and Tau value (-0.01) and SSE (-0.36 mm/month) imply that rainfall amount has decreased moderately over the study period. However, the confidence interval for the Sen's Slope (-5.13 to 2.69) is quite wide, suggesting some variability in the data, which could be influenced by large-scale climatic factors. For example, Madden Julian Oscillation (MJO) has been found to enhance monsoon convection over West Africa (Lavender and Matthews, 2009). Also, global teleconnections such as El Niño may affect global atmospheric composition and may impact dust emissions and transport in the Sahel and Saharan regions (Prospero and Nees, 1986). Therefore, seasonal and interannual variability of climate and atmospheric chemical composition in the region plays a critical role in influencing both the local ecosystem dynamics and the deposition patterns of pollutants at Lamto (Delon et al., 2010). Despite this variability, the overall trend points to an increase in rainfall at Lamto from January 1994 to December 2015. This increasing trend in monthly rainfall amount could be indicative of changing climatic conditions in the region, which may have implications for local agriculture, water resources, and ecosystem dynamics.

In contrast to the rainfall amount, the average monthly concentrations of Na^+, K^+, and Cl^- ions show no significant trends over the study period. The SMK trend test applied to Na^+ shows a p-value of 0.46, with a Z-score of 0.74 and a Tau value of 0.03, indicating no statistically significant trend. The estimated SSE is minimal at 0.01 µg/m^3 per month, with a confidence interval ranging from -0.41 to 0.46 µg/m^3/month, suggesting that sodium levels have remained relatively stable. Moreover, K^+ records no significant trend, with a p-value of 0.12, a Z-score of 1.55, and a Tau value of 0.07. The SSE is slightly negative at -0.03 µg/m^3 per month, with a confidence interval of -0.47 to 0.11 µg/m^3 per month, indicating stable or slightly decreasing potassium levels. Similarly, Cl^- shows no significant trend, with a p-value of 0.20, a Z-score of 1.27, and a Tau value of 0.06. The estimated SSE is 0.04 µg/m^3 per month, with a confidence interval from -0.49 to 0.47 µg/m^3 per month, indicating stable chloride concentrations over time.

Conclusion

This study conducted a comprehensive analysis of the variability and trends of sodium (Na^+), potassium (K^+), and chloride (Cl^-) ions in rainwater in the rural site of Lamto in Côte d'Ivoire over a 22-year period between 1994 and 2015. By applying non-parametric statistical analyses to in-situ rainfall measurements and satellite-based reanalysis of CHIRPS precipitation data, we established a robust understanding of rainfall patterns and the

influence of the WAM on these ion concentrations at Lamto. Our findings reveal statistically significant increasing trends in all three ion concentrations in rainwater at the event scale over the 22-year observation period, suggesting a complex interplay of natural and anthropogenic factors. The absence of a significant trend in rainfall points to a stable rainfall regime over this period. Also, the increasing trends in ion concentrations in rainwater highlight the impact of local and regional environmental changes and are likely driven by changes in local land use, increased human activity, and broader climatic factors. The strong positive correlations between Na^+ and Cl^- suggest a common source for these two ions, likely related to the marine aerosols transported by the WAM. The increasing trend in K^+ shows the influence of local anthropogenic activities, such as biomass burning, agricultural practices, and building infrastructure in this region.

The strong correlation between CHIRPS and in-situ rainfall data emphasises the potential utility of such reanalysis datasets for both air quality and climate studies across data-scarce regions. No significant trends were detected at the monthly and annual scales for rainfall height, Na^+, Cl^-, and K^+ in rainwater samples from Lamto, Côte d'Ivoire. This may reflect the high variability of aggregated data, the episodic and non-linear nature of event-scale processes, or the smoothing effects of temporal aggregation, which can obscure finer-scale variations. These scale-dependent trends, far from being contradictory, actually complement each other. When we examine trends based on specific events, we detect early signs of changes in the concentrations of Na^+, Cl^-, and K^+ ions in rainwater in Lamto. On the other hand, analyses of monthly and annual trends offer us a broader and more sustainable perspective. They average out short-term fluctuations to reveal deeper, longer-term trends that, although slower to emerge, are crucial for understanding sustainable change. Together, these approaches provide a comprehensive view, highlighting both short- and long-term changes. The results of monthly and annual trends are essential for predicting the impacts of natural (such as WAM) and anthropogenic (such as biomass fires) sources on the chemistry of rainfall in Lamto.

Moving forward, continuous monitoring and enhanced modeling efforts are crucial for better understanding the long-term environmental variability and impacts of Na^+, K^+, and Cl^- in rainwater at Lamto. These results highlight the importance of long-term monitoring atmospheric composition in the region, especially as climate change and human activities continue to shape the environment. These changes in the environment and activities, and their impact on Lamto, need to be assessed in future work.

Data availability

The INDAAF data can be retrieved from the website. The ion concentration data from Lamto rainwater used in this study are available for download at https://doi.org/10.25326/276. Interested persons may contact corinne.galy-lacaux@aero.obs-mip.fr or beatrice.marticorena@lisa.ipsl.fr for these data.

Author contributions

Conceptualization and methodology, N.E.T., M.O.G. and Y.F.; analysis , N.E.T., M.O., A.B., A.K.A., Y.F and V.Y; writing – original draft preparation, N.E.T., M.O.G., A.B., Y.F, A.K.A. and V.Y.; writing – review and editing, N.E.T., M.O.G., A.B., Y.F., A. K.A and V.Y. All authors have read and agreed to the published version of the manuscript.

Competing interests

The authors declare no conflict of interest.

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