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Measurement report: Long-term measurements of ozone concentrations in semi-natural African ecosystems

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Abstract. In the framework of the International Network to study Deposition and Atmospheric chemistry in Africa (INDAAF) program, we present the seasonal variability of atmospheric ozone concentrations at the regional scale. The contributions of local atmospheric chemistry and meteorological parameters to ozone photochemistry are investigated, as are long-term trends in ozone concentrations. Fourteen measurement sites were identified for this study, representative of the main African ecosystems: dry savannas (Banizoumbou, Niger, Katibougou and Agoufou, Mali, Bambey and Dahra, Senegal), wet savannas (Lamto, Côte d'Ivoire, Djougou, Benin), forests (Zoétélé, Cameroon, Bomassa, Republic of Congo) and semi-agricultural/arid savanna (Mbita, Kenya, Louis Trichardt, Amersfoort, Skukuza and Cape Point, South Africa). Over the period 1995-2020, monthly ozone concentrations were measured at these sites using passive samplers. Monthly averages of surface ozone range from 4.7±1.4 ppb (Bomassa) to 31.0±10.5 ppb (Louis Trichardt). Ozone levels in the wet season (in dry savanna) are higher and comparable to concentrations in the dry season (in wet savanna and forest). In East Africa, ozone levels show no marked seasonality, with higher levels in Southern Africa. In the Sahel, under the influence of temperature, ozone formation is closely linked to biogenic Volatile Organic Carbon (VOC) emissions. It is also sensitive to nitrogen monoxide (NO) emissions in the presence of high precipitation and humidity. Biogenic VOC (BVOC) emissions, anthropogenic NOx, temperature and radiation are the dominant contributors to O_3 formation in wet savannas and forests. At the southern African sites, the most important parameters influencing surface ozone concentrations are humidity, temperature, VOC emissions (anthropogenic and biogenic) and NOx. At the annual scale, Katibougou and Banizoumbou sites (dry savanna) experienced a significant decrease in ozone concentrations respectively around -0.24 ppb yr^{-1} (-2.3 % yr^{-1}) and -0.15 ppb yr^{-1} (-1.9 % yr^{-1}). Seasonal Kendall statistical tests revealed decreasing trends of -0.07 ppb yr^{-1} in Banizoumbou and -0.24 ppb yr^{-1} in Katibougou. These decreasing trends are consistent with those observed for nitrogen dioxide (NO_2) and biogenic VOCs. A significant increasing trend is observed

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in Zoétélé, with the Sen slope estimated at 0.1 ppb yr^{-1} and at Skukuza (Sen slope = 0.3 ppb yr^{-1}). The increasing trends are consistent with the increase in biogenic emissions at Zoétélé and NO_2 levels at Skukuza. Very few surface O_3 measurements exist in Africa, and long-term results presented in this study are the most extensive for the studied ecosystems.

1 Introduction

Ozone (O_3) is a greenhouse gas, difficult to observe and quantify on a global scale due to its acute spatial variability resulting from its variable photochemical lifetime (between 20 and 25 days) (Cooper at al., 2020; Young et al., 2013). It has phytotoxic effects altering key plant physiological processes that can significantly reduce the productivity of agricultural crops and ecosystems (Dufour at al., 2021; Lelieveld et al., 2015; Mills et al., 2018; Monks et al., 2015). At the local scale, its presence in high concentrations in the lower troposphere is harmful to human health, notably through irritation of the upper airways (Camredon and Aumont, 2007; Schultz et al., 2017). O_3 is a secondary air pollutant, meaning that it is not emitted directly but formed in the troposphere as a result of oxidative chemical reactions of precursor gases such as nitrogen oxides (NOx), carbon monoxide (CO) and volatile organic compounds (VOCs) (Lu et al., 2019; Schultz et al., 2017). It is chemically lost by photodissociation or by surface deposition and uptake by plant stomata (Silva and Heald, 2018), or by heterogeneous reactions involving aerosols. Stomatal uptake of O_3 and subsequent damage to plants can lead to changes in biosphere-climate interactions (Sadiq et al., 2017). Mitigating its negative impacts on health requires reducing both pollutant concentrations and population exposure (Petetin et al., 2022). These changes are compounded by the variation of O_3 precursors, which in recent decades have shifted from high and mid-latitudes to low latitudes, where O_3 production efficiency is greater (Zhang et al., 2016). These variations are particularly significant in tropical regions, where seasonal cycles linked to natural and anthropogenic sources of gas and particle emissions are well marked (Adon et al., 2010). Air quality forecasts could therefore be used to warn the population of the potential occurrence of a pollution episode (Petetin et al., 2022). Its long-term importance for atmospheric chemistry has been investigated by several studies on air quality (Monks and Leigh 2009) and atmosphere-biosphere interactions (Fowler et al., 2009; Laban et al., 2018).

International Global Atmospheric Chemistry Project (IGAC) has produced the Tropospheric Ozone Assessment Report (TOAR) on the global Measures for Climate Change, Human Health and Crop/Ecosystem Research (www.igacproject.org/TOAR). This report stated that free tropospheric O_3 has increased during the industrial era and in recent decades (Gaudel et al., 2018; Tarasick et al., 2019). Despite these years of regional and global surface O_3 research and monitoring, many regions of the world such as Africa, South America, the Middle East and India, remain under sampled, leading to incomplete knowledge of the horizontal, vertical and temporal distribution of O_3 (Cooper et al., 2014; Lin et al., 2015; Mills et al., 2018; Oltmans et al., 2013; Sofen et al., 2016). Although Africa is considered as one of the most sensitive continents to air pollution and climate change, it is the least studied (Laakso et al., 2012; Swartz et al., 2020a). Indeed, very little work has been done on regional atmospheric chemistry and the dominant atmospheric processes related to surface O_3 formation in Africa. With the exception of a few sites, O_3 variability is not yet sufficiently documented on this continent (Gaudel et al., 2018; Fleming et al., 2018; Mills et al., 2018).

From this perspective, long term measurement programs play a vital role in studies of air pollution and the various changes in the chemical composition of the atmosphere. These long-term assessments are crucial for posing the most topical research questions on atmospheric chemistry (Vet et al., 2014), in order to provide the right answers for relevant decision-making at local and global scale. In situ, satellite, O_3-sonde and aircraft observations provide a substantial amount of information on the current distribution of tropospheric O_3, its variability and trends (Gaudel et al., 2018; Tarasick et al., 2019). They are well suited to improve our understanding of emissions, transport, chemical reactions, deposition processes and the impacts of atmospheric species on human health, vegetation and climate change (Lefohn et al., 2018). Numerous projects and programs have therefore sprung up in several places around the world, for decades.

In Africa, the International Network to Study Deposition and Atmospheric Composition in AFrica (INDAAF; https://indaaf.obs-mip.fr), operational since 1995, is dedicated to study the evolution of the chemical composition of the

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atmosphere and deposition fluxes. INDAAF is a national observatory (Service National d'Observation, SNO) of the Institut National des Sciences de l'Univers (INSU) of the Centre National de Recherche Scientifique (CNRS) and of the Institut de Recherche pour le Developpement (IRD), and a labelled component of the European research infrastructure Aerosols, Clouds and Trace gases Research Infrastructure (ACTRIS). The INDAAF long term monitoring network is also labelled by Global Atmosphere Watch (GAW) program of the World Meteorological Organization (WMO) as a contributing network and is a component of the DEBITS (Deposition of Biogeochemically Important Trace Species) activity of IGAC (International Global Atmospheric Chemistry).

Previous existing studies have considered surface ozone levels in Africa. Adon et al. (2010) characterized the ozone concentration levels (together with several atmospheric pollutants), from 2002 to 2007, at seven remote sites in West and Central Africa, while Martins et al. (2007) investigated O_3 concentrations in Southern Africa over a period of 9 to11 years (1995-2005). In South Africa, Swartz et al. (2020a) assessed long-term seasonal and interannual trends of O_3 based on a 21-years (1995-2015) dataset at the Cape Point station. This work was continued at 3 historic IDAF-DEBITS sites (Amersfoort, Louis Trichardt, Skukuza, Swartz et al., 2020b). Laban et al. (2018) reported O_3 levels in northeastern South Africa, and characterized the links between observed NOx and O_3 concentrations. Other works were carried out by Bencherif et al. (2020), Gaudel et al. (2020), Hamdun and Arakaki (2015), Ihedike et al. (2023), Josipovic et al. (2010), Khoder (2009), Lee et al. (2021), Ngoasheng et al. (2021), Zunckel et al. (2004) in different locations in Africa to characterize O_3 pollution levels. Other projects such as POLCA (POLlution des Capitales Africaines) and DACCIWA (Interactions Dynamique-Aérosols-Chimie-Nuages en Afrique de l'Ouest), have also been implemented in African capitals such as Bamako, Dakar and Yaoundé (Adon et al., 2016), Abidjan, Cotonou (Bahino et al., 2018) and have provided O_3 concentration surface measurements.

However, very little information is available on the long-term evolution of O_3 chemistry in Africa. The impact of meteorological parameters (temperature, humidity, rainfall, radiation) and atmospheric chemistry (NOx and VOCs concentrations) on the seasonality of O_3 concentrations, and the analysis of long-term O_3 trends remain unexplored. Further work is therefore needed to fill the data gaps in Africa, and better understand the mechanisms of O_3 formation as a function of ecosystems and their long-term evolution.

As part of the INDAAF program, this study aims to improve the long-term assessment of surface O_3 in the western, central, eastern and southern African regions. In the first objective, we first document the long-term (1995-2020 depending on the site) monthly, seasonal and interannual variability of O_3 concentrations on a regional scale at fourteen sites grouped by ecosystem (dry savannas, humid savannas, forests and agricultural/semi-arid savannas), followed by a comparative study with existing references. The study goes further by discussing the seasonal architecture of anthropogenic and biogenic O_3 precursors based on meteorological parameters and emission inventories. In the second objective, we study the impact of meteorological parameters (temperature, humidity, precipitation, radiation) and atmospheric chemistry precursors (NOx and VOC) on photochemical O_3 production, using principal component analysis. In this section, we establish the correlation matrix between O_3, its gaseous precursors and meteorological parameters. An attempt is made to assess their different contributions to photochemical O_3 production in each ecosystem. In the third objective, we use non-parametric statistical tests to assess long-term seasonal and annual trends in O_3, and discuss the results according to trends in anthropogenic and biogenic emissions of precursors. For the first time, the chemical evolution of tropospheric O_3 is examined over the long term at all INDAAF and companion sites. This study provides a robust regional mapping of the long-term seasonal cycle O_3 formation at the continental scale.

2. Materials and methods

2.1 Sampling sites

Fourteen O_3 measurement sites located in different African ecosystems have been selected for this long-term study of tropospheric O_3 chemistry (Fig. 1), among which 8 stations of the INDAAF long-term monitoring network located in 7 West and Central Africa countries (Mali, Niger, Ivory Coast, Senegal, Benin, Congo and Cameroon). These sites are characteristic

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of dry savanna, humid savanna and forest ecosystems (Table 1). A detailed description of INDAAF monitoring stations and land use classes is available in Adon et al. (2010, 2013). Other sites implemented through INDAAF's companion projects and using the same O_3 measurements protocols were also selected for this study. The site of Dahra in Senegal, part of the 'Cycle de l'Azote entre la Surface et l'Atmosphère en afriQUE" (CASAQUE) project is located in dry savanna and used for grazing (Bigaignon et al., 2020). The site of Mbita, part of the Integrated Nitrogen Management system (INMS) is located in East Africa. In South Africa, four long-term DEBITS sites (Louis Trichardt, Skukuza, Cape Point and Amersfoort) are considered. They are regionally representative of the specific ecosystems of southern Africa. Full descriptions of these South African sites can be found in the works of Conradie et al. (2016), Laakso et al. (2012) and Swartz et al. (2020a,b). All study sites are representative of semi natural rural sites in remote regions. Table 1 presents geographical coordinates and some ecological and climatological characteristics of the sites. In the remainder of the text, the measuring stations will be referred to using the following abbreviations: Banizoumbou (Ba), Katibougou (Ka), Agoufou (Ag), Bambey (Bb), Dahra (Da), Lamto (La), Djougou (Dj), Zoetele (Zo), Bomassa (Bo), Mbita (Mb), Louis Trichardt (LT), Skukuza (Sk), Cape Point (CP) and Amersfoort (Af).

Table 1. Geographical, ecological and climatic characteristics of the study sites

Ecosystem	Station	Latitude, Longitude	Country	Land cover classes	Climate
Dry savanna	Banizoumbou (Ba)	13°18' N, 02°22' E	Niger	Open grassland with sparse shrub and culture	Sahelian
	Katibougou (Ka)	12°56' N, 07°32' W	Mali	Deciduous shrubland with sparse trees	Sudano-Sahelian
	Agoufou (Ag)	15°20' N, 01°29' W	Mali	Open grassland with sparse shrub and trees	Sahelian
	Bambey (Bb)	14°42' N, 16°28' W	Senegal	Cultivated grass land with sparse trees	Sahelian
	Dahra (Da)	15°24' N, 15°26' W	Senegal	Open grassland with sparse shrub and trees, sylvopastoral area	Sahelian
Wet savanna	Lamto (La)	06°13' N, 05°02' W	Cote d'Ivoire	Mosaic forest/savanna	Guinean
	Djougou (Dj)	09°39' N, 01°44' E	Benin	Deciduous open woodland	Sudano-Guinean
Forest	Zoetele (Zo)	03°10' N, 11°49' E	Cameroun	Dense evergreen lowland forest	Guinean
	Bomassa (Bo)	02°12' N, 16°20' E	Congo		Guinean
Agricultural field	Mbita (Mb)	0°25' S, 34°12' E	Kenya	Tropical agricultural area	Subtropical
	Louis Trichardt (LT)	22°59' S, 30°01' E		Cultivated/Semi-arid regional savanna	Subtropical
	Skukuza (Sk)	24°59' S, 31°35' E		Semi-arid regional background site surrounded by natural bushveld in a protected area Southern	

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Regional savanna/ Semi-arid	Cape Point (CP)	34°21' S, 18°29' E	South Africa	Hemispherical marine background site, rocky and sparsely vegetated, Fynbos biome	Mediterranean
	Amersfoort (Af)	27°04' S, 29°52' E		Semi-arid regional savanna, impacted by anthropogenic activities	Warmtemperate

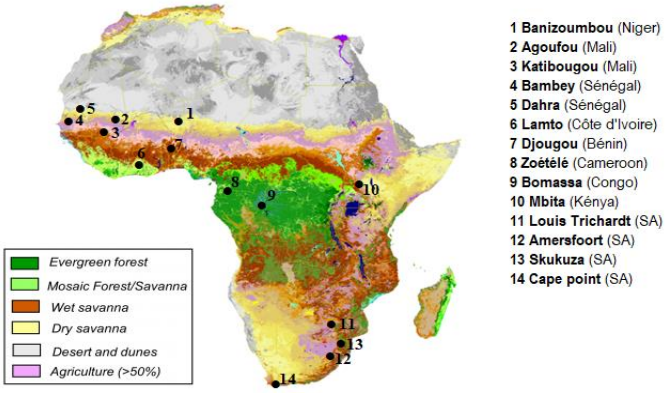

Figure 1. Location of the 14 measurement studied sites in Africa on a vegetation map (adapted from Mayaux et al., 2004)

2.2 Passive sampling and chemical analysis

O_3 concentrations were measured using passive samplers developed at the Laboratoire d'Aérologie (LAERO) in Toulouse in the framework of the INDAAF program, and at the North West University in Potchefstroom in South Africa. They are based on the passive sampling technique, which relies on laminar diffusion and the chemical reaction of the atmospheric pollutant under consideration (Adon et al., 2010; Ferm, 1991 and Martins et al., 2007). These sensors have been tested and validated in different tropical and subtropical regions (Carmichael et al., 2003; Martins et al., 2007). The measurement protocols including passive samplers deployment, analysis by Ionic chromatography and the calculation method of concentrations have been widely described in previous studies (Adon et al., 2010; Bahino et al., 2018; Carmichael et al., 2003; Ferm and Rodhe, 1997; Galy-Lacaux et al., 2009; Galy-Lacaux and Modi, 1998; Ossohou et al., 2019; Swartz et al., 2020b). In this paper, we use monthly database of O_3 concentrations. The concentration measurement period runs from the start date of measurements at each site to 2015-2020 (Table 2). O_3 concentration collection efficiency on the sampling period (ratio of the number of valid concentrations to the number of filters analysed) was assessed at each of the 14 sites (Table 2). All wet and dry seasons duration are indicated in Table 2. These proportions are fairly representative of high quality measurements as indicated in the work of Laakso et al. (2008, 2012), Laban et al. (2018) and Petäjä et al. (2013). Measurements of O_3 concentrations are continuing at the various sites and are referenced in the INDAAF website (https://indaaf.obs-mip.fr).

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Table 2. Sampling period and concentration data collection efficiency

Ecosystem	Station	Sampling period	Detection limit (ppb)	Data collection efficiency	Season	Measurement altitude (m)
Dry savanna	Ba Ka Ag Bb Da	2000-2020 2001-2020 2005-2018 2016-2020 2012-2020	0.1	232/248 208/240 109/132 47/50 87/104	Dry season: Oct-May Wet season: Jun-Sep	1.5
Wet savanna	La Dj	2001-2020 2005-2020		226/240 172/186	Dry season: Nov-Mar Wet season: Apr-Oct	1.5
Forest	Zo	2001-2020		208/240	Dry season: Dec-Fev and July-Aug Wet season: Mar-Jun and Sept-Nov	3
	Bo	2001-2020		164/240	Dry season: Dec-Fev Wet season: Mar-Nov	
Agricultural field	Mb	2017-2020		41/43	Dry season: Jun-Oct and Jan-Fev Wet season: Mar-May and Nov-Dec	1.5
Regional savanna/semi-arid	LT Sk Af	1995-2015 2000-2015 1997-2015		236/248 165/192 189/221	Dry season: Apr-Sep Wet season: Oct-Mar	1.5
	CP	1995-2020		225/248	Dry season: Oct-Mar Wet season: Apr-Sep	

2.3 Meteorological parameters and leaf area index

In order to characterize each measurement site, classical meteorological parameters are used such relative humidity, ambient air temperature, rainfall, radiation and leaf area index (LAI). At Ba, Bb, La, Dj and Zo sites, the data on ambient air temperature, relative humidity and rainfall are extracted from the AMMA-CATCH database (Analyse Multidisciplinaire de la Mousson Africaine - Couplage de l'Atmosphère Tropicale et du Cycle Hydrologique; www.amma-catch.org/) and the Observatoire de recherche en environnement "Bassins versants tropicaux expérimentaux" (SO BVET; bvet.obs-mip.fr/) (Ossohou et al., 2019). The measuring devices used at Ka, La and Bo is described in the same work (Ossohou et al., 2019). At Bb site, relative humidity, temperature and rainfall are collected in the INDAAF database (https://indaaf.obs-mip.fr). At Da site, measurements of meteorological parameters come from a measuring station installed by the University of Copenhagen (Bigaignon et al., 2020). In Ka, Ag, Bo, Mb, LT, Af, Sk and CP, the meteorological data are provided by the intermediate reanalysis archive (ERA 5) of the European Center for Medium-Range Weather Forecasts (ECMWF). The time series of global solar radiation used in this study at all sites except Dahra are also ERA 5 reanalysis data obtained from ECMWF. LAI data are obtained from MODIS (Moderate Resolution Imaging Spectroradiometer) with a resolution of 0.25 km² for an 8-day time scale centred around each station (Ossohou et al., 2019). All these parameters are collected at the same sampling period as O_3 concentrations, with the exception of LAI measurements, which began in 2000 (https://modis.ornl.gov/data.html).

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2.4 NO_X and VOC emissions

The emissions of volatile organic compounds (VOCs) and nitrogen oxides (NO_X) from biomass combustion were downloaded from the ECCAD (Emissions of atmospheric Compounds and Compilation of Ancillary Data) database in the GFED4 inventories for 0.25°x 0.25° grid cells. BVOC emissions are extracted from the MEGAN-MACC inventory in the ECCAD database (eccad.aeris-data.fr). The biogenic NO fluxes used are model outputs in reference to the work of Delon et al. (2010, 2012). They were filtered in the eastern grid from 5° S to 20° N in latitude, and 20° W to 30° E in longitude over the period from 2002 to 2007 and cover only the Ba, Ka, Ag, La, Dj, Zo and Bo sites. ECCAD platform is the emissions database of the international GEIA project (Global Emission InitiAtive: geiacenter.org) has been developed within the framework of the French atmospheric data center AERIS (http://www.aeris-data.fr) (Darras et al., 2022). The GFED4 inventory is based on satellite data of fire activities and vegetation productivity observed since 1997 (eccad.aeris-data.fr). MEGAN (Model of Emissions of Gases and Aerosols from Nature) inventory quantifies net biogenic emissions of isoprene and other gases emitted by vegetation into the atmosphere (Guenther et al., 2006; Sindelarova et al., 2014). Isoprene, α-pinene and β-pinene, which account for the largest proportion of BVOCs emitted by vegetation in Africa (Ferreira et al., 2010; Jaars et al. 2016; Liu et al., 2021; Saxton et al., 2007; Serça et al., 2001), were identified and used in this study. A more detailed description of these emission inventories is discussed in the work of García-Lázaro et al. (2018), Guenther et al. (2006) and Vitolo et al. (2018). For each emission category, NO_x (kg m^{-2} s^{-1}) and VOC (kg m^{-2} s^{-1}), we use the sum of fluxes from all biomass combustion sources (agricultural, waste combustion, savanna, grassland, scrubland, boreal forest, temperate forest, tropical deforestation, peat degradation and peat fires) at a monthly scale and over the study period for each site.

2.5 Statistical analysis

We use Principal Component Analysis (PCA) and Mann-Kendall test to investigate respectively the different contributions of O_3 precursors from photochemical formation and the monotonic trends in the time series of each site. PCA multivariate analysis method (Jolliffe, 1986), is often used in air quality analyses to identify the major sources of pollutants emitted into the atmosphere (Bruno et al., 2001; Lanz et al., 2007). It specifies the relationships between variables, the phenomena behind these relationships and the similarities between individuals (Mouissi and Alayat, 2016). In this study, the statistical analysis of meteorological data and atmospheric chemical pollutants was carried out on a data matrix made up of ten to eleven variables depending on the site, and 12 individuals representing the months of the year. For PCA implementation, R programming software was used. More detailed information on the calculation of PCA can be found in the literature (Aït-Sahaliaa and Xiu, 2019; Baglama et al., 2021; Duriš et al., 2021; Tsuyuzaki et al., 2020). The Mann-Kendall and seasonal Kendall tests, associated with the calculation of Sen's slope (Sen, 1968) is applied to all sites with at least 10 years of measure using XLSTAT 2022.2.1.1313 software. The trend is said to be statistically significant when the p-value of the test is less than 5%. In the case of Kendall's seasonal test, the seasonal nature of the series is taken into account. The literature provides extensive information on Mann Kendall trends calculations (Frimpong et al., 2022; Hirsch et al., 1982; Kendall, 1975; Merabtene et al., 2016). Another non-parametric breakpoint test (Pettitt test) is carried out using Khronostat 1.01 software to assess possible breaks in homogeneity in the O_3 concentration series and for optimal application of the trend test.

3. Results and discussion

3.1 Meteorological and biophysical parameters variation

The monthly variations of meteorological parameters and leaf area indexes (LAI) are shown in Fig. 2 for all sites. In dry savanna, the rainfall regime is unimodal, with the greatest amounts of rain recorded from July to September corresponding at the maxima of LAI. Mean air temperature ranged from 22.1 ± 0.9°C to 34.9 ± 0.4°C, with air relative humidity from 68% to 82%. The most elevated solar radiation is found at Ag (23.1 ± 0.5 $MJ.m^{-2}$). In wet savanna and forest sites, the rainfall pattern and LAI follow a quasi-bimodal distribution. The mean annual LAI varies from 1.2 ± 0.3 $m^2.m^{-2}$ (Dj) to 4.7 ± 0.7 $m^2.m^{-2}$ (Bo). The most significant monthly variations in relative humidity are found in Dj (23.2 - 84.1%). At Mb site, the maximum rainfall

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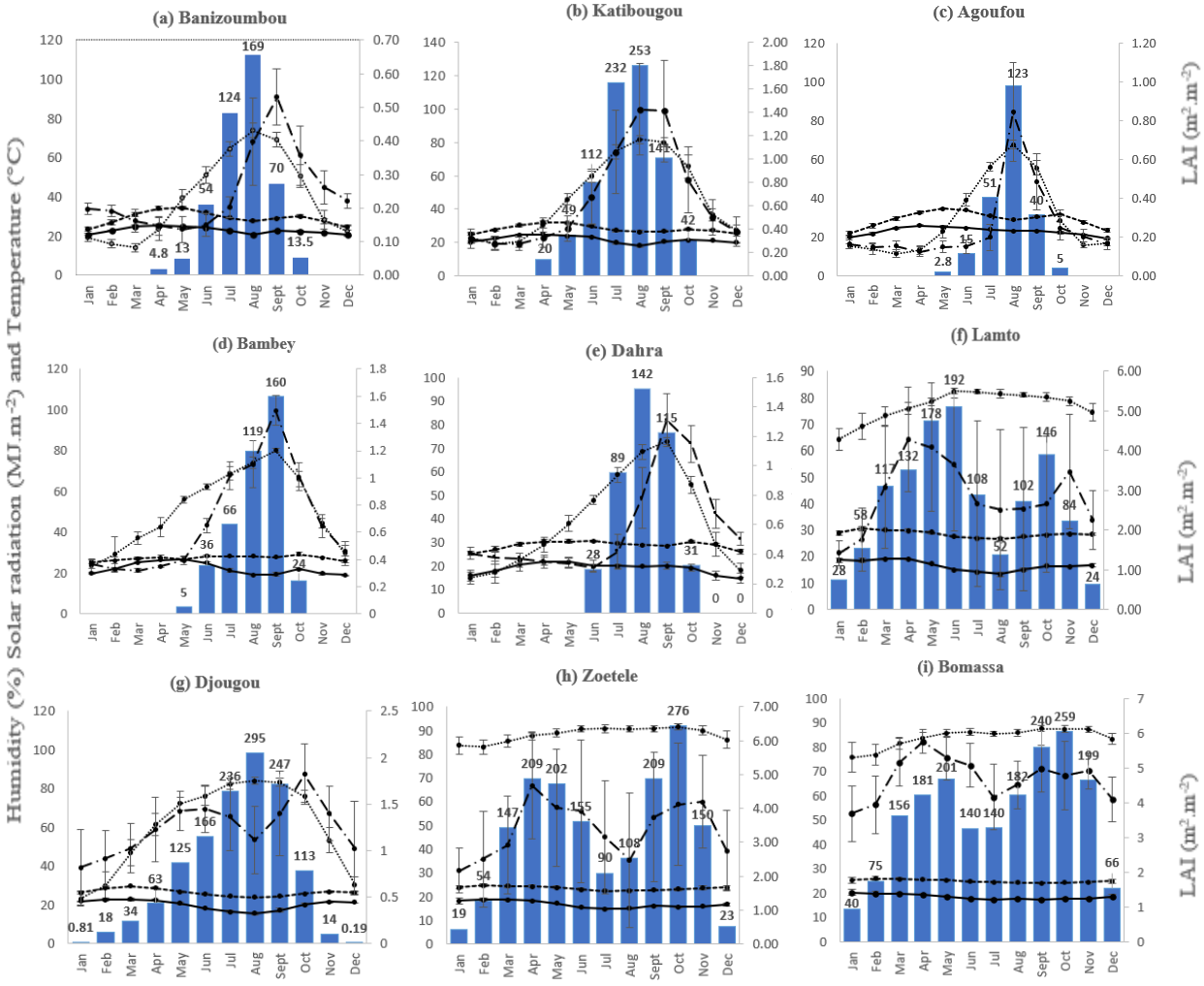
occurs between March and May, reaching 255 mm in April. The vegetation cover is denser at the end of the first wet season (1.3 ± 0.3 $m^2.m^{-2}$ in May) with an average value of humidity around 70%. In Southern Africa, the humidity varies from 13% to 22% year-round except at CP where variations are very low. The maximum of rain is collected between December and January (432 mm on average) at LT, Af and Sk sites and in August at CP (343 mm). LAI maxima are of the order of 1.6 ± 0.2 $m^2.m^{-2}$ at Af ; 3.1 ± 0.6 $m^2.m^{-2}$ at LT; 4.0 ± 0.1 $m^2.m^{-2}$ at CP and 1.7 ± 0.2 $m^2.m^{-2}$ at Sk in wet season. In wet savanna and forest, the temperature variations are low, as well as in Mb (23.6 ± 0.5°C). On the other hand, at the South African sites, the temperature reaches amplitudes ranging from 6°C to 10°C. From wet savanna to semi-arid savanna (South Africa), the average solar radiation is below at 22 $MJ.m^{-2}$. Along the north-south transect for the study sites, the gradient of humidity, leaf area index and rainfall are positive, whereas it is negative for temperature and radiation.



