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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

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ABSTRACT

On Quaternary time scales, the global biogeochemical cycle of silicon is interlocked with the carbon cycle through biotic enhancement of silicate weathering and uptake of dissolved silica by vascular plants and aquatic microalgae (notably diatoms, for which Si is an essential nutrient). Large tropical river systems dominate the export of Si from the continents to the oceans. Here, we investigate variations in Si cycling in the upper White Nile basin over the last 15 ka, using sediment cores from Lakes Victoria and Edward. Coupled measurements of stable O and Si isotopes on diatom separates were used to reconstruct past changes in lake hydrology and Si cycling, while the abundances of lipid biomarkers characteristic of terrestrial/emergent higher plants, submerged/floating aquatic macrophytes and freshwater algae document past ecosystem changes. During the late-glacial to mid-Holocene, 15–5.5 ka BP, orbital forcing greatly enhanced monsoon rainfall, forest cover and chemical weathering. Riverine inputs of dissolved silica from the lake catchments exceeded aquatic demand and may also have had lower Si-isotope values. Since 5.5 ka BP, increasingly dry climates and more open vegetation, reinforced by the spread of agricultural cropland over the last 3–4 ka, have reduced dissolved silica inputs into the lakes. Centennial-to millennial-scale dry episodes are also evident in the isotopic records and merit further investigation.

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1. Introduction

The biogeochemical cycles of essential nutrients such as carbon, nitrogen, phosphorus and silicon are major determinants of global climate and ecophysiology on Quaternary time scales (Berner and Berner, 2012; Kump et al., 2013; Schlesinger and Bernhardt, 2013). The Si cycle is interlocked with the carbon cycle (Conley, 2002; Street-Perrott and Barker, 2008) through several key processes: enhancement of silicate rock weathering by biotic activity (Kelly et al., 1998; Lucas, 2001); pumping of dissolved silica (DSi) by higher plants to form biogenic silica (BSi) particles known as opal phytoliths, which are deposited by litterfall and actively recycled by

soil processes (Carey and Fulweiler, 2012; Derry et al., 2005; Sommer et al., 2006; Song et al., 2012; Struyf and Conley, 2012); and uptake of DSi by siliceous algae, notably diatoms, in rivers, lakes, reservoirs and oceans (Conley, 1997; De La Rocha and Tréguer, 2012; Frings et al., 2014a; Humborg et al., 2000; Smetacek, 1999). The global DSi flux to the oceans is dominated by large tropical rivers (Gaillardet et al., 1999) and by chemical weathering of basalts (Dessert et al., 2003; Dupré et al., 2003). Since DSi is an essential nutrient for marine diatoms, which largely control the export of organic carbon to the deep ocean (Smetacek, 1999; