

Page Proof Instructions and Queries

Journal Title: The Holocene

Article Number: 678466

Greetings, and thank you for publishing with SAGE. We have prepared this page proof for your review. Please respond to each of the below queries by digitally marking this PDF using Adobe Reader (free at https://get.adobe.com/reader).

Please use only the circled tools to indicate your requests and responses, as edits via other tools/methods are not compatible with our software. To ask a question or request a formatting change (such as italics), please click the tool and then choose "Text Callout." To access the necessary tools, choose "Comment" from the right-side menu.



















No.	Query
	Please confirm that all author information, including names, affiliations, sequence, and contact details, is correct.
	Please review the entire document for typographical errors, mathematical errors, and any other necessary corrections; check headings, tables, and figures.
	Please ensure that you have obtained and enclosed all necessary permissions for the reproduction of artworks (e.g. Illustrations, photographs, charts, maps, other visual material, etc.) Not owned by yourself. Please refer to your publishing agreement for further information.
	Please note that this proof represents your final opportunity to review your article prior to publication, so please do send all of your changes now.
	Please confirm that the acknowledgement and funding statements are accurate.
1	Strikethrough texts in some of the words/phrases/sentences/paragraphs in the original MS have been removed. Please check.
2	'Marchant and Taylor (1998)' has been changed to 'Marchant and Taylor (2000)' to match the reference list. Please check and confirm.
3	'Robertson et al. (2006)' is not listed in the references. Please provide reference details or delete the citation.
4	'Gambaro et al. (2009)' has been changed to 'Gambaro et al. (2008)' to match the reference list. Please check and confirm.
5	Please provide the location details for 'Thermo Fisher Scientific' and 'Supelco'.
6	Please check whether the inserted expansions for 'DCM', 'BSTFA', 'TMCS' and 'RSD' are correct.
7	'Reznikov et al. (2015)' has been changed to 'Reznikov and Adzhiev (2015)' to match the reference list. Please check and confirm.
8	'Kiage (2006)' has been changed to 'Kiage and Liu (2006)' to match the reference list. Please check and confirm.
9	'Ashley et al. (2010)' is not listed in the references. Please provide reference details or delete the citation.
10	Please note that the reference details of 'Maley et al. (2012)' and 'Neumann et al. (2012)' appear to be identical except the author name. Please check.
11	'Stager et al. (2009)' is not cited in text. Please indicate where a citation should appear or delete the reference.
12	'Stager et al. (2005)' is not cited in text. Please indicate where a citation should appear or delete the reference.
13	Please provide publisher details and page range for 'Thompson and Young (1999)'.
14	Please note that the given URL in 'Weninger et al. (2011)' does not lead to the desired web page. Please provide an active URL.
15	Please provide an appropriate caption for Table 1.
16	Please provide an appropriate caption for Table 2.



Fire and human record at Lake Victoria, East Africa, during the Early Iron Age: Did humans or climate cause massive ecosystem changes during the Early Iron Age in East Africa?

The Holocene
I-II
© The Author(s) 2016
Reprints and permissions:
sagepub.co.uk/journalsPermissions.nav
DOI: 10.1177/0959683616678466
hol.sagepub.com

\$SAGE

Dario Battistel, 1,2 Elena Argiriadis, 1 Natalie Kehrwald, 2,3 Maddalena Spigariol, 1 James M Russell 2 and Carlo Barbante 1,2 [AQ: 1]

Abstract

Organic molecular markers determined in a sediment core (V95-IA-IP) from Lake Victoria (East Africa) were used to reconstruct the history of human impact and regional fire activity during the Early Iron Age (~2400 to ~1100 yr BP). Fire history was reconstructed using levoglucosan and polycyclic aromatic hydrocarbons (PAHs) as markers for biomass burning that demonstrate two distinct fire periods peaking at 1450–1700 and 1850–2050 cal. yr BP. A partial correlation between levoglucosan and PAHs is interpreted as different transport behaviors and burn temperatures affecting the proxies. A fecal sterol index (CoP-Index) indicates the presence of humans near the lakeshore, where the CoP-Index lags a few centuries behind the fire peaks. The CoP-Index peaks between 1850 and 1950 cal. yr BP and between 1400 and 1500 cal. yr BP. Retene, a PAH that indicates softwood combustion, differs from other PAHs and levoglucosan by abruptly increasing at ~1650 cal. yr BP and remaining high until 1200 cal. yr BP. This increase may potentially signal human activity in that the development of metallurgy and/or ceramic production requires highly efficient fuels. However, this increase in retene occurs at the same time as severe drought events centered at ~1500 and ~2000 yr BP where the droughts and associated woodland to grassland transition may have resulted in more intense fires. The grassland expansion could have created favorable conditions for human activities and triggered settlement growth that in turn may have created a positive feedback for further landscape opening.

Keywords

biomarkers, East Africa, fire activity, human impact, Iron Age, Lake Victoria

Received I August 2016; revised manuscript accepted 30 September 2016

Introduction

The African Great Lakes region has an extensive human history with known human habitation for thousands of years. However, the connections between society and a changing climate during the East African Iron Age are not well known. Increased fire activity and grassland expansion is documented in the literature beginning approximately 2500 yr BP (Colombaroli et al., 2014; Finch and Marchant, 2011; Nelson et al., 2012; Thevenon et al., 2003; Vincens et al., 2005) which coincides with regional human migration. Bayon et al. (2012) argue that Bantu colonists caused major vegetation changes centered around ~2500 yr BP, while Maley et al. (2012) ascribe the regional 'rainforest crisis' to natural climatic factors. Here, we use biomarkers in Lake Victoria sediment cores to examine the following hypothesis: Did the natural increase in fire activity create open spaces that consequently encouraged human settlements or could a small group of humans trigger large fires that, in turn, created open spaces and thereby promote the growth of human societies?

During the Iron Ages, the development of increasingly structured and technologically advanced societies may have significantly impacted the environment through land use for agricultural and herding activities. The Early Iron Age in East Africa is defined as ~2400 to ~1100 yr BP, coincident with the shift of the ceramic tradition from Urewe to roulette decorated ceramics (Ashley, 2010; Sinclair et al., 1993). The term 'Iron Age' is generally used for convenience since significant regional and/or chronological variations occur in East Africa (Ashley, 2010). Archaeological and historical evidence demonstrated that human presence during this period was mainly because of the migrations of Bantu-speaking

Corresponding author:

Dario Battistel, Department of Environmental Sciences, Informatics and Statistics, Ca' Foscari University of Venice, Dorsoduro 2137, 30123 Venice, Italy.

Email: dario.battistel@unive.it

Department of Environmental Sciences, Informatics and Statistics, Ca' Foscari University of Venice, Italy

²Institute for the Dynamics of Environmental Processes – CNR, Ca' Foscari University of Venice, Italy

³Geosciences and Environmental Change Science Center, U.S. Geological Survey, USA

⁴Earth, Environmental and Planetary Sciences, Brown University, USA

people (Huffman, 1989; Li et al., 2014; Russell et al., 2014). As a consequence, the increase in human activities combined with the development of metallurgy (Bower, 1991; Schmidt and Childs, 1995) led to an increase in population (Vansina, 1995) and resulting forest clearance from slash-and-burn practices (Marchant and Taylor, 2000; Vincens et al., 2003). [AQ: 2]

Possible anthropogenic impacts on the environment are, however, superimposed on a period of severe drought throughout tropical East Africa, as suggested by lacustrine and terrestrial records (Chritz et al., 2015; Halfman et al., 1994; Ricketts and Johnson, 1996; Russell and Johnson, 2005). Sediment, pollen and diatom records indicate a diffuse expansion of grassland and drier montane forests of *Podocarpus* trees between 3000 and 1700 yr BP in the Lake Victoria region (Kendall, 1969; Ssemmanda and Vincens, 2002; Stager et al., 2003), and a similar expansion of dry montane forests was also observed in sedimentary pollen records from the neighboring Albertine Rift sites, from 4100 to 900 yr BP (McGlynn et al., 2013).

Drier climate conditions limit primary production of herbaceous litter but at the same time promote efficient burning of grassdominated savannahs when dry fuel is available (Colombaroli et al., 2014; Nelson et al., 2012; Thevenon et al., 2003). Regional, annual, and interannual natural fire frequency is a result of the latitudinal movement of the intertropical convergence zone (ITCZ), which allows vegetation growth during the rainy seasons and favors burning events throughout the dry season from June to September (Pyne, 2001). Past East African fire regime reconstructions based on pollen and charcoal records showed a decrease in fire activity since ~5000 yr BP because of limited fuel, followed by a widespread increase since ~2000 yr BP (Colombaroli et al., 2014; Finch and Marchant, 2011; Nelson et al., 2012; Rucina et al., 2009; Thevenon et al., 2003; Vincens et al., 2005). Although grassland expansion and increased fire activity are sometimes ascribed to anthropogenic activities (Finch and Marchant, 2011; Kendall, 1969; Stager and Johnson, 2000; Talbot and Laerdal, 2000; Thevenon et al., 2003; Vincens et al., 2005), scientists actively debate whether the influence of humans on East African fire during the Iron Age and a shift in the ITCZ resulting in drought are the primary causes of fire activity during this period.

Previous studies (Cockerton et al., 2015; D'Anjou et al., 2012; Schüpbach et al., 2015; Zou et al., 2000) proposed the use of molecular markers in lake sediments or soil for obtaining information on the evolution of human settlements in relation to herding and fire activities. These methods are based on determining different classes of chemical compounds present in the organic fraction of sediments. Among these markers, polycyclic aromatic hydrocarbons (PAHs) and monosaccharide anhydrides (MAs) are molecular proxies for fire (Gambaro et al., 2008; Kuo et al., 2011; Robertson et al., 2006; Simoneit, 1999, 2002; Simoneit et al., 1993). [AQ: 3][AQ: 4] Levoglucosan (Lvg), the most representative MA, is considered a source-specific proxy for biomass burning since it is generated solely during cellulose and lignin combustion (Simoneit, 2002; Zangrando et al., 2013). PAHs are less specific markers, except for the three-ring retene which is a specific marker for coniferous wood combustion (Fine et al., 2002; Hays et al., 2002, 2011; Lu et al., 2013; Muri et al., 2003; Ramdahl, 1983; Schauer and Cass, 2000; Vincente et al., 2011; Wakeham et al., 1980) or the low-temperature transformation of chemical precursors from coniferous plants (Muri et al., 2003; Wakeham et al., 1980). Pollen grains of crop plants, soil erosion, and archaeological evidence indicate the anthropogenic influence on fire activity and landscape (Dotterweich et al., 2012; Moskaldel Hoyo et al., 2015).

The use of fecal sterols (FeSts) as specific molecular markers for human and/or livestock presence (D'Anjou et al., 2012) is an attractive alternative to other discontinuous and fragmented historical indicators. While FeSts can demonstrate the presence of



Figure 1. Map of East Africa, showing the location of Lake Victoria and the position of the collected sediment core V95-1A-1P used in this study.

ruminants, and thereby can be used to determine past herding, a large population of wild grazers including animals present in East Africa may also affect certain FeSt concentrations. FeSts can be determined in the same sediment sample as other molecular markers, thus ensuring the coincident timing of the recorded signals, even when core dating presents uncertainty. The stability of these compounds in sediments after burial is supported by studies where significant concentrations of Lvg, FeSts, and PAHs are present in soil and lacustrine sediments with ages older than 10 kyr BP (D'Anjou et al., 2012; Johnsen et al., 2005; Schüpbach et al., 2015), thus suggesting that degradation, if it occurs, is a low-kinetic process in these archives.

The use of these molecular markers for paleoenvironmental reconstructions is promising. For example, D'Anjou et al. (2012) were able to reconstruct human migration in relation with environmental changes during the last 7000 yr BP in Southern Norway, while Schüpbach et al. (2015) correlated the fire history of Petén Itza (Guatemala) with Mayan agricultural activities. However, to the best of our knowledge, Lvg and FeSts have never been studied in an African sequence.

We use a multi-biomarker method to analyze PAHs, FeSts, and Lvg in one section of the piston core V95-1P collected in 1995 from Lake Victoria during the *International Decade for the East African Lakes* (IDEAL) multidisciplinary project (Berke et al., 2012; Johnson et al., 2000; Johnson and Odada, 1996). We investigate relative changes in different biomarker records to examine the role of humans in the increased fire activity during the Early Iron Age in East Africa. This time period encompasses both dramatic vegetation shifts from forests to grasslands, increased fire activity, and the migration of Bantu-speaking populations, but the connections between climate, land use, and human history are not yet clear.

Study site

Lake Victoria is located at 1134 m a.s.l. It is the largest African lake (Figure 1) with a surface area of ~69,000 km² and a maximum water depth of~80 m (Crul, 1995; Johnson et al., 2000; Stager et al., 2003). Core V95-1A-1P was recovered from a water

depth of 65 m at (1°13.9′S, 33°11.9′E). The lake is located between the eastern and western branches of the East African Rift System and straddles the equator (Cockerton et al., 2015). The Kagera River and small tributaries are the principal inflow, and the only outflow occurs at Jinja (Cockerton et al., 2015). Therefore, Lake Victoria acts as a huge pluviometer as the lake level is mainly regulated by the precipitation—evaporation (P:E) balance (Johnson et al., 2000).

Methods

Samples and age model

A total of seven piston cores of different lengths were collected from Lake Victoria in the multidisciplinary IDEAL project (1995), where the cores were later curated in refrigerated storage by the US National Lacustrine Core (LacCore) Facility at the Limnological Research Center, University of Minnesota. In this study, we use section 1 (0–45 cm) from the V95-IA-1P core. Details on the coring are reported at http://www.ngdc.noaa.gov/geosamples/showsample.jsp?fac=LacCore&cru=Victoria&smp=Victoria-LV95-1A&dev=downhole%20coring&inst= and in the literature (Berke et al., 2012; Johnson et al., 2000). The sediment core was subsampled into 1-cm samples taken every other centimeter, resulting in a total of 20 samples. Samples were shipped to the University of Venice, where they were freeze-dried, milled, homogenized, and stored at -20° C until extraction.

V95-1P section 1 spans the interval from 2400 to 1200 cal. yr BP, using the age-depth model proposed in the literature (see Figure 1B in Berke et al., 2012; Johnson et al., 2000) and data available at ftp://ftp.ncdc.noaa.gov. As reported, all ages were calibrated using CalPal (Weninger et al., 2011), and since the uppermost sediments were not recovered, the core top was estimated to date back to ~1200 cal. yr BP, where the more recent dating point (5 cm depth) date back to 1342 ± 49 cal. yr BP (Johnson et al., 2000). The sedimentation rate of ~500 mm 1000 yr⁻¹ observed in V95-1P is consistent with sedimentation values obtained in parallel cores (V95-2P and V95-3P) that resulted in rates of 700 and 600 mm 1000 yr⁻¹, respectively (Talbot and Laerdal, 2000). Holocene Lake Victoria sediments, including those studied here, are fine-grained, dark diatomaceous mud (see also Talbot and Laerdal, 2000; Verschuren et al., 2002) rich in organic matter (~20%) with high C/N ratios (15-20) that may result from predominantly phytoplankton source (Johnson et al., 2000).

Chemical analysis

In this study, we analyzed three different classes of compounds: PAHs, FeSts, and MAs. A detailed list of each compound is reported in Table 1. Standard PAH native compounds were purchased from Dr Ehrenstorfer GmbH (Germany), and FeSts and Lvg were obtained from Sigma Aldrich (St Louis, MO). The determination of each compound was carried out using the internal standard method, employing ¹³C-labeled standards: ¹³C₆-acenaphthylene and ¹³C₆-phenanthrene for PAHs (¹³C-PAH), purchased from Cambridge Isotope Laboratories, Inc (Andover, MA), ¹³C₆-Lvg (Cambridge Isotope Laboratories, Inc), and ¹³C₃-cholesterol (Sigma Aldrich).

Table I. [AQ: 15]

Class of compound	Name	Abbreviation
PAHs	Naphthalene	Nph
	Acenaphthylene	Acy
	Acenaphthene	Ace
	Fluorene	Flu
	Phenanthrene	Phe
	Anthracene	Ant
	Fluoranthene	Fla
	Benzo(a)anthracene	BaA
	Chrysene	Chr
	Retene	Ret
	Pyrene	Pyr
FeSt	Coprostanol	CoP
	epi-Coprostanol	e-CoP
	Cholesterol	Chol
	5α-Cholestanol	5α -Ch
	Stigmastanol	Stg
MAs	Levoglucosan	Lvg

Uppsala, Sweden). The cleanup and separation of three fractions (PAHs, FeSt, and Lvg) was performed using solid phase extraction cartridges (Discovery SPE DSC-Si silica tube 12 mL, 2 g; Supelco). Cartridges were topped with aluminum oxide (~1 g) and anhydrous sodium sulfate (~1g) for removing trace water in the extracts. The cartridges were previously cleaned and conditioned with 40 mL DCM:Hex 1:1. The pre-concentrated sample was loaded onto the cartridge, and the PAH fraction was eluted with 20 mL DCM:Hex 1:1, FeSt fraction with 70 mL DCM, and Lvg fraction with 20 mL acetonitrile. The FeSt and Lvg fractions were evaporated to dryness, and the residues were redissolved in DCM and pyridine, respectively. Prior to the analyses by gas chromatography-mass spectrometry (GC-MS), FeSts and Lvg samples were derivatized by adding 100 µL of N,O-bis(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylsilyl chloride (TMCS) and heated at 70°C for 1 h. We created and tested extraction blanks after every three sample extractions to ensure the absence of contaminants prior to analyzing the samples.

Concentrations are converted into flux values by considering the sedimentation rate (500 mm 1000 yr⁻¹) obtained from the age depth model (Berke et al., 2012; Johnson et al., 2000)

Instrumental analysis

PAHs, FeSt, and Lvg fractions were analyzed by GC-MS (GC: 7890A GC System; MS: Agilent 5975C; Agilent Technologies, Santa Clara, CA), using the analytical methods from Gambaro et al. (2004), Battistel et al. (2015), and Medeiros and Simoneit (2007). Separation was performed on a HP5-MS column (60 m length, 0.25 mm inside diameter, and 0.25 µm film thickness; Agilent Technologies). Helium was used as a carrier gas with a flow of 1 mLmin⁻¹ where 2 µL of each sample was injected in split/ splitless mode (splitless time 1.5 min). The temperature program was designed and optimized for each single fraction as follows: PAHs: 70°C (held for 1.5 min) up to 150°C with a rate of 10°C min⁻¹ and then increased to 300°C at 3°C min⁻¹ and held for 15 min. FeSt: 150°C (held for 1 min) up to 220°C with a rate of 30°C min⁻¹ and then increased to 300°C at 2°C min⁻¹ and held for 10 min. Lvg: 110°C (held for 5.5 min) up to 210°C with a rate of 15°C min⁻¹ and then increased to 220°C at 2°C min⁻¹ and held for 5 min. An electron ionization source was used for mass detection. Single-ion monitoring mode was performed for quantification, based on target ions and qualifiers. Target (qualifiers) m/z ratios for each compound were as follows: Nph: 128, Acy: 152, Ace: 154, Flu: 166, Phe: 178, Ant: 178, Fla: 202, Pyr: 202, BaA: 228,

Chr: 228, Ret: 234, CoP: 370 (215), e-CoP: 370 (215), Chol: 368 (353), 5α -Ch: 460 (355), Stg: 473 (215), and Lvg: 204 (217). The uncertainty of each single measurement was estimated as the uncertainty of the analytical method from extracting four replicates of diatomaceous earth fortified with low (10 ng) and high (100 ng) amounts of native standard compounds. The uncertainty of the method resulted in 9–16% for PAHs, 12–18% for FeSt, and 13–16% for Lvg.

Results

Lvg is a specific marker for biomass burning and is widely considered as a proxy for regional fire activity (Kirchgeorg et al., 2014; Simoneit, 2002) since it is produced by cellulose and lignin combustion (Kuo et al., 2011; Robinson et al., 2006; Simoneit et al., 1993). Along the core section, Lvg flux values in Lake Victoria (Figure 2) ranged from 1 to 8 ng cm⁻² yr⁻¹. These values are slightly higher than, but comparable to, other Holocene sediment records, such as Lake Petén Itzá (Guatemala), where MAs fluxes ranged from 0.5 to 3 ng cm⁻² yr⁻¹ (Schüpbach et al., 2015). Figure 2 demonstrates higher Lvg values in two distinct zones between 14-20 and 31-40 cm (corresponding to about 1500-1700 and 1850-2050 cal. yr BP, respectively) with the maximum values observed at 14 and 32 cm (~1550 and ~1900 cal. yr BP) followed by a sharp decrease. In these two zones, the Lvg signal is significantly different from the rest of the core, as highlighted by the corresponding gray error bands in Figure 2. All Lvg and PAH values are reported as fluxes in order to allow a direct comparison with other records (Figure 2).

Individual PAH values at different depths are reported in Table 2. Among the investigated PAHs, low-molecular-weight Nph, Acy, Ace, Flu, and Phe are the most abundant and constitute 65–90% (average 82%, relative standard deviation (RSD) 7%) of the total PAH signal measured, while high-molecular-weight PAHs are less abundant. Several diagnostic ratios, such as Ant/(Ant+Phe), have been proposed for assessing combustion source type since Ant/(Ant+Phe) ratios >0.10 are associated to pyrogenic sources rather than petrogenic (Yunker et al., 2002). The majority of samples had ratios >0.10. In samples where diagnostic ratios were below 0.1, these values were still quite close to this indicative threshold.

The sum of individual PAH concentrations in Lake Victoria sediments varied from hundreds to thousands of nanograms per gram. In general, total PAH fluxes (Σ PAHs) in Lake Victoria (Figure 2) significantly increased between 6 and 16 cm (1400–1600 yr BP), where this trend is similar to Lvg.

Lvg and ΣPAHs records reported in Figure 2 do not significantly correlate (r = 0.434, p = 0.055). This difference suggests that these proxies, although they are both indicative of fire, do not necessarily describe the characteristics and aspects of the same fire event. The increasing number of rings in PAHs relate to burning temperatures, where the greater the number of rings, the higher the burning temperature required (McGrath et al., 2003). In contrast, Lvg is produced at combustion temperatures centered around 250°C, but concentrations diminish at higher temperatures (Kuo et al., 2011). Thus, since fire intensity affects the pyrolitic by-products, $\Sigma PAHs$ and Lvg do not strictly correlate. However, Lvg flux correlates better with the sum of the low-molecularweight PAHs (Nph+Acy+Ace+Flu; r = 0.523, p = 0.018) rather than with the sum of high-molecular-weight PAHs (Phe+Ant+Fl a+Pyr+BaA+Chr+Ret; r=0.105, p=0.661). This correlation is consistent with the occurrence low-temperature fire events. Although PAHs do not unambiguously differentiate between grass and wood fires, the higher abundance of low-molecular-weight PAHs instead of high-molecular-weight PAHs suggests a possible predominance of grass rather than wood fires, with a possible increase in grass fires during the last ~400 years of the record.

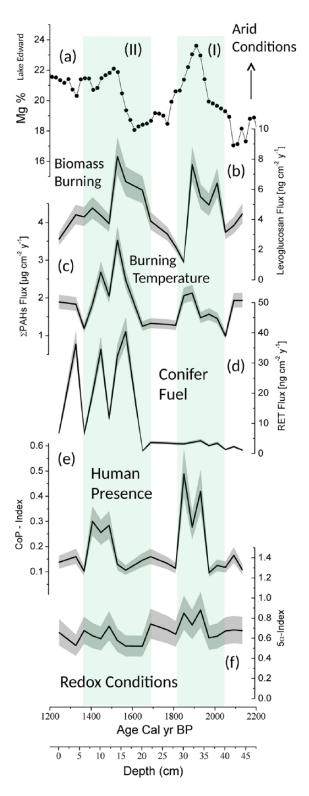


Figure 2. (a) %Mg in calcite from Lake Edward (from Russell and Johnson, 2005). Organic molecular proxy records from V95-1A-1P (Lake Victoria): (b) levoglucosan, (c) Σ PAHs, (d) retene, (e) CoP-Index calculated as (CoP+e-CoP)/Chol, and (f) 5α -Index calculated as 5α -Ch/Chol.

Retene is one of the few source-specific PAHs. The retene profile differs from the other PAHs in that it demonstrates negligible except for at 20 cm (~1650 yr BP), where a significant, although fluctuating, increase was observed. The increase in retene could indicate a possible change in fire source, as retene derives from abietic acid-like structures, and it is a marker for conifer wood combustion (Fine et al., 2002; Hays et al., 2002, 2011; Lu et al., 2013; Muri et al., 2003; Schauer and Cass, 2000; Vincente et al.,

<mark>16]</mark>	
[AQ:	
Table 2.	

Depth (cm)	Age (cal. yr BP)	Class of	Class of compound															
		PAHs											FeSts					MAs
		Concenti	Concentration $({\sf ngg^{-1}})$	(1-														
		Nph	Acy	Ace	Flu	Phe	Ant	Fla	Pyr	BaA	Chr	Ret	CoP	e-CoP	Chol	5a-Ch	Stg	Lvg
0.5	1252	236.2	40.5	165.5	253.0	564.9	115.7	323.8	136.3	9.3	37.1	7.0	110.4	21.6	1129.1	739.5	1332.3	238.3
4.5	1332	249.1	71.0	378.4	380.0	465.9	56.4	120.6	45.3	8.4	25.5	32.9	225.0	29.1	1850.8	976.3	1391.7	380.7
6.5	1373	313.5	47.5	195.6	154.5	292.8	39.9	95.4	31.8	4.8	7.0	6.5	65.1	36.7	1209.1	816.8	1280.0	368.0
8.5	1413	393.9	9.06	462.7	234.5	454.0	51.0	85.0	24.9	8.0	5.1	18.3	407.4	29.8	1644.8	1026.6	1566.4	419.8
10.5	1453	314.7	65.5	266.9	415.6	0.686	131.0	218.5	94.2	9.61	132.9	31.4	516.8	54.8	2538.4	1511.1	1883.6	377.3
12.5	1494	368.1	83.6	481.4	270.6	512.6	73.2	155.4	52.4	12.5	27.5	Ξ	300.5	27.5	1305.7	939.9	1448.0	329.4
14.5	1534	756.5	110.0	678.0	1063.0	554.8	58.9	161.9	61.5	18.7	34.5	29.5	156.3	51.3	1901.5	0.9601	1782.2	720.6
16.5	1574	369.5	104.4	499.5	497.3	200.0	87.8	139.8	65.2	7.0	168.2	36.5	133.8	53.9	2136.4	1116.4	1637.1	574.7
20.5	1655	186.3	55.1	195.4	229.5	268.1	54.9	123.9	39.1	4.	82.7	9.1	196.4	39.5	1900.7	989.3	1420.9	525.4
22.5	1695	232.5	1.65	373.4	171.3	318.2	54.0	47.5	15.9	27.7	20.5	4.2	6.911	14.9	962.1	713.6	1207.9	343.4
26.5	1776	159.3	54.0	315.7	228.2	318.0	33.0	93.4	29.9	26.8	35.5	4.0	112.6	15.0	1130.7	772.7	1163.2	268.2
28.5	1816	86.2	48.8	258.1	285.1	280.4	49.5	139.4	43.9	26.0	50.4	3.9	108.4	15.1	1299.2	831.7	1118.5	193.0
30.5	1857	179.2	8	427.5	541.0	478.3	1.65	153.1	48.7	31.5	1.95	3.8	198.3	26.0	511.1	434.4	973.0	103.5
32.5	1897	300.4	74.7	467.1	440.0	483.8	94.2	156.3	50.4	14.6	4.4	4.2	182.6	48.7	943.9	690.5	963.4	673.9
34.5	1937	274.9	36.6	257.2	238.3	308.4	38.1	63.7	24.9	2.4	223.4	4.7	281.8	52.5	890.0	783.5	1340.0	485.1
36.5	1978	285.9	56.9	271.6	259.1	382.4	37.4	151.0	45.7	21.3	9.05	3.3	0.4.0	1.5	1469.7	1.788	1105.4	438.7
38.5	2018	127.2	26.8	295.8	330.5	345.8	42.7	138.7	44.7	20.5	49.2	4.0	97.2	31.8	1227.5	761.1	938.7	565.2
40.5	2058	267.6	36.9	226.2	149.6	197.9	28.3	45.6	9.91	4.9	16.5	2.1	69.2	21.0	914.9	615.1	882.8	278.6
42.5	2098	195.5	64.8	417.7	486.0	511.6	5.3	139.3	43.5	1.61	47.7	3.0	137.6	55.7	1363.0	931.0	2025.3	318.0
44.5	2139	141.3	73.7	469.0	502.9	449.6	67.2	125.2	37.9	22.3	42.6	2.0	105.7	41.9	1643.0	8.6011	1236.4	384.8

2011; Wakeham et al., 1980). This increase in conifer burning is consistent with the timing of the increase in the Σ PAHs (Figure 2), but retene concentrations spike again at 1300 yr BP while Σ PAHs and Lvg concentrations remain relatively low suggesting that conifer burning may have had a bigger impact during this time period than during the rest of the record. Individual or groups of PAHs, therefore, provide more information than is possible through only examining the Σ PAHs record.

When comparing individual or summed PAHs and Lvg, transport mechanisms and stability must also be considered. Both PAHs and Lvg can be present in gas and particle phases, with different gas/particle partitioning (George et al., 2016; Xie et al., 2014; Yang et al., 2007). Since the atmospheric lifetimes of PAHs range from 1-3 h (gas phase) to 4-5 days (particulate phase; Lammel et al., 2009; Stier et al., 2005), and the lifetime of Lvg ranges between 1 and 26 days (Bai et al., 2013; Lai et al., 2014; Slade and Knopf, 2013), both PAHs and Lvg may be potentially subject to long-range transport but with different dynamics. A previous study of a small North American lake indicated that although PAHs can successfully detect recent fire events within 0.5 km, PAHs failed in detecting known fire events occurring 1-2km away from the lake site (Denis et al., 2012). Similar studies have not yet been performed for Lvg in sediments, but Lvg has been detected in polar ice dating back 10 kyr BP (Zennaro et al., 2014), suggesting that Lvg survives long-range transport.

Within-lake transport could also affect the stability of these proxies prior to deposition. In previous studies carried out in Lake Baikal (Russia) which is deeper than Lake Victoria, sediments from a location with several hundreds of meters of water above the interface, demonstrate similar organic compounds (several PAHs and steroids), are not subject to substantial alteration under these conditions (Reznikov and Adzhiev, 2015; Tani et al., 2009). [AQ: 7] In Lake Victoria, surface wave activity can affect sediment accumulation for water depths shallower than ~50 m (Johnson et al., 2000). The core studied in this paper was collected at a water depth of ~70 m, and so, we assume that such wave action did not affect the samples.

Also, different transport mechanisms between atmospheric (fire proxies) and terrestrial (FeSts) should be considered. Atmospheric transport may act faster than terrestrial transport, introducing an artificial lag between these classes of proxies. However, it is beyond the scope of this study to quantify such a lag. Therefore, we assume that atmospheric and terrestrial inputs are basically synchronous and that possible lags are slight enough to not strongly affect our interpretations.

Although FeSts can also be reported as fluxes, we opted to use several fecal indexes obtained from sterol concentrations (Table 2). Fecal indexes are generally used as pollution indicators in sediments. However, since in situ anaerobic processes can produce coprostanol through hydrogenation reactions (Fattore et al., 1996) and/or e-CoP from CoP epimerization (Bull et al., 2002), it has been recognized that sterol ratios are a more robust approach for assessing human inputs (Tse et al., 2014). We used the fecal index (CoP+e-CoP)/Chol (hereafter referred to as the CoP-Index) as an indicator of human presence (Leeming et al., 1996). The CoP-Index ranged from 0.08 to 0.44 with maximum values in two distinct zones at 8-12 cm (1400-1500 cal. yr BP) and at 29-35 cm (1850-1950 cal. yr BP), indicating a higher CoP input during these periods. Further information on redox conditions in the sedimentary columns can be obtained from 5α-Ch/Chol ratios (Vane et al., 2010; referred to as the 5α-Index). In anaerobic sediments, bacteria can reduce cholesterol to 5α-cholestanol, and therefore, higher 5α-Index ratios describe more reducing environments. However, 5α-Index (Figure 2) fluctuates from 0.5 to 0.9 (mean: 0.7 ± 0.1). Similarly, 5α -Index values in shallow UK lakes (Vane et al., 2010) were interpreted as indicators of a slightly reducing environment.

The FeSt Stg is a marker of the presence of ruminants and, therefore, allows inferring pastoralism (D'Anjou et al., 2012). In the Lake Victoria samples (Table 1), Stg values have large associated errors negating the possibility of determining significant variations through time. Wild fauna (Bloesch, 2008) may influence the Stg signal, overlaying any pastoralism signal. For this reason, we do not further discuss the Stg signal in this paper.

Discussion

Lake Victoria is the largest lake in Africa with a surface area of almost 70,000 km² and a maximum depth of ~80 m. This size suggests that Lake Victoria is suitable for collecting atmospheric and terrestrial signals from a large region. The absence of substantial tributaries restricts external perturbations to sedimentation processes and the evaporation/precipitation balance regulates lake levels. The constant sedimentation rate of ~500 mm 1000 yr⁻¹ observed in V95-1P is consistent with sedimentation values obtained in parallel cores (V95-2P and V95-3P), which have sedimentation rates of 700 and 600 mm 1000 yr⁻¹, respectively (Talbot and Laerdal, 2000). This core section encompasses a reducing environment over its entire length, which minimizes the occurrence of oxidizing reactions that may cause diagenetic transformations that can alter biomarkers. These overall characteristics make Lake Victoria a suitable catchment for collecting both a large spatial scale atmospheric signal (fire) as well as a terrestrial signal (human presence) and for reconstructing human and fire history with a suitable temporal resolution. Considering the vast area of the lake, the terrestrial input may be more representative of the development of large-scale settlements and migrations affecting much of the lake's perimeter rather than reflecting localized settlements. Therefore, we consider both the human history and fire results to reflect regional changes.

This fire history, as determined by Lvg flux, indicates a peak in biomass burning between 1850 and 2050 cal. yr BP (Figure 2). High-resolution charcoal fluxes in neighboring lakes such as Lake Katinda (Colombaroli et al., 2014) demonstrate a similar increase in biomass burning magnitude and frequency at 1850-2150 cal. yr BP coincident with a vegetation transition to open savannah (Figure 3). A regional transition to drier conditions occurs at ~1850 cal. yr BP as evidenced by Lake Edward %Mg and biogenic silica records (Cockerton et al., 2015; Russell and Johnson, 2005) and carbonate maxima content in Lake Katinda. Although the PAH profile in Lake Victoria marks the same relatively intense fire period, this peak is not as pronounced as in the Lvg record (Figure 2). As previously discussed, PAHs and Lvg do not necessarily correlate because of differences in the fire temperatures and transport. However, the relatively greater correlation between Lvg and lowmolecular-weight PAHs suggests the occurrence of low-temperature combustion episodes during this time.

The second fire period (1500–1700 cal. yr BP) is consistent with the later transition from wetter to drier conditions observed in the biogenic silica decrease and %Mg increase at ~1500 cal. yr BP in Lake Edward (Russell and Johnson, 2005). During this period, Lvg better matches the overall Σ PAHs record, and particularly the low-molecular-weight PAHs while also agreeing with the high-resolution charcoal record in Lake Katinda (Colombaroli et al., 2014). These observations support a regional increase in low-temperature fire events and are consistent with the wet-to-dry climate transitions observed in other records.

The retene record provides more specific information on fire sources. As evident from Figure 2, the retene record abruptly increases at $\sim\!1650$ cal. yr BP, indicating the input of resinous softwood vegetation to biomass burning. The retene signal remains elevated, although fluctuating, even when the Lvg and $\Sigma PAHs$ signals decrease. Thus, retene records softwood burning independent of $\Sigma PAHs$ and Lvg. This signal is supported by the progressive

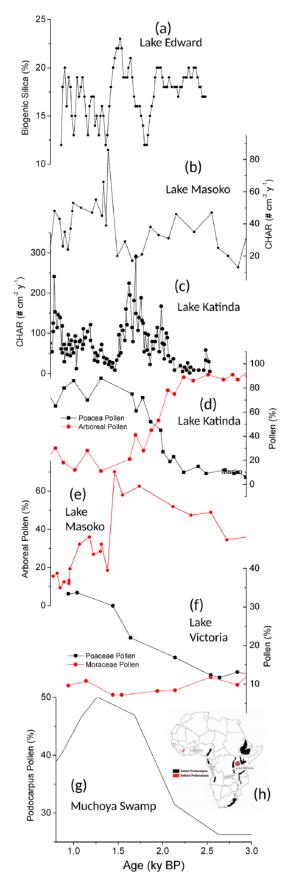


Figure 3. (a) Biogenic silica from Lake Edward (Russell and Johnson, 2005); charcoal record from (b) Lake Masoko (Thevenon et al., 2003) and (c) Lake Katinda (Colombaroli et al., 2014); pollen record from (d) Lake Katinda (Colombaroli et al., 2014), (e) Lake Masoko (Thevenon et al., 2003), and (f) Lake Victoria (Chritz et al., 2015); (g) Podocarpus pollen from Muchoya Swamp (Taylor, 1990); (h) map of Podocarpus distribution in Africa (Adie and Lawes, 2010).

increase in Podocarpus pollen (Kendall, 1969; Kiage and Liu, 2006) and suggests a change in biomass burning dynamics where the anthropogenic influences cannot be excluded, based on the increase in phytoliths observed in Lake Victoria during this same time period (Stager and Johnson, 2000). [AQ: 8] Several conifers such as *Podocarpaceae* (*Podocarpus* and the endemic *Afrocarpus*) and Cupressaceae (i.e. Juniperus procera) are predominant in afromontane forests, though sparse specimens can be found at lower altitudes (Adie and Lawes 2010; Katende et al., 1995; Mumbi et al., 2008; Thompson and Young, 1999). The present-day distribution indicates that modern mountain forests are located several tens of kilometers from the lake. Thus, if the retene derives from natural fire, then the signal is likely because of long-distance transport, where this deduction agrees, with the pollen record (Figure 3). However, an alternate possibility also exists. As detailed later in the paper, conifer wood could have been used for obtaining charcoal and transported from the mountain forests to the lake and used for burning at settlements close to the shore.

The anthropogenic contribution to forest clearance has been claimed by several authors and is mainly supported by historical evidence and pollen records (Finch and Marchant, 2011; Kendall, 1969; Thevenon et al., 2003; Vincens et al., 2005). The CoP-Index record (Figure 2) supports the idea of a connection between the increase in fire activity and human presence. The increase in human activity in the local area as inferred from the CoP-Index lagged the initial increase in fire signal by 1–2 centuries. Although it must be kept in mind that the FeSts signal is potentially very local, we propose two possible interpretations: (a) the natural increase in fire activity created open spaces that consequently encouraged human settlements and (b) even a small group of humans could trigger large fires that contributed to create open spaces and promote the growth of human societies. A dynamic similar to the latter possible interpretation was proposed for the South Island of in New Zealand where the arrival of a limited human community was sufficient to trigger a significant increase in fire activity (McWethy et al., 2014).

In Lake Victoria, both of these dynamics may be possible during 1800-2000 yr BP (I) and 1400-1600 yr BP (II; see Figure 2), where events I and II preferentially support hypotheses a and b, respectively. Arid conditions extended across much of East Africa during the time period encompassing event I, as evidenced by the ratio of Mg to Ca in authigenic calcite (%Mg) in Lake Edward where this ratio serves as a robust indicator of drought (Russell and Johnson, 2005, 2007; Figure 2). Lake Tanganyika also records a prolonged drought during this time period (Cohen et al., 2005; Figure 1). This drought may have acted as the main forcing for increased fire activity, while human inputs may be a secondary effect. In equatorial Africa, the ITCZ passes over the region twice a year, creating rainy seasons in October to December and again in March to May (Nicholson, 1996; Russell and Johnson, 2007). Changes in the biannual migration of the ITCZ influence the amount of precipitation deposited over East Africa. Over multidecadal to millennial timescales, these fluctuations in the ITCZ, coupled with the changes in the Indian Ocean Monsoon to the east and the Congo Air Boundary to the west can together create prolonged droughts over East Africa (Nicholson, 1996; Russell and Johnson, 2007). The spatial extent of the 1800–2000 yr BP present drought suggests that this regional aridity resulted in heightened fire activity, as reflected in the increased Lvg concentrations in Lake Victoria (Figure 2) and increased CHAR in Lake Katinda (Figure 3). The CoP-Index indicates that human communities were present near the lakeshore during this time period, but this index can only detect relatively local settlements (Figure 2). Hypothesis a - where regional opening of land by fire encouraged human settlements – is, therefore, more likely during this time period because of the spatial extent and climatic drivers of drought-associated fires.

However, event II, from 1400 to 1600 yr BP, proposes a different scenario. In event II, drought, fires, and the presence of humans do not all occur at the same time. The %Mg identifies a drought between ~1500 and 1600 yr BP which may be partially responsible for a coincident increase in biomass burning as identified by a peak in Lvg concentrations (Figure 2). The presence of humans in the area lags these fires by approximately a century where human settlements appear to be independent of or at least not linearly dependent on the other variables. The development of technological innovations (as described later in greater detail) suggested by the retene record and the evidence of increasingly elaborate archaeological findings (Ashley et al., 2010) suggest the development of a more complex society during this time period. [AQ: 9] Retene concentrations remarkably differ before and after ~1600 yr BP (Figure 2). A retene signal is completely absent during event I, but conifer burning becomes elevated around ~1600 yr BP and then has three major peaks after this initial change. This conifer fuel burning coincides with the major decrease in arboreal pollen in Lake Masoko beginning at ~1600 yr BP and remains low until ~900 yr BP while grassland pollen dominates the signal during this time period (Figure 3). This combination suggests that the increased conifer burning is not because of any increased availability of conifer vegetation, as forest vegetation declines during this time period. Lvg concentrations demonstrate that general biomass burning is relatively low at ~1500 yr BP, while the human presence and conifer burning are high, suggesting the preferential burning of this fuel and supporting hypothesis b.

The increase in human presence in events I and II differs since the parallel increase in retene during event II may be linked with technological innovations. Historical evidence suggests the development of early metallurgy and ceramic production that required high-temperature fires and burning fuels that provide sufficiently high heat of combustion for processing metals and clays. Available resinous softwood may have provided such a fuel source (Oremusova et al., 2014), which is consistent with an analogous increase in retene that was interpreted as a possible anthropogenic input in Central Europe (Musa Bandowe et al., 2014). For example, in forested areas of Kenya, coniferous wood (Juniperus procera) was preferentially used for obtaining charcoal for fuel supplies for iron production (Thompson and Young, 1999). The spatial and temporal diffusion of metallurgy and clay processing is still uncertain in East Africa. Thompson and Young (1999) proposed 2800 yr BP as a possible initiation of metallurgy, but the expansion was limited to only certain settlements in East Africa. It seems reasonable that the intensification of these practices approximately 1000 years later took advantage of the earlier techniques.

However, in both events I and II, the human presence near Lake Victoria does not appear to be permanent. The increase and subsequent decrease in the CoP-Index suggest that the growth of human settlements is followed by drops in population that can be interpreted as possible migration events that could have also been triggered by other factors, such as resource availability, social dynamics, or even because of the 'woodland tsetse belt', as argued by some archaeologists (Cecchi et al., 2008; Chritz et al., 2015; Ford, 1971). The fluctuation of human settlements enforces the idea that humans responded to variations in climate, and their impact on environment was limited during this time period. This interpretation supports the hypothesis that the creation of natural openings and woodland corridors in the forest was triggered by natural climatic conditions, which, in turn, allowed Bantu-speaking people to spread throughout Central Africa and East Africa (Maley et al., 2012; Neumann et al., 2012).

Conclusion

In this study, we used a multi-proxy approach for reconstructing fire and human history at Lake Victoria using PAHs and Lvg as molecular markers for biomass burning and FeSts as markers of human presence. A total of two peak fire periods occurred at 1400-1700 and 1850-2050 cal. yr BP and were dominated by low-temperature fires. The presence of humans near the lakeshores, reconstructed using the CoP-Index, mimics the fire signal although human settlements lagged behind fire peaks by a few centuries. The retene record differs from other PAHs and Lvg in that it abruptly increases beginning at ~1650 cal. yr BP. This peak may correlate with human activity and, in particular, with anthropogenic fire usage for the development of metallurgy and/or for ceramic production, both of which require the types of fires associated with softwood combustion such as Podocarpus trees. However, the occurrence of severe drought events peaking at ~1500 and 2000 cal. yr BP combined with grassland expansion may have intensified fires and overlapped with human activity. The onset of favorable conditions for human activities could have been created by new climate conditions and may have triggered human settlement growth that, in turn, extended the open spaces. Further insights on this climate and land-use relationship could be obtained by investigating a high-resolution sediment core spanning the pre-Iron Age period and/or also including smaller lakes in the region, especially where archaeological studies have been already conducted. Such studies would allow better determining human impacts on land use, fires, and vegetation distribution in East Africa. Discerning the contributions of anthropogenic versus natural forcing on the climate system before the Industrial Revolution remains a major climate science goal. These human impacts vary both regionally and through time, but such studies improve our understanding of how humans interacted with and changed their environments before industrialization.

Acknowledgements

Cores are archived at LacCore (National Lacustrine Core Facility), Department of Earth Sciences, University of Minnesota-Twin Cities. We thank Professor Thomas Johnson and Professor Melissa Berke for their help in elucidating the dating of the sediment core.

Funding

This work was financially supported by the Early Human Impact ERC Advanced Grant of the European Commission's VII Framework Program, grant number 267696, contribution no. 14.

References

- Adie H and Lawes M (2010) Podocarps in Africa: Temperate zone relicts or rainforest survivors? *Smithsonian Contributions to Botany* 95: 79–100.
- Ashley CZ (2010) Towards a socialised archaeology of ceramics in Great Lakes Africa. *African Archaeological Review* 27: 135–163
- Bai J, Sun X, Zhang C et al. (2013) The OH-initiated atmospheric reaction mechanism and kinetics for levoglucosan emitted in biomass burning. *Chemosphere* 93: 2004–2010.
- Battistel D, Piazza R, Argiriadis E et al. (2015) GC-MS method for determining faecal sterols as biomarkers of human and pastoral animal presence in freshwater sediments. *Analytical and Bioanalytical Chemistry* 407: 8505–8514.
- Bayon G, Dennielou B, Etoubleau J et al. (2012) Intensifying weathering and land use in Iron Age Central Africa. *Science* 335: 1219–1222.
- Berke MA, Johnson TC, Werne JP et al. (2012) Molecular records of climate variability and vegetation response since the Late Pleistocene in the Lake Victoria basin, East Africa. *Quaternary Science Reviews* 55: 59–74.
- Bloesch U (2008) Thicket clumps: A characteristic feature of the Kagera savanna landscape, East Africa. *Journal of Vegetation Science* 19: 31–44.

Bower J (1991) The pastoral Neolithic of East Africa. *Journal of World Prehistory* 5: 49–82.

- Bull ID, Lockheart MJ, Elhmmali MM et al. (2002) The origin of faeces by means of biomarker detection. *Environment Inter*national 27: 647–654.
- Cecchi G, Mattioli RC, Slingenbergh J et al. (2008) Land cover and tsetse fly distribution in sub-Saharan Africa. Medical and Veterinary Entomology 22: 364–373.
- Chritz KL, Marshall FB, Zagal ME et al. (2015) Environments and trypanosomiasis risk for early herders in the later Holocene of the Lake Victoria basin, Kenya. *Proceedings of the National Academy of Sciences* 112: 3674–3679.
- Cockerton HE, Street-Parrott FA, Barker PA et al. (2015) Orbital forcing of glacial/interglacial variations in chemical weathering and silicon cycling within the upper White Nile basin, East Africa: Stable-isotope and biomarker evidence from Lakes Victoria and Edward. *Quaternary Science Reviews* 130: 57–71.
- Cohen AS, Lezzar KE, Cole J et al. (2005) Late-Holocene linkages between decade-century scale climate variability and productivity at Lake Tanganyika, Africa. *Journal of Paleolimnology* 35: 189–209.
- Colombaroli D, Ssemmanda I, Gelorini V et al. (2014) Contrasting long-term records of biomass burning in wet and dry savannas of equatorial East Africa. Global Change Biology 20: 2903–2914.
- Crul RCM (1995) *Limnology and Hydrology of Lake Victoria*. UNESCO/IHP-IV project M-5.1. Paris: UNESCO.
- D'Anjou RM, Bradley RS, Balascio NL et al. (2012) Climate impacts on human settlement and agricultural activities in northern Norway revealed through sediment biogeochemistry. Proceedings of the National Academy of Sciences of the United States of America 100: 20332–20337.
- Denis EH, Toney JL, Tarozo R et al. (2012) Polycyclic aromatic hydrocarbons (PAHs) in lake sediments record historic fire events: Validation using HPLC-fluorescence detection. Organic Geochemistry 45: 7–17.
- Dotterweich M, Rodzik J, Zgłobicki W et al. (2012) High resolution gully erosion and sedimentation processes, and land use changes since the Bronze Age and future trajectories in the Kazimierz Dolny area (Nałeczów Plateau, SE-Poland). *Catena* 95: 50–62.
- Fattore E, Benfenati E, Marelli R et al. (1996) Sterols in sediment samples from Venice Lagoon, Italy. *Chemosphere* 33: 2383–2393.
- Finch J and Marchant R (2011) A palaeoecological investigation into the role of fire and human activity in the development of montane grassland in East Africa. *Vegetation History and Archaeobotany* 20: 109–124.
- Fine PM, Cass GR and Simoneit BRT (2002) Chemical characterization of the fine particle emissions from fireplace combustion of woods grown in the southern United States. *Environmental Science and Technology* 36: 1442–1451.
- Ford J (1971) The Role of the Trypanosomiases in African Ecology: A Study of the Tsetse Fly Problem. Oxford: Clarendon Press.
- Gambaro A, Manodori L, Moret I et al. (2004) Determination of polychlorobiphenyls and polycyclic aromatic hydrocarbons in the atmospheric aerosol of the Venice Lagoon. *Analytical* and *Bioanalytical Chemistry* 378: 1806–1814.
- Gambaro A, Zangrando R, Gabrielli P et al. (2008) Direct determination of levoglucosan at the picogram per milliliter level in Antarctic ice by high-performance liquid chromatography/electrospray ionization triple quadrupole mass spectrometry. Analytical Chemistry 80: 1649–1655.
- George IJ, Black RR, Geron CD et al. (2016) Volatile and semivolatile organic compounds in laboratory peat fire emissions. *Atmospheric Environment* 132: 163–170.

- Halfman JD, Johnson TC and Finney B (1994) New AMS dates, stratigraphic correlations and decadal climate cycles for the past 4 ka at Lake Turkana, Kenya. *Palaeogeography, Palaeo-climatology, Palaeoecology* 111: 83–98.
- Hays MD, Geron CD, Linna KJ et al. (2002) Speciation of gasphase and fine particle emissions from burning of foliar fuels. *Environmental Science and Technology* 36: 2281–2295.
- Hays MD, Gullett B, King C et al. (2011) Characterization of carbonaceous aerosol emitted from outdoor wood boiler. *Energy & Fuels* 25: 5632–5638.
- Huffman TN (1989) Ceramics settlements migrations. *African Archaeological Review* 7: 155–182.
- Johnsen AR, Wick LY and Harms H (2005) Principles of microbial PAH-degradation in soil. Environmental Pollution 133: 71–84.
- Johnson TC and Odada EO (eds) (1996) The Limnology, Climatology, and Paleoclimatology of the East African Lakes. Amsterdam: Gordon and Breach, 664 pp.
- Johnson TC, Kelts K and Odada EO (2000) The Holocene history of Lake Victoria. *Ambio* 29: 1–11.
- Katende AB, Birnie A and Tengnas B (1995) Useful Trees and Shrubs for Uganda: Identification and Management for Agricultural and Pastoral Communities. Kampala; Nairobi: Regional Soil Conservation Units (RSCU), Swedish International Development Cooperation Agency (Sida).
- Kendall RL (1969) An ecological history of the Lake Victoria basin. *Ecological Monographs* 39: 121–176.
- Kiage LM and Liu K-B (2006) Late Quaternary paleoenvironmental changes in East Africa: A review of multiproxy evidence from palynology, lake sediments and associated records. *Progress in Physical Geography* 30: 633–658.
- Kirchgeorg T, Schupbach S, Kehrwald N et al. (2014) Method for the determination of specific molecular markers of biomass burning in lake sediments. *Organic Geochemistry* 71: 1–6.
- Kuo L-J, Louchouarn P and Herbert BE (2011) Influence of combustion conditions on yields of solvent-extractable anhydrosugars and lignin phenols in chars: Implication for characterization of biomass combustion residues. *Chemosphere* 85: 797–805.
- Lai C, Liu Y, Ma J et al. (2014) Degradation kinetics of levoglucosan initiated by hydroxyl radical under different environmental conditions. *Atmospheric Environment* 91: 32–39.
- Lammel G, Sehili AM, Bond TC et al. (2009) Gas/particle partitioning and global distribution of polycyclic aromatic hydrocarbons A modeling approach. *Chemosphere* 76: 98–106.
- Leeming R, Ball A, Ashbolt N et al. (1996) Using fecal sterols from humans and animals to distinguish fecal pollution in receiving water. *Water Research* 30: 2893–2900.
- Li S, Schlebusch C and Jakobsson M (2014) Genetic variation reveals large-scale population expansion and migration during the expansion of Bantu-speaking peoples. *Proceedings of the Royal Society of London B: Biological Sciences* 281: 20141448. Available at: http://dx.doi.org/10.1098/rspb.2014.1448.
- Lu Y, Hautevelle Y and Michels R (2013) Determination of the molecular signature of fossil conifers by experimental palaeochemotaxonomy – Part 1: The Araucariaceae family. *Biogeo*sciences 10: 1943–1962.
- McGlynn G, Mooney S and Taylor D (2013) Palaeoecological evidence for Holocene environmental change from the Virunga volcanoes in the Albertine Rift, central Africa. *Quaternary Science Reviews* 61: 32–46.
- McGrath TE, Chan WG and Hajaligol MR (2003) Low temperature mechanism for the formation of polycyclic aromatic hydrocarbons from pyrolysis of cellulose. *Journal of Analytical and Applied Pyrolysis* 66: 51–70.
- McWethy DB, Wilmshrurst JM, Whitlock C et al. (2014) A highresolution chronology of rapid forest transitions following Polynesian arrival in New Zealand. *PLoS ONE* 9: e111328.

Maley J, Giresse P, Doumenge C et al. (2012) Comment on: 'Intensifying weathering and land use in iron age central Africa'. *Science* 337: 1040. [AQ: 10]

- Marchant R and Taylor D (2000) Pollen representivity of montane forest taxa in south-west Uganda. *New Phytologist* 146: 515–525.
- Medeiros PM and Simoneit BRT (2007) Analysis of sugars in environmental samples by gas chromatography mass-spectrometry. Journal of Chromatography A 1141: 271–278.
- Moskal-del Hoyo M, Lityńska-Zajac M, Korczyńska M et al. (2015) Plants and environment: Results of archaeobotanical research of the Bronze Age settlements in the Carpathian Foothills in Poland. *Journal of Archaeological Science* 53: 426–444
- Mumbi CT, Marchant R, Hooghienstra H et al. (2008) Late quaternary vegetation reconstruction from the Eastern Arc Mountains, Tanzania. *Quaternary Research* 69: 326–341.
- Muri G, Wakeham SG and Faganelli J (2003) Polycyclic aromatic hydrocarbons and black carbon in sediments of a remote alpine lake (Lake Planina, Northwest Slovenia). *Environmental Toxicology and Chemistry* 22: 1009–1016.
- Musa Bandowe BA, Srinivasan P, Seelge M et al. (2014) A 2600year record of past polycyclic aromatic hydrocarbons (PAHs) deposition at Holzmaar (Germany). *Palaeogeography, Pal*aeoclimatology, *Palaeoecology* 401: 111–121.
- Nelson DM, Verschuren D, Urban M et al. (2012) Long-term variability and rainfall control of savanna fire regimes in equatorial East Africa. *Global Change Biology* 18: 3160–3170.
- Neumann K, Eggert MKH, Oslisly R et al. (2012) Comment on: 'Intensifying weathering and land use in iron age central Africa'. Science 337: 1040.
- Nicholson SE (1996) A review of climate dynamics and climate variability in eastern Africa. In: Johnson TC and Odata EO (eds) The Limnology, Climatology, and Paleoclimatology of the East African Lakes. Amsterdam: Gordon and Breach, pp. 25–56.
- Oremusova E, Terenova L and Reh R (2014) Evaluation of grass and net calorific value of the selected wood species. *Advanced Materials Research* 1001: 292–299.
- Pyne SJ (2001) The fires this time, and next. *Science* 294: 1005–1006.
- Ramdahl T (1983) Retene A molecular marker of wood combustion in ambient air. *Nature* 306: 580–582.
- Reznikov SA and Adzhiev RA (2015) Persistent organic pollutants in bottom sediments in the North of lake Baikal-Amur mainline. Russian Meteorology and Hydrology 40: 207–214.
- Ricketts RD and Johnson TC (1996) Climate change in the Turkana basin as deduced from a 4000 year long δ¹⁸O record. Earth and Planetary Science Letters 142: 7–17.
- Robinson AL, Subramanian R, Donahue NM et al. (2006) Source apportionment of molecular markers and organic aerosol, 2: Biomass smoke. *Environmental Science and Technology* 40: 7811–7819.
- Rucina SM, Muiruri VM, Kinjanjui RN et al. (2009) Late Quaternary vegetation and fire dynamics on Mount Kenya. *Palaeogeography, Palaeoclimatology, Palaeoecology* 283: 1–14.
- Russell JM and Johnson TC (2005) A high-resolution geochemical record from Lake Edward, Uganda Congo and the timing and causes of tropical African drought during the late-Holocene. *Quaternary Science Reviews* 24: 1375–1389.
- Russell JM and Johnson TC (2007) 'Little Ice Age' drought in equatorial Africa: Intertropical Convergence Zone migrations and El Nino-Southern Oscillation variations. *Geology* 35(1): 21–24
- Russell T, Silva F and Steele J (2014) Modelling the spread of farming in the Bantu-speaking regions of Africa: An archaeology-based phylogeography. *PLoS ONE* 9: e87854.

- Schauer JJ and Cass GR (2000) Source apportionment of wintertime gas-phase and particle-phase air pollutants using organic compound tracers. *Environmental Science and Technology* 34: 1821–1832.
- Schmidt PR and Childs ST (1995) Ancient African iron production. *American Scientist* 83: 524–533.
- Schüpbach S, Kirchgeorg T, Colombaroli D et al. (2015) Combining charcoal sediment and molecular markers to infer a Holocene fire history in the Maya Lowlands of Peten, Guatemala. *Quaternary Science Reviews* 115: 123–131.
- Simoneit BRT (1999) A review of biomarker compounds as source indicators and tracers for air pollution. *Environmental Science and Pollution Research* 6: 159–169.
- Simoneit BRT (2002) Biomass burning A review of organic tracers for smoke from incomplete combustion. Applied Geochemistry 17: 129–162.
- Simoneit BRT, Rogge WF, Mazurek MA et al. (1993) Lignin pyrolysis products, lignans and resin acids as specific tracers of plant classes in emissions from biomass combustion. *Environmental Science and Technology* 27: 2533–2541.
- Sinclair P, Shaw T and Andah B (1993) Introduction. In: Shaw T, Sinclair P, Andah B, et al. (eds) *The Archaeology of Africa: Food, Metals and Towns*. London: Routledge, pp. 1–31.
- Slade JH and Knopf DA (2013) Heterogeneous OH oxidation of biomass burning organic aerosols surrogate compounds: Assessment of volatilization products and the role of OH concentration on the reactive uptake kinetics. *Physical Chemistry Chemical Physics* 15: 5898–5915.
- Ssemmanda I and Vincens A (2002) Vegetation changes and their climatic implications for the Lake Victoria region during the Late-Holocene. In: Odada EG and Olago DO (eds) *The East African Great Lakes: Limnology, Palaeolimnology and Biodiversity*. Dordrecht: Kluwer Academic Publishers, pp. 509–523.
- Stager JC and Johnson TC (2000) A 12,400 ¹⁴C yr offshore diatom record from east central Lake Victoria, East Africa. *Journal of Paleolimnology* 23: 373–383.
- Stager JC, Cumming BF and Meeker LD (2003) A 10,000-year high-resolution diatom record from Pilkington Bay, Lake Victoria, East Africa. *Quaternary Research* 59: 172–181.
- Stager JC, Cocquyt C, Bonnefille R et al. (2009) A late-Holocene paleoclimatic history of Lake Tanganyika, East Africa. *Quaternary Research* 72: 47–56. [AQ: 11]
- Stager JC, Westwood J, Grzesik D et al. (2005) A 5,500-year environmental history of Lake Nabugabo, Uganda. *Palaeogeography*, *Palaeoclimatology*, *Palaeoecology* 218: 347–354. [AQ: 12]
- Stier P, Feichter Kinne S, Kloster S et al. (2005) The aerosolclimate model ECHAM5-HAM. *Atmospheric Chemistry and Physics* 5: 1125–1156.
- Talbot MR and Laerdal T (2000) The late Pleistocene Holocene palaeolimnology of lake Victoria, East Africa, based upon elemental and isotopic analyses of sedimentary organic matter. *Journal of Paleolimnology* 23: 141–164.
- Tani Y, Nara F, Soma Y et al. (2009) Phytoplankton assemblage in the Plio-Pleistocene record of Lake Baikal as indicated by sedimentary steryl chlorine esters. *Quaternary International* 205: 126–136.
- Taylor DM (1990) Late Quaternary pollen records from two Uganda mires: Evidence for environmental change in the Rukiga highlands of southwest Uganda. *Palaeogeography, Palaeoclimatology, Palaeoecology* 80: 283–300.
- Thevenon F, Williamson D, Vincens A et al. (2003) A late-Holocene charcoal record from Lake Masoko, SW Tanzania: Climatic and anthropologic implications. *The Holocene* 13: 785–792.
- Thompson G and Young R (1999) Chapter 18: Fuels for furnace. In: Van der Veen M (ed.) *The Exploration of Plants Resources in Ancient Africa*. [AQ: 13]

Tse TJ, Codling G, Jones PD et al. (2014) Reconstructing longterm trends in municipal sewage discharge into a small lake in northern Manitoba, Canada. *Chemosphere* 103: 299–305.

- Vane CH, Kim AW, McGowan S et al. (2010) Sedimentary records of sewage pollution using faecal markers in contrasting peri-urban shallow lakes. The Science of the Total Environment 409: 345–356.
- Vansina J (1995) The roots of African cultures. In: Curtin P, Feierman S, Thompson L, et al. (eds) African History: From Earliest Times to Independence. Harlow: Longman, pp. 1–28.
- Verschuren D, Johnson TC, Kling HJ et al. (2002) History and timing of human impact on Lake Victoria, East Africa. Proceedings of the Royal Society of London B: Biological Sciences 269: 289–294.
- Vincens A, Buchet G, Williamson D et al. (2005) A 23,000 yr pollen record from Lake Rukwa: New data on vegetation dynamics and climate in Central Eastern Africa. Review of Palaeobotany and Palynology 137: 147–162.
- Vincens A, Williamson D, Thevenon F et al. (2003) Pollen-based vegetation changes in southern Tanzania during the last 4200 years: Climate change and/or human impact. *Palaeogeogra*phy, *Palaeoclimatology*, *Palaeoecology* 198: 321–334.
- Vincente A, Alves C, Monteiro C et al. (2011) Measurement of trace gases organic compounds in the smoke plume from a wildfire in Penedono (central Portugal). Atmospheric Environment 45: 5172–5182.

- Wakeham SG, Schaffner C and Giger W (1980) Polycyclic aromatic-hydrocarbons in recent lake-sediments, 2. Compounds derived from biogenic precursor during early diagenesis. *Geochimica et Cosmochimica Acta* 44: 415–429.
- Weninger B, Joris O and Danzeglocke U (2011) CalPal-2007. Available at: http://www.calpal-online.de/.[AQ: 14]
- Xie M, Hannigan MP and Barsanti KC (2014) Gas/Particle partitioning of 2-methyltetrols and levoglucosan at an urban site in Denver. *Environmental Science and Technology* 48: 2835–2842.
- Yang C, Lin T and Feng-Hsiang C (2007) Particle size distribution and PAHs concentration of incense smoke in a combustion chamber. *Environmental Pollution* 145: 606–615.
- Yunker MB, Macdonald RW, Vingarzan R et al. (2002) PAHs in the Fraser River basin: A critical appraisal of PAH ratios as indicators of PAH source and composition. *Organic Geochemistry* 33: 489–515.
- Zangrando R, Barbaro E, Zennaro P et al. (2013) Molecular markers of biomass burning in Arctic aerosols. *Environmental Science and Technology* 47: 8565–8574.
- Zennaro P, Kehrwald N, McConnell JR et al. (2014) Fire in ice: Two millennia of boreal forest fire history from the Greenland NEEM ice core. *Climate of the Past* 10: 1905–1924.
- Zou S, Li R, Xie S et al. (2000) Paleofire indicated by polycyclic aromatic hydrocarbons in soil of Jinluojia archaeological site, Hubei, China. *Journal of Earth Science* 21: 247–256.