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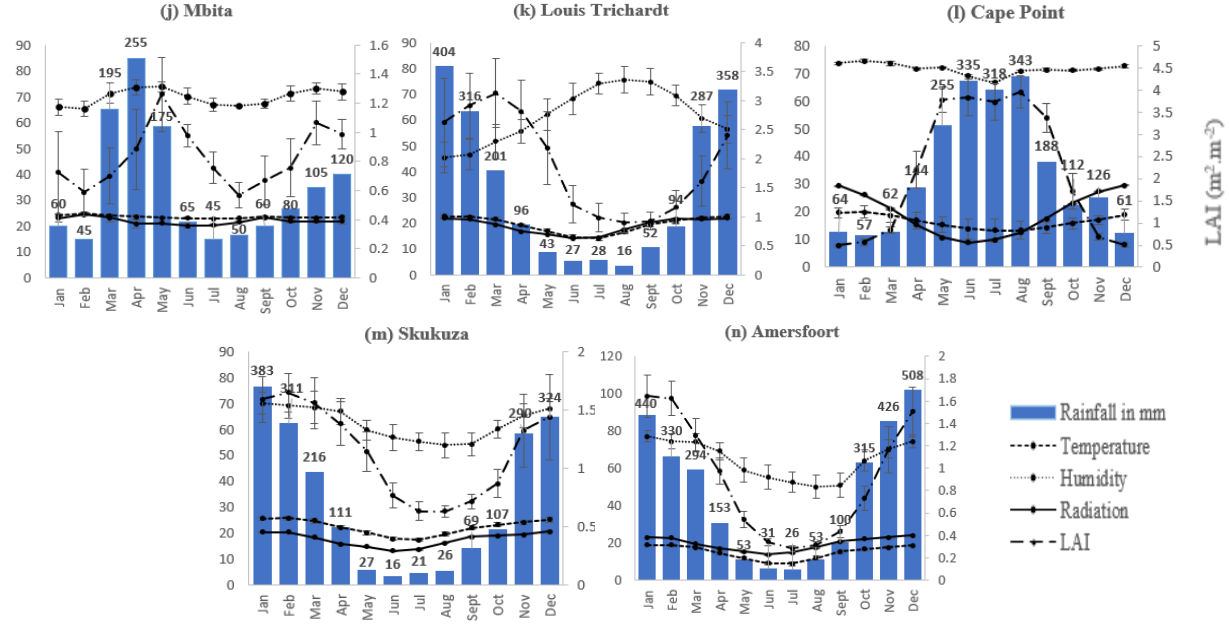

Figure 2. Mean monthly variation in air temperature (°C), rainfall (mm), relative humidity (%), solar radiation (MJ.m^{-2}) and leaf area index (m².m^{-2}). The mean absolute deviation is represented by the vertical bars.

3.2 Characterization of O_3 levels

3.2.1 Seasonal and annual variation in O_3 levels

3.2.1.1 Dry savanna

Figure 3 presents monthly O_3 surface concentrations measured in Ba, Ka, Ag (Fig. 3a), Bb and Da (Fig. 3b) representative of dry savannas in Niger, Mali and Senegal. The seasonal variability of O_3 is well marked: O_3 levels during the wet season are higher than in the dry season (Table 3). On most sites, from January to May, the O_3 concentrations gradually increase to finally reach annual peaks at the start of the wet season (May-June-July). The mean annual cycle of monthly O_3 concentrations at Ba, Ka, Ag (Fig. 4a), Bb and Da (Fig. 4b) are obtained from averages of monthly in situ measurements over the whole studied period. The annual distribution is similar to the regional rainfall pattern. O_3 concentrations decrease as the rainy season progresses, but remain at higher levels compared to the dry season. At dry savanna sites, monthly average surface O_3 concentrations range from 6.1±2.4 ppb to 14.5±2.6 ppb during the dry season, and from 13.9±5.1 ppb to 19.4±3.9 ppb during the rainy season (Table 3). From dry to wet season, O_3 levels increased from 18.7% to 68.5%. The annual O_3 concentrations ranged from 10.5±5.4 ppb at Ka to 14.8±4.3 ppb at Bb (Table 3). The high O_3 concentrations observed at the start of the rainy season are due to soil humidification during this period, which generates biogenic NO emissions pulses in the region. Indeed, the accumulated nitrogen in soils (in the form of ammonium and nitrate ions) from traditional agricultural practices, such as grazing, manure spreading and decomposition of crop residues (Delon et al., 2015; Laville et al., 2005) is released to the atmosphere when the first rains fall on dry soils. Bacterial nitrification is thus activated, leading to nitrogen consumption and consequent release of large pulses of NO (Adon et al. 2010; Delon et al., 2015; Jaegle et al., 2004; Laville et al., 2005; Ludwig et al., 2001; Ossohou et al., 2019). During the wet season, the decrease in O_3 levels may be attributed to a decrease in NOx concentrations. Indeed, soil mineral N is used by plants during their root growth phase, and is therefore less available for the production of NO to be released to the atmosphere (Homyak et al., 2014). These observations in dry savanna are confirmed

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by Stewart et al. (2008) who correlated O_3 production in the Sahel during the wet season with high NOx concentrations attributed to biogenic emissions during the AMMA (Analyse Multidisciplinaire de la Mousson Africaine) campaign.

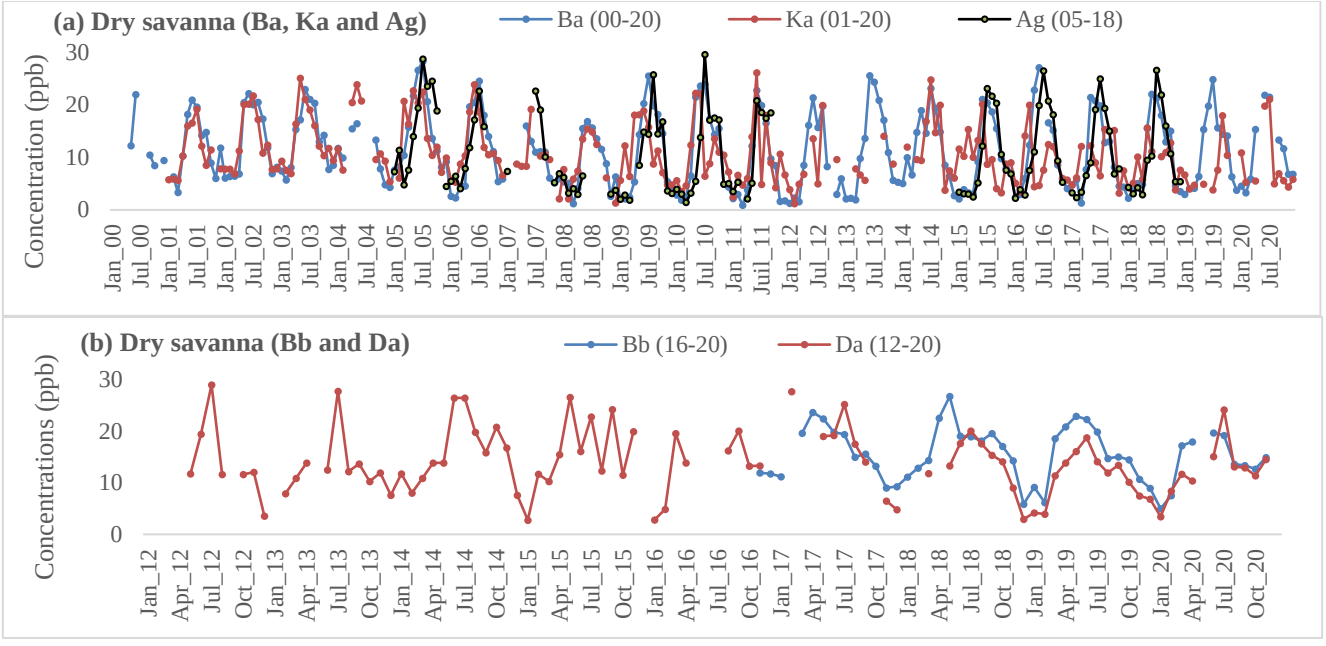
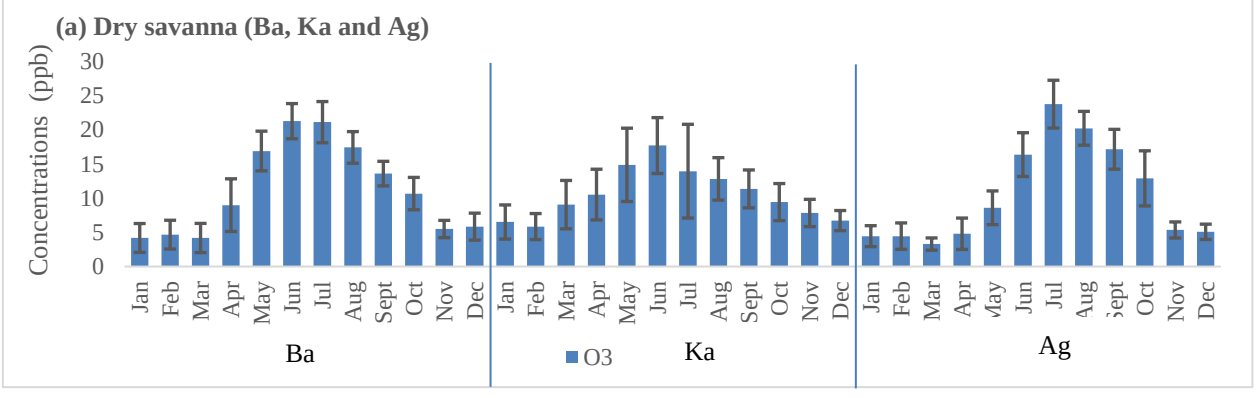


Figure 3. Monthly evolution of O_3 concentrations (ppb) in dry savanna (**a**) Ba (Niger), Ka, Ag (Mali) and (**b**) Bb, Da (Senegal).



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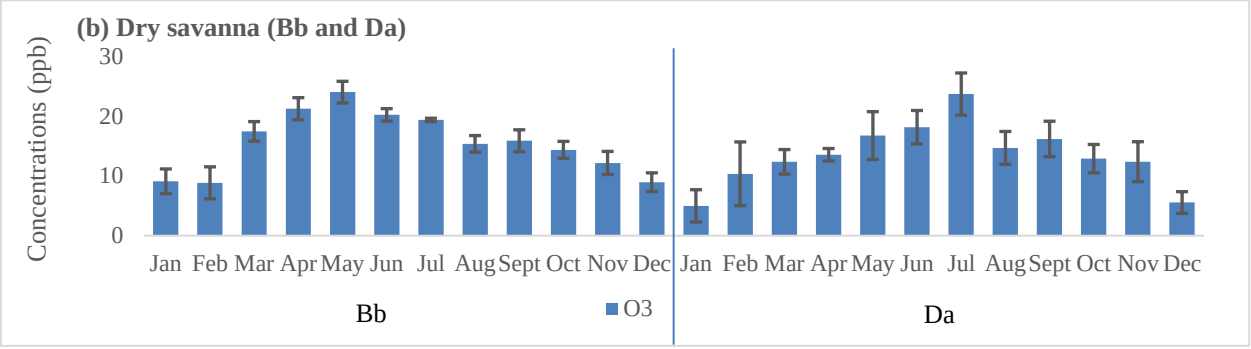

Figure 4. Mean monthly averages of O_3 concentrations (ppb) in dry savanna (**a**) Ba (Niger), Ka, Ag (Mali) and (**b**) Ba, Da (Senegal). Bars represent mean absolute deviation.

Table 3. Minimum, maximum and average of monthly, annual and seasonal O_3 concentrations at dry savanna sites (1995-2020)

Ecosystem		Monthly		Annual	Dry season			Wet season		
		min	max	moy	min	max	moy	min	max	moy
Dry savanna	Ba	0.9	28.3	11.2±6.9	4.2±2.7	16.9±3.7	7.6±2.9	13.6±2.5	21.2±3.5	18.3±3.2
	Ka	1.2	26.1	10.5±5.4	5.8±2.6	14.9±6.6	8.8±3.7	11.3±3.8	17.7±5.4	13.9±5.1
	Ag	1.4	29.5	10.5±7.3	3.3±1.1	12.9±4.6	6.1±2.4	16.4±3.8	23.7±4.6	19.4±3.9
	Bb	4.96	26.7	14.8±4.3	8.8±3.5	24.0±2.4	14.5±2.6	15.4±1.9	20.2±1.4	17.7±1.6
	Da	2.8	29.0	13.9±6.3	5.0±3.8	16.7±5.4	11.1±4.0	14.7 ±3.1	23.7 ±4.8	18.2±4.0
Wet savanna	La	4.25	20.6	10.8±3.3	9.9±1.4	15.1±2.3	13.5±2.3	6.7±1.1	12.2±1.9	9.0±1.5
	Dj	3.3	24.8	13.5±4.8	10.8±4.5	18.7±2.7	14.1±4.0	9.0±2.0	18.4±2.4	13.2±2.8
Forest	Zo	1.2	11.1	5.2±2.1	7.1±2.1	7.8 ±1.6	7.5±2.1	3.5±1.3	6.6±1.9	4.6±1.6
	Bo	1.5	8.3	3.9±1.1	4.0±1.0	5.2±1.2	4.7±1.4	2.8±1.0	5.4±1.0	3.7±1.0
Semi-agricultural or arid savanna	Mb	10.5	30.2	19.9±4.7	13.8±3.4	25.7±5.7	20.9±4.0	14.1±5.0	22.5±2.7	18.5±3.9
	Sk	6.3	64.1	22.8±7.3	23.0±9.6	30.2±6.0	25.9±7.3	14.5±1.9	29.2±5.6	20.3±5.4
	Af	3.2	55.5	26.9±6.3	19.9±6.2	31.2±7.6	24.5±6.3	23.8±4.9	34.4±7.4	29.0±7.3
	CP	3.3	67.4	26.8±6.2	17.3±5.3	30.4±6.0	23.4±5.6	25.9±6.3	32.1±5.7	29.8±6.1
	LT	9.0	86.6	30.8±8.0	24.7±7.9	40.1±10.8	32.0±8.9	21.3±5.2	36.0±9.1	28.3±8.2

3.2.1.2 Wet savanna and forest

Figure 5 presents the mean monthly surface O_3 concentrations in Dj, La (Fig. 5a), Zo and Bo (Fig. 5b). O_3 concentrations present a seasonality during the year. The maximum of the data series in La is 20.6 ppb in March (dry season) and the minimum is 4.3 ppb in October (wet season). In Dj, the O_3 levels are higher than in La. The monthly highest value recorded in Dj is 24.8 ppb in April (start of the wet season). At the forested ecosystems sites, O_3 concentrations are lower than in dry, wet and semi-agricultural/semi-arid savannas (Table 3). In Zo and Bo, the highest annual peaks are found in February (11.1 ppb and 8.3 ppb respectively). Monthly averages in the dry season ranged from 4.7±1.4 ppb (Bo) to 14.1±4.0 ppb (Dj), and in the wet season from 3.7±1.0 ppb (Bo) to 13.2±2.8 ppb (Dj) (Table 3). The O_3 mean annual cycle is shown in Fig. 6.

The high O_3 concentrations in dry season in these two ecosystems could be related to the biomass burning source, which is generally recorded during the months of December-February in rural tropical environments. Indeed, Abbadie (2006), Adon et

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al. (2010) and Galanter et al. (2000) have linked the high O_3 concentrations recorded in the dry season to the presence of NOx emitted by biomass combustion in the wet savanna. According to Adon et al. (2010), Baldy et al. (1996), Clain et al. (2009), Cros et al. (1992), Hamdun and Arakaki (2015), Martins et al. (2007) , Oluleye et al. (2013) the biomass burning is likely to contribute significantly to O_3 production through precursor emissions (NOx and CO) in the dry season (wet savanna) with nearly 30% to 80% of the savanna ground surface burnt annually between December and February. In other forests such as in Nepal, Kiros et al. (2016) also attribute high O_3 concentrations to increased burning of agricultural residues and forest fires. The high O_3 concentrations measured in Tranquebar (India) have been linked to increased emissions of NOx and other precursors from various sources (Debaje et al., 2003). Compared to wet and dry savannas, forest sites recorded the lowest O_3 amounts due to significant dry deposition of O_3 on the ground, on foliage and trees (Mari et al., 2011; Rummel et al., 2007; Saunois et al., 2009). Tropical forests are shown to be important O_3 sinks. A strong gradient of O_3 between forest and dry savanna in West Africa has been observed from aircraft measurements (Saunois et al., 2009).

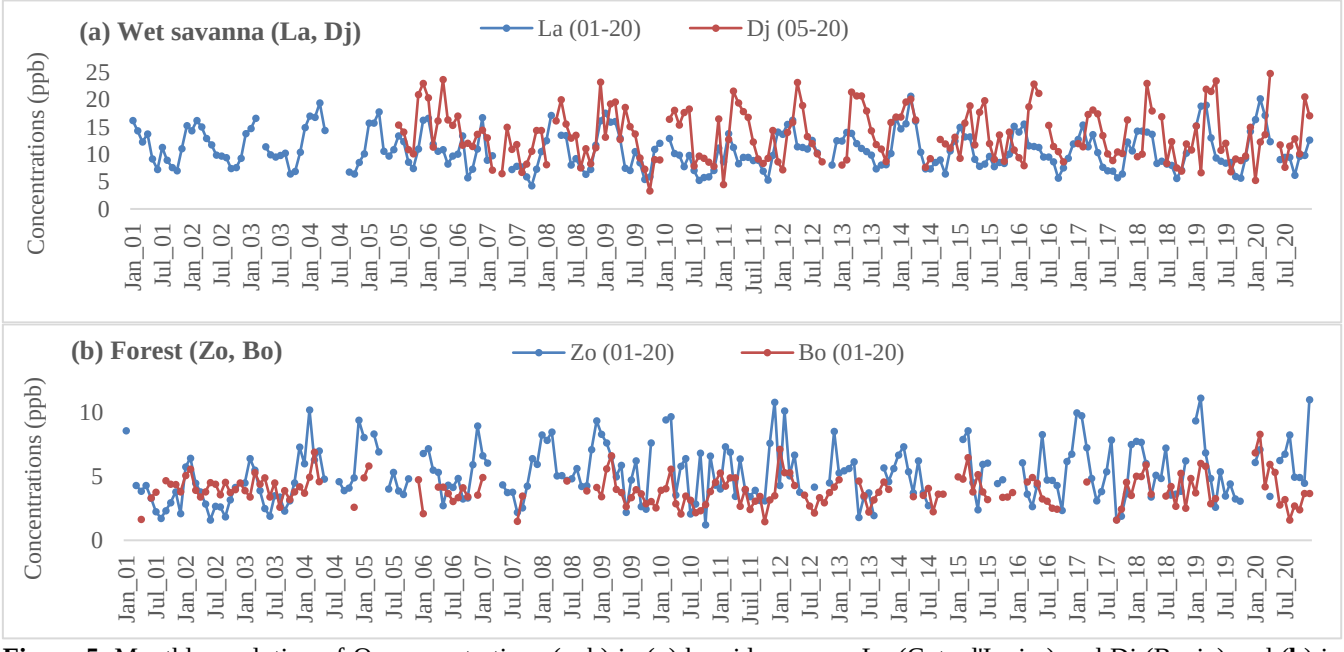

Figure 5. Monthly evolution of O_3 concentrations (ppb) in (**a**) humid savanna, La (Cote d'Ivoire) and Dj (Benin) and (**b**) in forest, Zo (Cameroon) and Bo (Congo).

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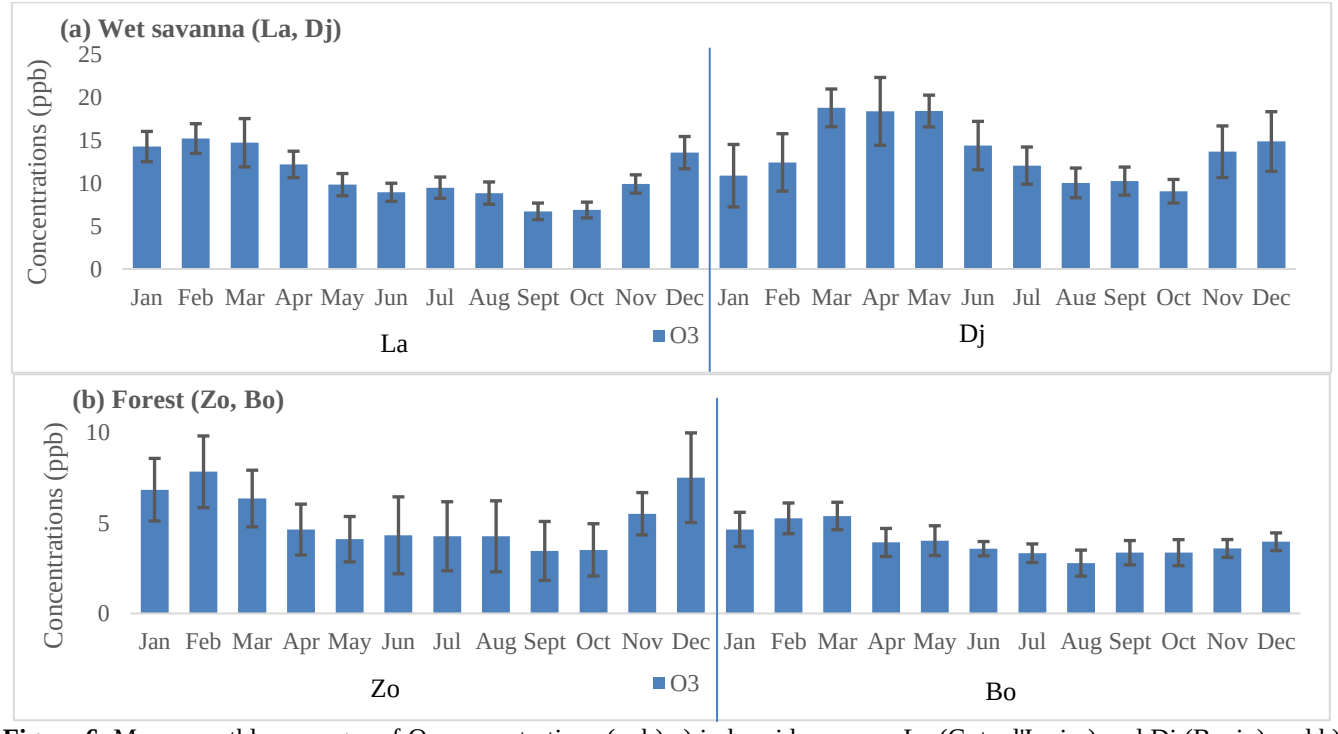

Figure 6. Mean monthly averages of O_3 concentrations (ppb) a) in humid savanna, La (Cote d'Ivoire) and Dj (Benin) and b) in forest, Zo (Cameroon) and Bo (Congo). Bars represent mean absolute deviation.

3.2.1.3 Agricultural and semi-arid savanna

Figures 7 presents the monthly evolution of surface O_3 concentration in agricultural site (Mb) and semi-arid savanna sites (LT, CP, Sk and Af). At Mb site, monthly O_3 concentrations do not exceed 30.2 ppb (Table 3). At the CP, LT, Sk and Af sites, O_3 levels are almost twice as high as in West African sites. The mean annual cycle of O_3 concentrations (Fig. 8a and b) shows that at Mb, O_3 levels are almost similar between seasons (Table 3). In southern African ecosystems, dry-season O_3 concentrations are the highest at LT and Sk. The annual averages are around 19.9±4.7 ppb at Mb; 22.8±7.3 ppb at Sk; 26.9±6.3ppb at Af; 26.8±6.2ppb at CP and 30.8±8.0ppb at LT (Table 3).

The O_3 levels observed in Mb during the dry season could be associated with biomass burning. Based on the analysis of burned surface areas, Bakayoko et al. (2021) indicated that Mb is strongly influenced by biomass burning from northern and southern sides during both dry seasons. High O_3 levels measured at Mb site during the dry season show similar values as in Nairobi, Kenya, during the same months (Kimayu et al., 2017). At southern African sites, O_3 levels could be attributed to a combination of regional and local influences, including emissions from industrial, vehicular and domestic biomass combustion to biomass combustion events in sub-Saharan Africa (Mozambique, Zambia, Zimbabwe and Angola) recirculated by anticyclonic air mass processes (Baldy et al., 1996; Laban et al., 2018; Martins et al., 2007; Swap et al., 2003; Tiitta et al., 2014). Biomass combustion is considered as a major source of O_3 precursors in South Africa (Ngoasheng et al., 2021; Vakkari et al., 2013) and in Southern Africa (Heue et al., 2016) and may explain the O_3 levels observed in dry season at LT and Sk. High O_3 levels in the wet season are thought to be due to soil microbial activity (Swartz et al., 2020a) as well as long-range transport of air pollutants emitted from the industrialized Highveld region (Abiodun et al., 2014; Ojumu, 2013). During the austral winter, O_3 concentrations in the boundary layer are higher (e.g. at CP and Af) due to a systematic increase in O_3 precursors from households, combustion for space heating (Bencherif et al., 2020; Laban et al., 2018; Lourens et al., 2011; Oltmans et al.,

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2013; Swartz et al., 2020b). The high concentrations measured at Af could also be due to industrial activities located near this site (Lourens et al., 2011).

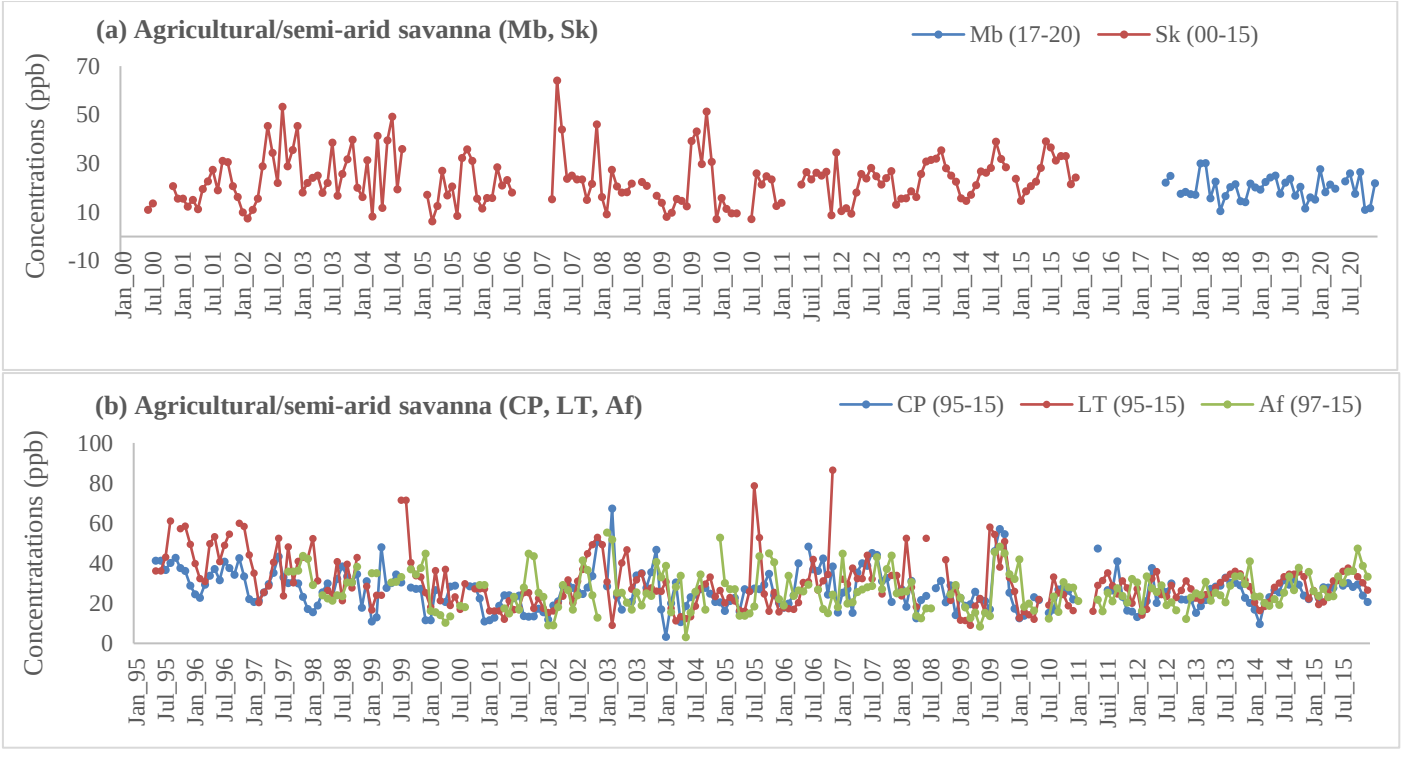
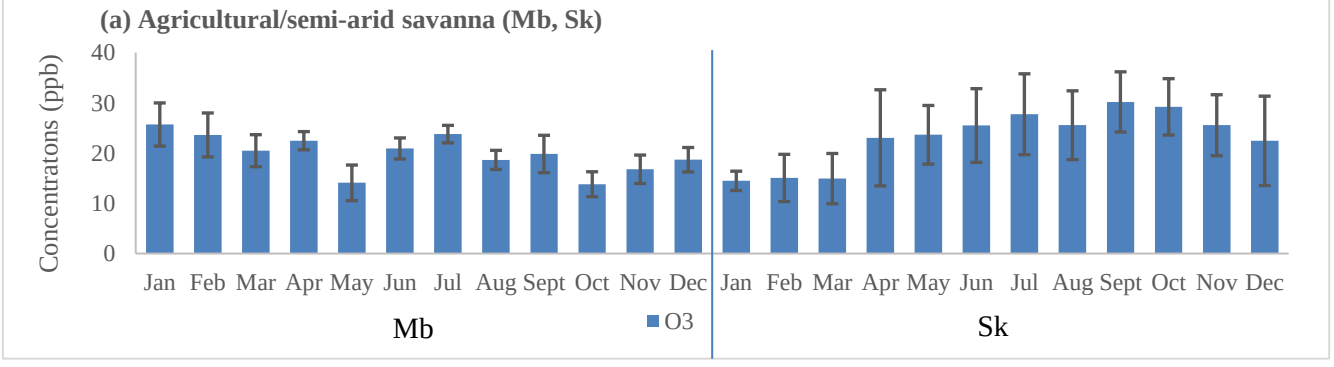


Figure 7. Monthly evolution of O_3 concentrations (ppb) in Agricultural/semi-arid savanna (**a**) Mb (Kenya) and Sk (South Africa) and (**b**) CP LT and Af (South Africa).



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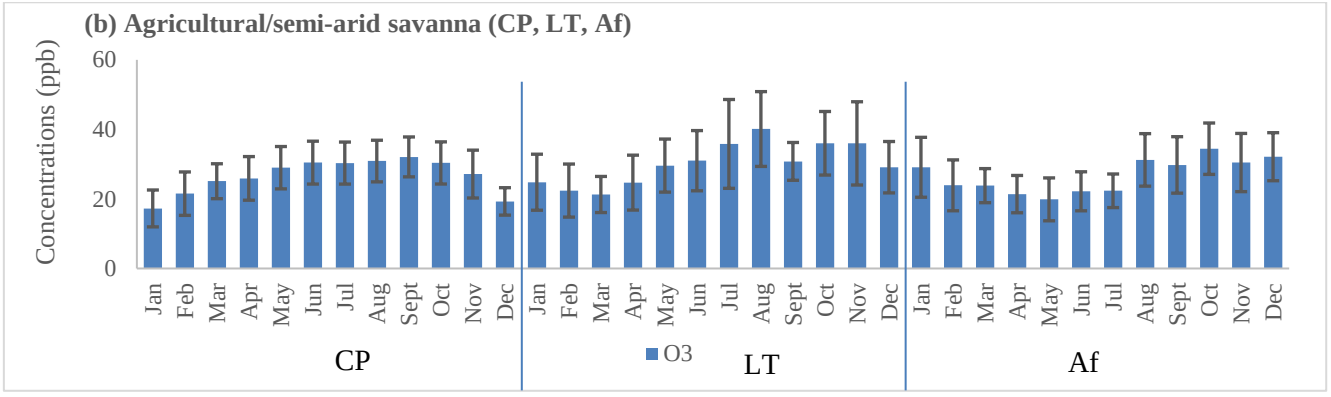

Figure 8. Mean monthly averages of O_3 concentrations (ppb) in Agricultural/semi-arid savanna (**a**) Mb (Kenya) and Sk (South Africa) and (**b**) CP, LT and Af (South Africa). Bars represent mean absolute deviation.

3.2.2 O_3 levels in Africa, set in a global context

O_3 concentrations measured at the 14 studied site are mapped on a seasonal and annual scale (Fig. 9).

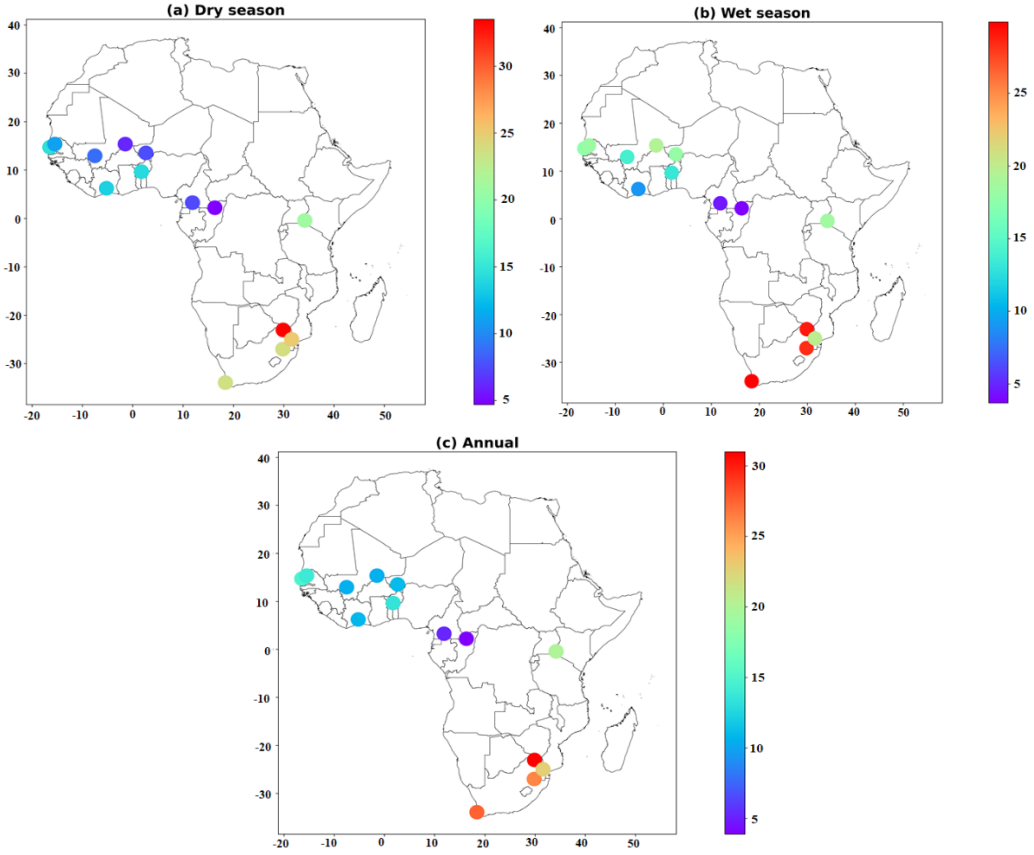

Figure 9. Seasonal and annual mapping of O_3 concentration levels in (**a**) Dry season, (**b**) Wet season and (**c**) Annual over the 14 studied sites.

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We compared African ozone levels related in this study with studies carried out in Africa and around the world over the last 20 years (Table 4, Fig. 10). The bibliographical synthesis takes into account studies where data measurement methodology has been clearly described. Sites where concentrations have been measured by passive samplers are listed. We have identified among others, sites in Nepal, North America, North-Eastern Europe, Asia and Africa. Figure 10 focuses more on O_3 monitoring studies in Africa.

Table 4. O_3 concentrations at various sites worldwide as reported in literature

Sites	Type	Period	O_3 (ppb)	References
India, Tranquebar	Rural	May 1997 -Oct 2000	17±7 - 23 ± 9	Debaje et al. (2003)
Sweden, Malmö (20 sites)	Rural	(16–24 Apr 2012 ; 28 May–4 Jun; 20–27 Aug 2012	37.0±5.4	Hagenbjörk et al. 2017
	Urban		35.0±3.9	
	Traffic		33.6±3.5	
Sweden, Umeå (20 sites)	Rural		27.7±8.4	
	Urban		26.7±7.3	
	Traffic		25.2±6.9	
China, Waliguan mountain	Rural	Sept 99-May 2001	44.9	Carmichael et al. 2003
Taiwan, Shui-Li	Rural		25.0	
Malaysia, Tanah Rata	Rural		16.0	
Indonesia, Bukit Kototabang	Rural		10.7	
Inde, Agra	Rural		30.8	
Argentina, Isla Redonda	Rural		15.9	
Brazil, Arembepe	Rural		19.2	
Turkey, Camkoru	Rural		35.4	
Arab Emirates, Al-Ain	Rural	Apr 2005-Apr 2006	8.7	Salem et al. 2009
	Industrial		5.9	
	Traffic		4.4	
	Commercial		5.9	
	Residential		7.3	
American Samoa Island, Tutuila	Urban and Rural	1990-2015	13.7 ± 1.0	Lu et al. 2018
Chili, El Tololo			32.0 ± 1.3	
South Africa, Cape point			24.3 ± 1.1	
Australia, Cape grim			24.9 ± 0.8	
New Zealand, Baring head			21.4 ± 1.6	
Syowa			25.2 ± 0.9	
Neumayer-G			24.3 ± 1.5	
Arrival heights			25.9 ± 1.5	
South Pole			28.4 ± 1.7	
Nepal, Bode	Urban		50.8 ± 14.1	
Nepal, Indrachowk			37.9 ± 6.2	
Nepal, Maharajgunj			46.4 ± 12.0	
Nepal, Mangal Bazaar			40.3 ± 8.0	
Nepal, Suryabinayak			41.3 ± 5.4	
Nepal, Bhaisepati	Suburban		47.7 ± 9.1	
Nepal, Budhanilkantha			50.3 ± 11.3	

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Nepal, Kirtipur		23 March -18 May 2013	46.9 ± 9.8	Kiros et al. 2016
Nepal, Lubhu			50.7 ± 8.6	
Nepal, Bhimdhunga	Rural		59.7 ± 11.9	
Nepal, Nagarkot			70.1 ± 9.7	
Nepal, Naikhandi			53.5 ± 13.0	
Nepal, Nala Pass			43.6 ± 10.5	
Nepal, Sankhu			57.6 ± 11.9	
Nepal, Tinpiple			54.4 ± 13.4	
Barrow Atmospheric Baseline Observatory	Remote site	1973-2015	15-44	Cooper et al. 2020
Mauna Loa Observatory (MLO)	Remote site	1973-2015	26-65	
American Samoa Observatory	Remote site	1973-2015	5-20	
South Pole Observatory	Remote site	1973-2015	17-40	
China	Rural site	2014-2017	34	Dufour et al. 2021
Central East China			36	
Beijing–Tianjin–Hebei region			39	
Yangtze River Delta			35	
Pearl River Delta			31	
North America, Europa and Est Asia (Korea et Japan)	3136 Rural sites	2010-2014	0-56 and more	Gaudel et al. 2018
North America, Europa and Est Asia	3348 Rural sites and 1453 urban sites	2010-2014	0-100 and more	Fleming et al. 2018
North America, Europa and Est Asia	Rural site	1996-2005	15-55	Young et al. 2018
North America, Europa and Est Asia	Rural and urban site	2010-2014	10-60	Schultz et al. 2017
Eastern North America	Rural site	2000-2014	26-38	Chang et al. 2017
	Urban site		28-38	

Several sites reported in Table 4 and Fig. 10 are exposed to high O_3 concentrations. These different levels observed in Africa and around the world are in most cases above values displayed in this study, with the exception of sites in southern Africa. The main reason for the discrepancies could be the proximity of these sites to O_3 precursor sources. They are generally located close to industrial, commercial and residential areas, not far from road traffic, garbage dumps, etc., where precursor emissions are high. Some sites may also be influenced by continental air masses containing gaseous pollutants. At urban sites such as Al-Ain, Bamako, Dakar, Abidjan and Cotonou, the low O_3 levels are due to the saturated NOx regime observed at these sites, which limits photochemical O_3 production (Adon et al., 2013; Bahino et al., 2018; Salem et al., 2009). At INDAAF sites, concentrations are lower because of their rural characteristics, generally far from anthropogenic sources, and much more influenced by biogenic activities from soils and vegetation. Results are fairly illustrative of the various mitigation or vigilance measures that need to be adopted to ensure the environmental well-being of each ecosystem. The additional efforts must therefore be made, through projects or programs, to densify monitoring networks for polluting gases in general and O_3 in particular, especially in Africa, where very few long-term monitoring exist.

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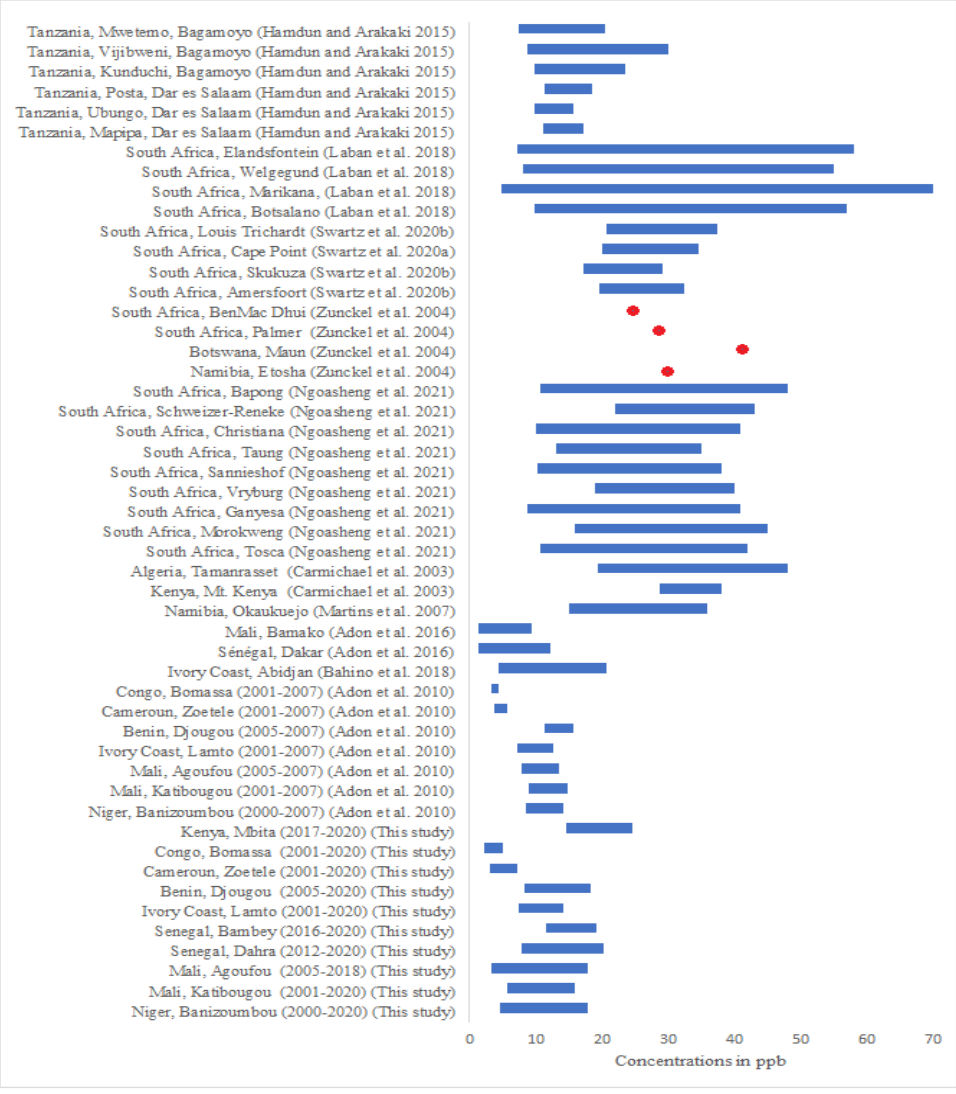

Figure 10. Overview of O_3 monitoring studies in Africa. Blue bars represent lower and upper range of means if reported. Red points represent average concentration of O_3.

3.3 Monthly variation in NOx and VOC anthropogenic and natural emissions

Tropospheric O_3 concentrations result from atmospheric chemistry involving precursors especially NOx and VOCs, and from transport (distance and time). Locating the sources of precursors (both biogenic and anthropogenic), and their amplitude, is therefore necessary to explain O_3 levels, on a local and regional scale.

3.3.1 NOx and VOC anthropogenic emissions

Monthly variation in anthropogenic NOx and VOC emissions are studied at all sites, with the exception of the Bb station, where emission data are not available (Fig. 11). In dry savanna, during the wet season, NOx and VOC fluxes are very low. On the other hand, maxima are observed in the dry season with the highest emissions found in Ka. Indeed, the monthly averaged

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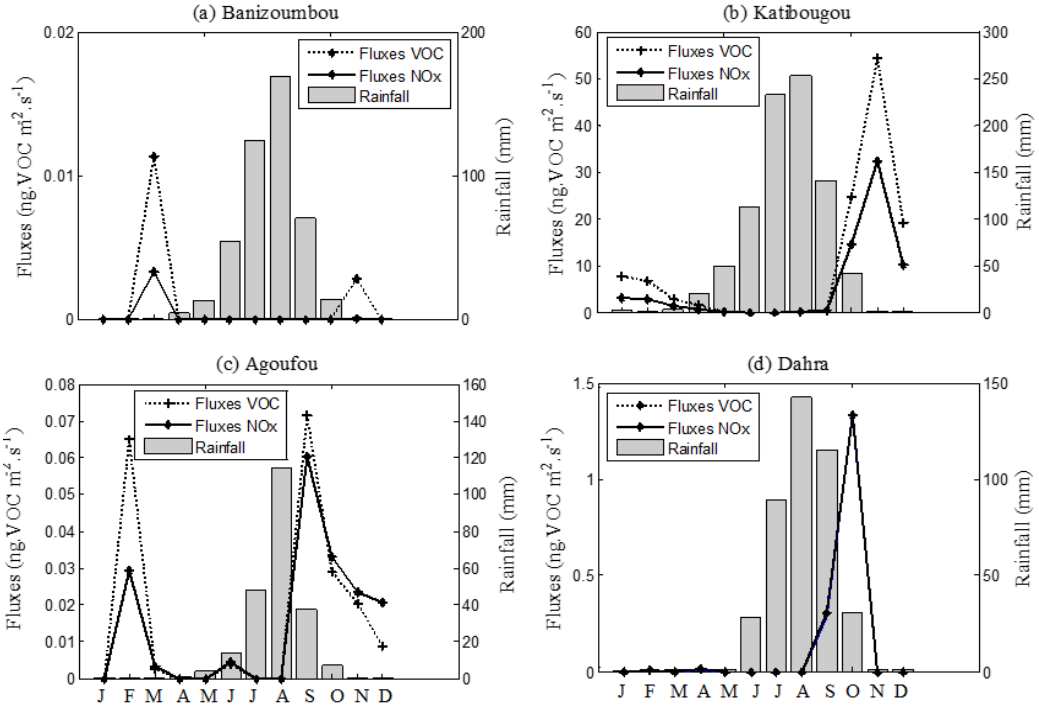
biomass combustion emissions (GFED4) over the 18-year period (1998-2015) in the Sahel show that Ka is significantly affected by the biomass combustion source in November (Ossohou et al., 2019). In wet savannas (La, Dj), and forest (Zo), NOx and VOC fluxes reach their maxima during the dry season. The mean flux estimates are respectively 14.5; 24.2; 4.9 $ng.m^{-2}.s^{-1}$ for NOx and 26.8; 29.4; 5.5 $ng.m^{-2}.s^{-1}$ for anthropogenic VOCs at La, Dj and Zo. High dry-season NOx emission rates observed are linked to biomass combustion sources (Abbadie, 2006; Adon et al., 2010; Akpo et al., 2015; Ossohou et al., 2019; Swartz et al., 2020b). In Bo, high NOx and VOC fluxes are observed in the wet season, unlike in Zo, corroborated by Ossohou et al (2019) over the period 1998-2015. The source of these recorded anthropogenic emissions could be biomass combustion. Indeed, according to the work of Sauvage et al. (2005), the period from August to September corresponds to a peak in biomass burning activity in the southern African countries (Mozambique, Zimbabwe, South Africa). Moving air masses over Central Africa via the northern edge of the continental anticyclone could explain such high emissions at Bo in August-September. NOx and VOC fluxes are quantifiable in February (dry season) at Mbita and are thought to be due to the combustion of biomass from agricultural activities and from biomass burning in savannas north of the site (South Sudan, Central African Republic and Democratic Republic of Congo) (Bakayoko et al., 2021; Boiyo et al., 2017b). At the South African sites, the mean fluxes vary from 0.3 $ng.m^{-2}.s^{-1}$ (LT) to 10.2 $ng.m^{-2}.s^{-1}$ (Sk) for NOx and from 2.9 $ng.m^{-2}.s^{-1}$ to 11.8 $ng.m^{-2}.s^{-1}$ (Sk) for VOCs with the maxima recorded in dry season. These values may be explained by significant anthropogenic emissions from domestic biomass burning and solid fuel combustion resulting in high levels of pollutants in South Africa's Highveld region (Kai et al., 2022). By developing an African regional inventory of anthropogenic emissions (wood and charcoal burning, charcoal manufacture, open waste burning etc.), Keita et al. (2021) estimate that Southern Africa is among the highest emitting regions for NOx, due to industrial sources and power plants. Using satellite data for NO_2 between 2005 and 2017, Oluleye (2021) has noted that nitrogen dioxide concentrations during the dry season were up to 200% higher than the values observed during the rainy season. The high NOx and VOC levels observed at the different sites during the dry season result from these different sources, releasing large quantities of precursors in the atmosphere, and not washed out by rainfall.



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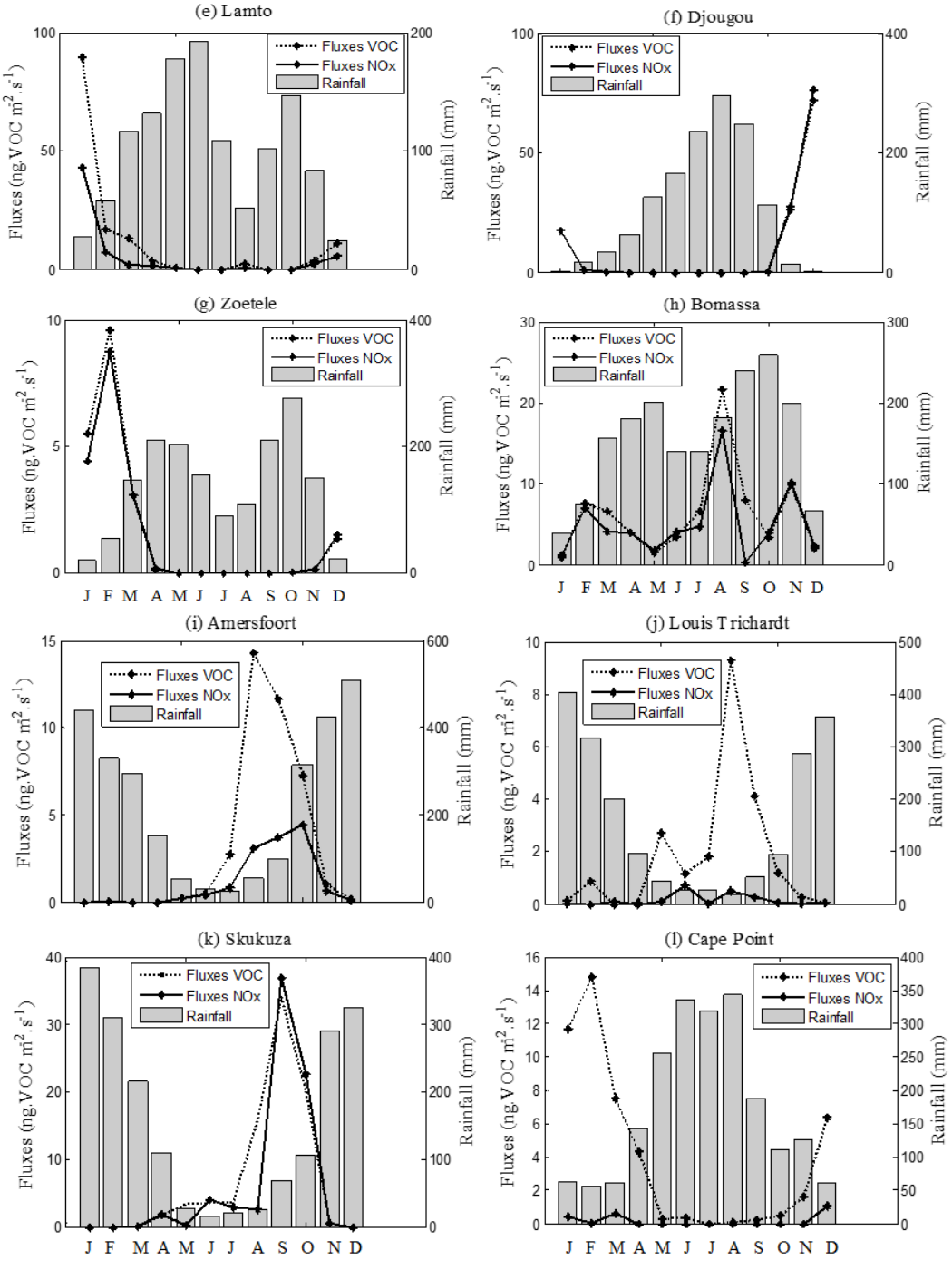

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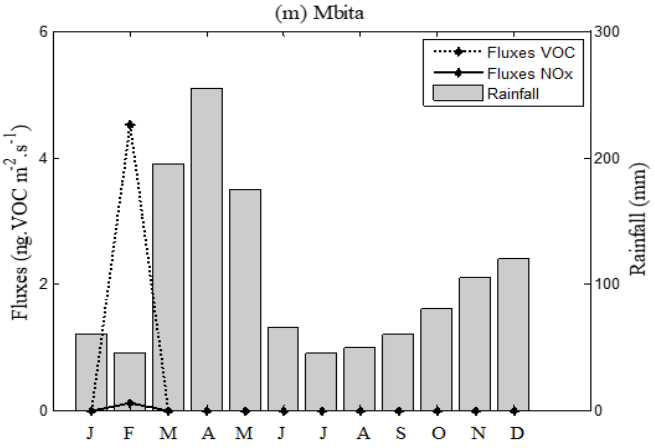

Figure 11. Mean monthly fluxes of NOx and VOC from biomass combustion estimated by the GFED4 inventory for 0.25° x 0.25° grid cells centered on each of dry savanna, wet savanna, forest and semi-Agricultural/arid savanna sites.

3.3.2 NOx and VOC natural emissions

Monthly variations in VOCs (α pinene, β pinene and isoprene) and biogenic NO around the studied sites are shown in Fig. 12. Biogenic NO fluxes in dry savanna show a bell-shaped variation, peaking in August (wet season). In wet savanna and forest, the highest values are also recorded during the wet season. Peaks range from 2.1 ± 0.1 (Bo) to 5.0 ± 0.6 $kgNha^{-1}year^{-1}$ (Ba) in the three ecosystems. These peaks of emission could be explained by the correlation between soil moisture, NO production in the soil and its release to the atmosphere, as shown in Delon et al., (2010), Galy-Lacaux et al., (2009) , Onojeghuo et al. (2017) and as illustrated in Sect. 3.2.1.1. The soil moisture content during the dry season is higher in wet savanna and forest than in dry savanna, which limits the NO pulse in these ecosystems (Delon et al., 2012). Monthly profile of BVOC fluxes (isoprene, α pinene and β pinene) in dry savanna shows a maximum at the end of the dry season/beginning of wet season at Ba, Ka and Ag, or during the wet season at Bb and Da. Isoprene fluxes are more obvious at Da (214.2 ± 30) $ng.m^{-2}.s^{-1}$ whereas α pinene, and β pinene exhibit larger values at Ka site (11,2 ± 1,8; 5.2± 0.8 $ng.m^{-2}.s^{-1}$ respectively). In La, Dj, Zo, Bo and Mb, BVOC maxima fluxes are also obtained at the end of the dry season/beginning of the wet season. A drop in these fluxes is then observed during the wet season. From dry savanna to forest, the transect gradient is positive for BVOC emissions, and is due to the abundance of vegetation in wet savanna (La) and forest. At Southern African sites, more specifically LT, Af and Sk, the highest values of BVOC are reached in the wet season: 189.6 ± 46.8 $ng.m^{-2}.s^{-1}$ (isoprene), 14.2 ± 2.7 $ng.m^{-2}.s^{-1}$ (α pinene) and 5.6 ±1.0 $ng.m^{-2}.s^{-1}$ (β pinene). At CP, the maximum emissions are measured in the dry months of January/February.

BVOC emission rates variation is a function of several factors, namely temperature, sunshine (Guenther et al., 1993), carbon dioxide (CO_2) concentration, precipitation, plant species (Jaars et al., 2016), crops, shrubs (Guenther et al., 1995), modulated on seasonal and interannual time scales by changes in land cover and canopy environment (Chen et al., 2018) as well as by climate change. In dry savanna, the peaks in BVOC observed at the end of the dry season (April/May) could therefore be due to strong radiation and high temperatures which could impact certain woody species (Zwarts et al., 2023), and cause biogenic emissions (Saxton et al., 2007). Indeed, woody species in grasslands are major sources of BVOC emissions (Jaars et al., 2016). Furthermore, positive correlations were obtained between isoprene emissions and air temperature (Jaars et al., 2016; Guenther et al., 1993; Saxton et al., 2007). In the forest, Serça et al. (2001) reported that the average ambient isoprene concentration for a tropical forest in northern Congo was the highest at the start of the rainy season and fell sharply at the end of the rainy season. At the Welgegund site (South Africa), which belongs to the same ecosystem as LT, Af and Sk, Jaars et al. (2016) also measured the highest BVOC concentrations during the rainy season as in this study, and increased BVOC concentrations were associated with high soil moisture. Gradual decrease in BVOC fluxes during the wet season observed at most of the sites is thought to be

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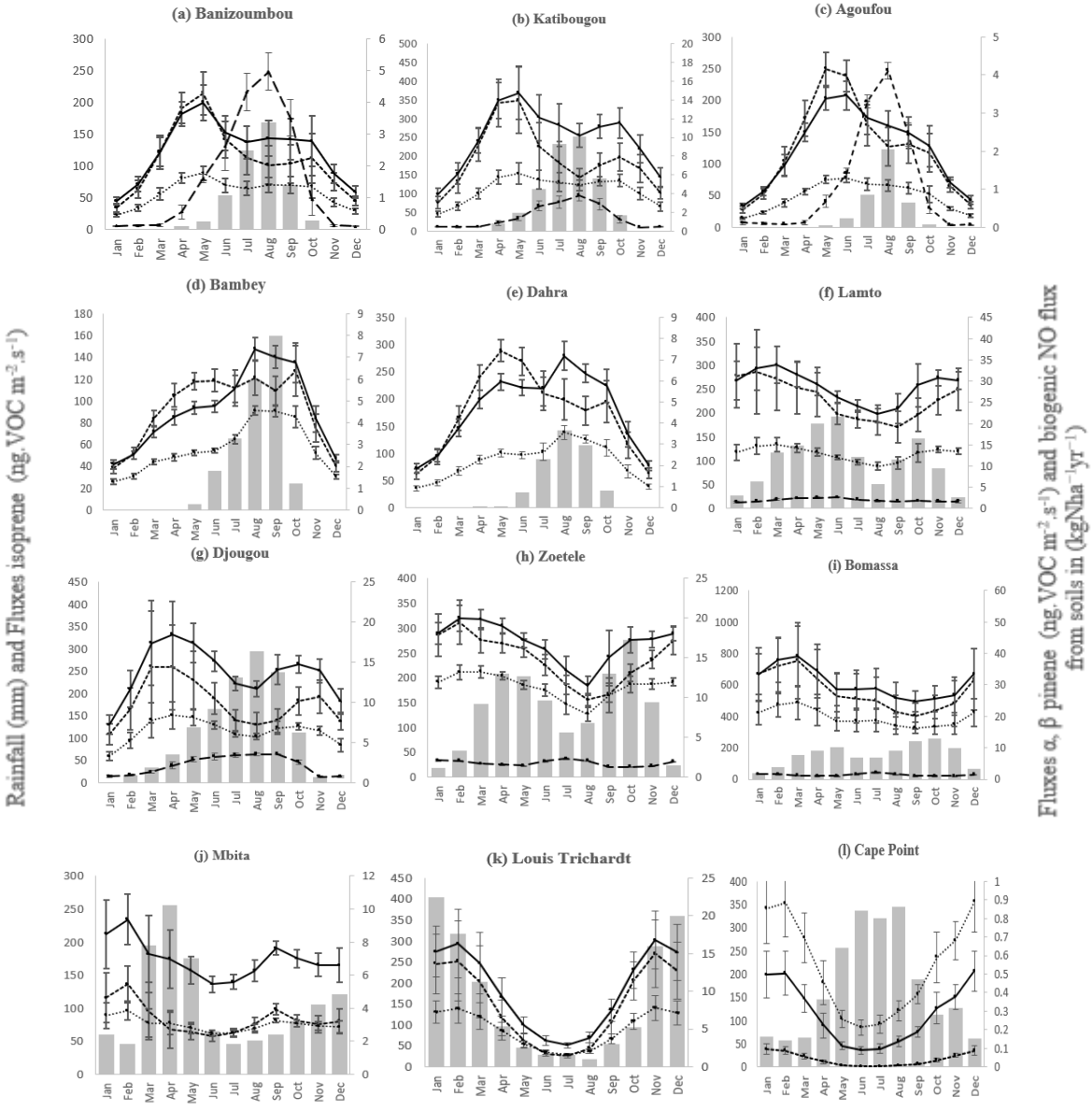
due to the uptake of VOCs by soils, thanks to the microbial activity that develops during this period. Soils could therefore play an important role as isoprene sinks (Gray et al., 2015).



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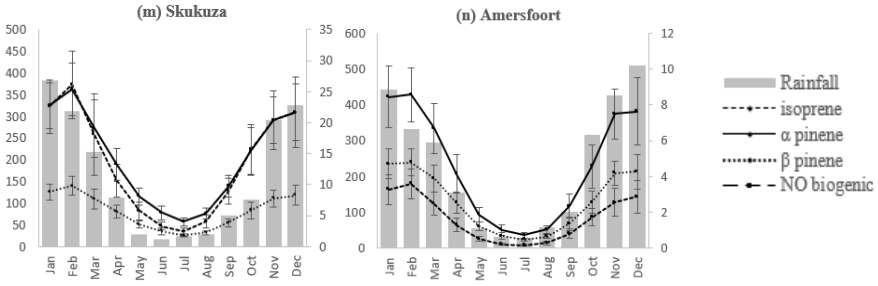

Figure 12. Mean monthly fluxes of BVOCs estimated by the MEGAN inventory for 0.25° x 0.25° centered meshes and biogenic NO in dry savanna, wet savanna, forest and semi-Agricultural/arid savanna.

3.4. Contributions of meteorological variables, NOx and VOC to O_3 production

In this section, we use PCA to estimate the relevant number of axes (Sect. 3.4.1) to identify the major sources of O_3 precursor emissions at each site, based on their contribution to the construction of the axis and the quality of their representation on the axis (Sect. 3.4.2).

3.4.1. Estimation of the inertia of the studied factors

At all sites, ten to eleven factors (meteorological variables and chemical pollutants) are studied. Each factor could correspond to a given axis. However, the cumulative inertia calculated indicated that the first two axes ranged from 79.9% (Ag) to 95.4% (Af) at all sites (Fig. 13). These values are well above the reference value of 54.2% (Tsuyuzaki et al., 2020). These axes therefore represent the main axes. The high inertia values obtained indicate that the factors studied are not independent and that there are strong links between them. The projection of the dataset onto the factorial plane (with two axes) is therefore a good approximation of the air quality at each site, as it contains most of the available information.

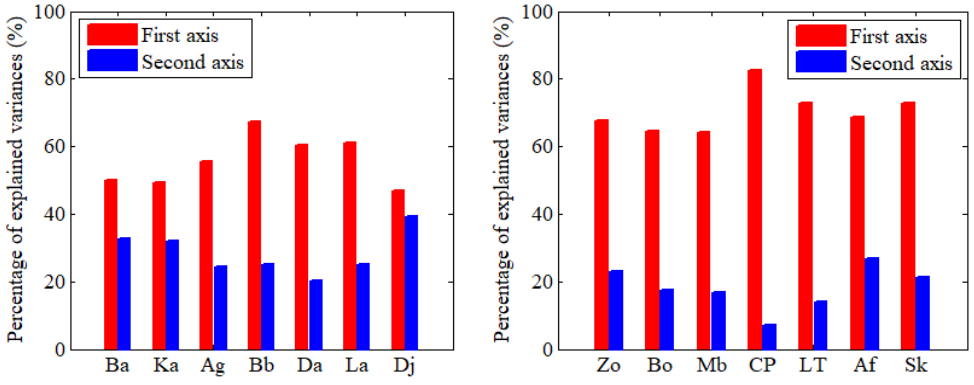

Figure 13. Decomposition of the total inertia of O_3 precursors on the factorial axes.

3.4.2. Characterisation of O_3 precursor emission sources and studies of correlations

3.4.2.1 Dry savanna

At the dry savanna sites, two groups of pollutants can be identified through their contribution to the two axes. O_3, α pinene, β pinene, isoprene and NO are strongly correlated with the first axis ($0.58 < r^2 < 0.98$) and NOx, VOCs anthropogenic fluxes correlate well with the second axis ($0.62 < r^2 < 0.92$). These results suggest the presence of two major sources of precursors: biogenic and anthropogenic activities. Calculating the Pearson correlation between the different pollutants, we observe a good

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dependence of O_3 only with all biogenic precursors ($0.51 < r < 0.95$) at Ba, Ka and Ag in the presence of high relative humidity and precipitation ($0.64 < r < 0.95$) (Table 5). On the other hand, at Bb and Da, isoprene correlates well with O_3 ($r = 0.79$). It appears as a dominant precursor under the influence of temperature and radiation, which are also well correlated with O_3 ($0.5 < r < 0.76$) (Table 5). The main contributions to the formation of the first axis are observed for BVOC (39.5% to 44.6%) with a good quality of representation (0.5 to 0.9) (Fig. 14). The contribution of biogenic NO is about 10%. The most relevant meteorological variables (humidity, temperature, precipitation, radiation) contribute significantly, ranging from 10.5% to 30.8%. In dry savanna, Oluleye et al. (2013) estimated that rain was responsible for 62% of the O_3 distribution in the West African region, excluding the precursors NO, CO and hydrocarbons, as also illustrated in our results. Saunois et al. (2009) have shown that soil NOx emissions, combined with the northward advection of volatile organic compounds (VOCs), play a key role in O_3 production in dry savanna regions. This large-scale impact of biogenic emissions has also been verified by Williams et al. (2009), who estimate that 2-45% of tropospheric O_3 over equatorial Africa may originate from NOx emissions from African soils. All these works are in agreement with the results of this study.

3.4.2.2 Wet savanna and forest

In wet savanna and forest, pollutants such as O_3, α pinene, β pinene, isoprene, anthropogenic NOx and VOCs (except at Bo and Dj) are positively correlated to the same axis ($0.62 < r^2 < 0.97$), while only NO is linked to the second axis. The major sources highlighted in this case could be anthropogenic activities and those linked to biogenic vegetation on the one hand, and biogenic soil activities on the other. Strong Pearson correlations are observed between O_3, NOx and the VOCs ($0.49 < r < 0.92$) (Table 5). Temperature and radiation are also well correlated with O_3 ($0.54 < r < 0.89$) in these two ecosystems. The dominant contributors are then BVOC (31% to 60.8%), temperature (11% to 12.1%), radiation (11.1% to 12.4%) (Fig. 14). Anthropogenic emissions at La and Zo make a significant contribution (5.8% to 10.2%). These results are corroborated by the literature. Indeed, radiation and humidity facilitate the propagation of radical chain reactions and the production of hydroxyl radicals (OH) at these sites (Graedel and Crutzen, 1993). According to several authors, O_3 levels tend to increase under warm, sunny conditions favorable to photochemical O_3 production (Hamdun and Arakaki, 2015; Morakinyo et al., 2020). Moreover, Aghedo et al. (2007), Mari et al. (2011), Saunois et al. (2009) and Saxton et al. (2007) have reported that vegetated areas emit large quantities of biogenic organic compounds that influence O_3 production in the presence of light and temperature.

3.4.2.3 Agricultural and semi-arid savanna

At the South African sites, the major sources of pollutant vary from one site to another. At Af, we identify a first group of pollutants (α pinene, β pinene, isoprene) strongly dependent on the first axis ($0.98 < r^2 < 0.99$), suggesting the presence of a source linked to biogenic activities originating from vegetation. On the other hand, the second group, comprising O_3, VOCs and NOx anthropogenic, indicates a source from combustion activities ($0.83 < r^2 < 0.89$), which favors O_3 photochemistry. At CP, LT and Sk sites, the pollutant category in which O_3 is found is linked to biogenic emissions (VOCs) and combustion sources. At these sites, temperature, humidity, precipitation and radiation are anti-correlated with O_3 ($-0.90 < r < -0.43$) (Table 5). High O_3 concentrations are therefore measured at these sites during the driest and coldest months (Swartz et al., 2020b). The main contributions are BVOCs, NOx and anthropogenic VOCs, temperature and rainfall at CP, VOCs, humidity, temperature at Sk, NOx and anthropogenic VOCs, and radiation at Af and anthropogenic VOCs, radiation, humidity at LT. These factors contribute between 5.1% and 39.4% (Fig. 14) to the formation of the axes, with a good representation quality (0.44 to 0.97) for most of them. Observations made at South African sites are confirmed by several recent studies. Indeed, Swartz et al. (2020b) attributed the decrease in O_3 concentrations from 1995 to 2001 to the decrease in NO_2 concentrations over this period at LT. The importance of humidity and temperature in O_3 photochemistry observed at almost all South African sites has been highlighted by Balachov et al. (2014), Laban et al. (2018, 2020). Laban et al. (2018) added that emissions from domestic combustion (in winter) and regional biomass combustion (in spring) are also responsible for the increase of O_3 levels. The contribution of VOCs to O_3 photochemistry in South Africa is confirmed by Jaars et al. (2016). At Mb, neither of the two

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main axes has a significant relationship with O_3, with a poor quality of representation on the main axes (0.14). Further studies on other precursors such as biogenic NO and carbon monoxide (CO) should be carried out, and biomass combustion on a continental scale should also be taken into account, in order to better assess the dominant chemical contributions to O_3 production at this site.

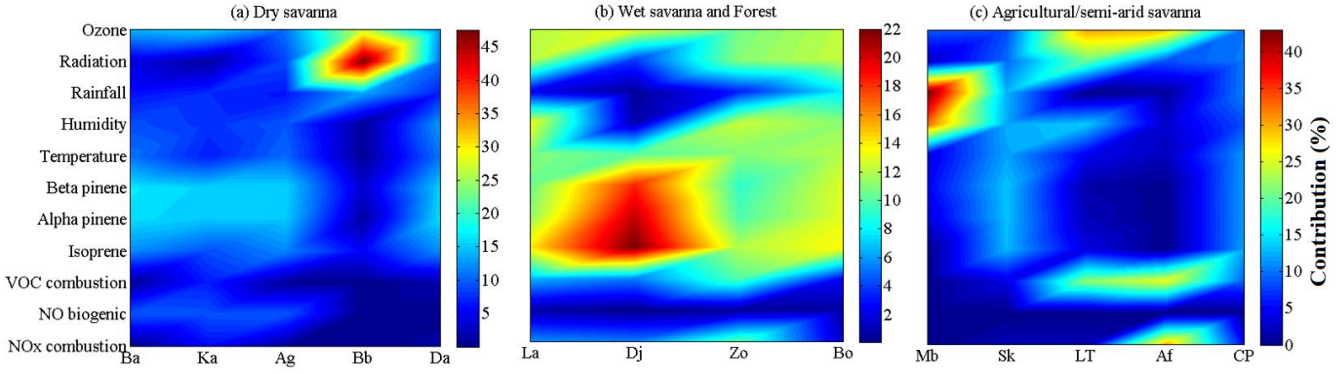

Figure 14. Contribution of different variables to photochemical O_3 pollution in (**a**) Dry savanna (**b**) West savanna and Forest (**c**) Agricultural/semi-arid savanna.

Table 5. Correlation r between O_3, its precursors and meteorological variables at different sites. Blank spaces in the table indicate the absence of data on this site for the precursor concerned over the study period.

Ecosystem	Dry savanna					Wet savanna		Forest		Agricultural/semi-arid savanna				
Sites	Ba	Ka	Ag	Bb	Da	La	Dj	Zo	Bo	Mb	LT	CP	Af	Sk
	O_3													
NO biogenic	0.85	0.73	0.92	-	-	-0.15	-0.24	0.43	-10^{-3}	-	-	-	-	-
NOx_C	-0.33	-0.43	0.04	-	0.001	0.49	0.059	0.80	-0.33	0.31	0.37	-0.67	0.62	0.61
VOC_C	-0.40	-0.47	-0.003	-	-5.10^{-4}	0.54	0.05	0.79	-0.40	0.31	0.60	-0.87	0.52	0.65
isoprene	0.51	0.54	0.46	0.79	0.78	0.92	0.81	0.80	0.92	0.42	-0.39	-0.91	0.29	-0.64
α pinene	0.67	0.76	0.66	0.45	0.77	0.74	0.64	0.63	0.90	0.36	-0.45	-0.86	0.29	-0.67
β pinene	0.70	0.79	0.72	0.34	0.72	0.70	0.56	0.60	0.89	0.33	-0.48	-0.85	0.27	-0.71
Temperature	0.45	0.47	0.42	0.51	0.76	0.72	0.69	0.68	0.89	0.47	-0.46	-0.90	0.49	-0.62
Humidity	0.82	0.64	0.95	0.52	0.70	-0.85	-0.19	-0.89	-0.79	-0.61	-0.79	-0.68	0.1	-0.80
Rainfall	0.74	0.64	0.75	0.15	0.51	-0.48	-0.39	-0.76	-0.54	-0.18	-0.49	074	053	-0.69
Radiation	0.16	0.15	0.26	0.75	0.69	0.73	0.54	0.65	0.87	0.21	-0.19	-0.76	0.71	-0.43

3.5 Annual and seasonal trends of O_3 and its precursors

3.5.1 Annual trends

Annual trends in O_3 concentrations were calculated according to Mann-Kendall test (Fig. 15). At the annual scale, Ka site in Mali shows a significant decrease in O_3 concentrations around -0.24 ppb yr^{-1} (-2.3 % yr^{-1}) from 2001 to 2020 (pvalue = 0.002) at the 95% confidence level. At the seasonal scale, this downward trend at Ka is confirmed in both the dry and wet seasons. O_3 concentrations decrease by -0.18 ppb (-2.1 % yr^{-1}; pvalue = 0.03) per year in the dry season and -0.33 ppb (-2.4 % yr^{-1}; pvalue < 0.01) in the wet season. At the same site, the trend in nitrogen oxide (NO_2) over the 1998-2020 period shows a decline in annual concentrations and annual seasonal means. Ossohou et al. (2019) observed a decrease in NO_2 concentrations at Ka

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in the wet season of -0.04 ppb yr^{-1} (-2.4 % yr^{-1}) over the period 1998-2015. These downward trends in NO_2 could therefore explain the downward trends in O_3. At the Ba site during the dry period, a significant downward trend is recorded, with O_3 decreasing by -0.15 ppb yr^{-1} (-1.9 % yr^{-1}; pvalue = 0.04). Calculation of trends on biogenic VOCs at Ba indicate a decrease in biogenic emissions of alpha pinene (τ = -0.37, p value = 0.020) and beta pinene (τ = -0.39, p value =0.01). Chen et al. (2018) indicated that trends in global tree cover from 2000 to 2015 have led to clear decreases, particularly in West Africa with a reduction of around 10% in regional BVOC emissions due to agricultural expansion. At the other sites, no significant trends were observed. The absence of annual trends at South African sites confirms the results obtained by Swartz et al. (2020a, 2020b) at the LT, Af, Sk and CP sites. Breaks in the annual concentration data were observed at Ba and Ka in 2006 as a result of the Pettitt test. During the dry and wet seasons, further breaks were recorded in the annual series in 2006 (Ka) and 2007 (Ba) (dry season) and in 2014 at Ka (wet season). However, no trend inversion was induced in these break years. At the other study sites, no breaks were observed.

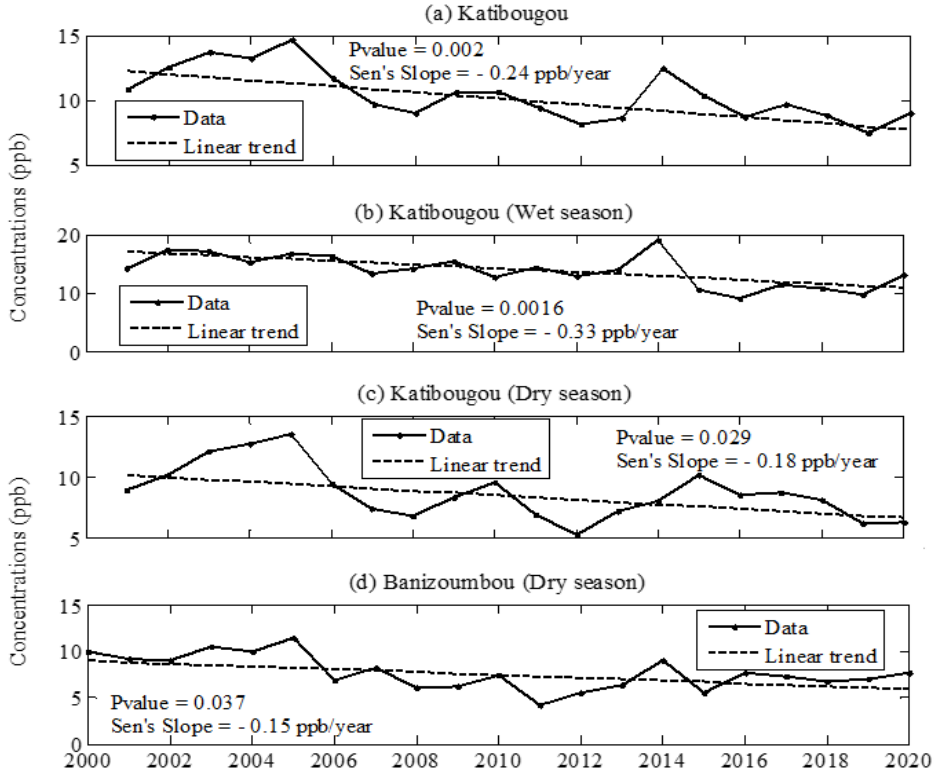

Figure 15. Significant long-term annual linear trend in in situ O_3 concentrations over the period 2000-2020

3.5.2 Seasonal trend

Trend tests are performed on monthly mean O_3 concentrations using seasonal Kendall test and significant trend results are presented in Fig. 16. The test reveals downward trends of -0.07 ppb yr^{-1} (-0.6 % yr^{-1}; pvalue = 0.02) at Ba and -0.24 ppb yr^{-1} (-2.3 % yr^{-1}; pvalue <0.001) at Ka for a 95% confidence level. A significant upward trend is reported at Zo in Cameroon (Sen slope = 0.1 ppb yr^{-1}; 1.4 % yr^{-1}; pvalue = 0.001), and at Sk in South Africa (Sen slope = 0.3 ppb yr^{-1}; 1.5 % yr^{-1}; pvalue = 0.001). To explain the trends observed at Zo, we apply the Kendall rank correlation between O_3 concentrations and its precursors. We obtain a significant positive rank correlation at the 95% confidence level. For NOx and anthropogenic VOCs,

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τ = 0.7 (pvalue= 0.03) while with biogenic VOCs, the correlation varies from 0.54 to 0.69 (pvalue <0.01). In addition, we observe increasing trends for biogenic VOCs. Isoprene increases by 1.8 $ng.m^{-2}s^{-1}$ per year (pvalue = 0.001), alpha pinene by 0.1 $ng.m^{-2}s^{-1}$ (pvalue < 0.001) and beta pinene by 0.04 $ng.m^{-2}s^{-1}$ (pvalue = 0.003). Similar trends were also observed in the wet season for alpha pinene and beta pinene, and in the dry season for isoprene. The significant rise in O_3 concentrations in Zo could therefore be explained by these increasing trends observed in isoprene, alpha pinene and beta pinene from one season to the next and in anthropogenic emissions in African forest regions. These results are corroborated by 18 years of satellite data (1998-2015) used by Andela et al. (2017) who noted an increasing trend in burned areas in dense canopy forests. At Sk, the upward trend is thought to be due to anthropogenic and biogenic emissions. At the Irene site in the North-eastern interior of South Africa, Bencherif et al. (2020) obtained an upward trend in the tropospheric O_3 column at a rate of around 2.4% per decade. In addition, an upward trend in NO_2 levels was also evident at Sk, signifying the influence of growing rural communities on the Kruger National Park border (Swartz et al., 2020b). This population growth and the associated increase in anthropogenic activities could therefore justify the upward trend obtained.

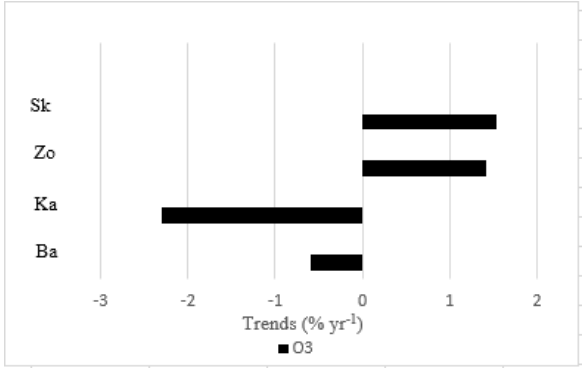

Figure 16 Significant Kendall's seasonal trend for O_3 Concentrations

4. Conclusion

This work presents an original database of long-term O_3 concentrations at fourteen African sites belonging to the INDAAF program and companion projects. This database gives a better understanding of O_3 concentration levels at remote tropical sites representative of the major African biomes. In this study, we establish a mean annual cycle by site and ecosystem type, and investigate the seasonal variability of O_3 concentrations over the period 1995-2020. Our analysis of the seasonality of anthropogenic and biogenic NOx and VOC emissions then highlights the significant factors contributing to O_3 formation. Finally, we calculate the O_3 long-term trends, which provide an insight into the long-term evolution of O_3 levels and the local and regional dynamics of the emission sources of its precursors.

The results indicated that O_3 levels are the highest during the rainy season in dry savannas and during the dry season in wet savannas and forests. In agricultural fields, no seasonal variations of O_3 concentrations are observed. In semi-arid savanna (South Africa), dry season O_3 levels are the highest at LT and Sk. At CP and Af sites, maxima occur during the rainy season. Mean annual O_3 concentrations range from 10.5±5.4 to 14.8±4.3 ppb in dry savannas, from 10.8±3.3 to 13.5±4.8 ppb in wet savannas, from 3.9±1.1 to 5.2±2.1 ppb in forest ecosystems and from 19.9±4.7 to 29.1±8.4 ppb in semi-arid/agricultural savannas. BVOC (under the influence of air temperature), NO emissions (in the presence of humidity) and precipitation, are the main contributors to O_3 formation in dry savannas. The seasonality of O_3 measurements and dominant precursors confirm the important role of microbial processes leading to high NO emissions at the beginning of the wet season for O_3 production. Furthermore, the influence of air temperature and solar radiation on woody emissions from shrubs in the Sahel, and the presence of sparse vegetation (short grasses, forbs and dicotyledonous shrubs with perennial ground cover) in this region could

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be at the origin of BVOC emissions. The photochemical O_3 regime in savannas and rainforests (heavily vegetated areas) is strongly linked to BVOC emissions from vegetation, and to temperature, radiation and humidity. At La and Zo, anthropogenic NOx and VOC also contribute to O_3 formation. The most dominant precursor species in southern Africa are mainly VOC emissions (anthropogenic and biogenic), humidity and temperature, as well as anthropogenic NOx at a few sites. They are due of Biomass and fuel combustion, large-scale transport of pollutants, domestic combustion in winter and biogenic emissions from vegetation. At INDAAF sites, which are rural sites far from many anthropogenic sources, O_3 concentrations are below most of the values reported in the literature. Annual and seasonal Mann-Kendall trends at all sites indicate that the Ka site in Mali and the Ba site in Niger experience a significant decrease in O_3 concentrations (around -2.3 % yr^{-1} and -0.6% yr^{-1}) over the period 2000 to 2020 justified by downward trends of NO_2 trends observed at Ka and the BVOC emissions at Ba. In contrast, a significant upward trend is reported at Zo (1.4% yr^{-1}) in Cameroon and Sk (1.5% yr^{-1}) in South Africa. These trends could be attributed to the increase in BVOCs in Zo and anthropogenic and biogenic emissions in Sk.

This study described in details the O_3 levels in representative African biomes, as well as the photochemical regimes and conditions leading to the observed concentrations. The results presented in this article constitute a robust database showing the importance of developing or maintaining long-term observation projects and observatories. This database could be used to assess the impact of O_3 dry deposition fluxes on African crops and the potential yield losses because of O_3 absorption by crops. An assessment of the various agricultural losses during the growing season will help to better orient actions to improve crop yield and achieve food security. In addition, this documentation is invaluable for modeling chemical processes in the atmosphere and for projecting future changes in tropospheric O_3. It could limit the uncertainties of these models and facilitate their validation, which is mainly based on data measured in situ.

Data availability. The INDAAF O_3 observations are available on the program website at https://indaaf.obs-mip.fr (https://doi.org/10.25326/608 ; https://doi.org/10.25326/604 ; https://doi.org/10.25326/610 ; https://doi.org/10.25326/609 ; https://doi.org/10.25326/606 ; https://doi.org/10.25326/275 ; https://doi.org/10.25326/605 ; https://doi.org/10.25326/603 ; https://doi.org/10.25326/607 ; https://doi.org/10.25326/642 ; https://doi.org/10.25326/646 ; https://doi.org/10.25326/645; https://doi.org/10.25326/644 ; https://doi.org/10.25326/647) . GFED4 (NOx, COV) and MEGAN-MACC (Isoprene, pinene-a, pinene-b) data are available from https://eccad.sedoo.fr/#/data. ERA5 reanalysis data are available from https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-single-levels?tab=overview.

Author contributions. CGL designed the study, wrote the protocol and edited the paper. HEVD conducted data processing, the statistical analysis and wrote the paper. CD made conceptual contributions and edited the paper. ABA and MO contributed at the statistical analysis, assisted in sample collection and edited the paper. VY, DL, MO-L, ON, PGVZ assisted in sample collection and edited the paper. EG and MDA analysed the samples.

Competing interests. The contact author has declared that none of the authors has any competing interests.

Disclaimer.

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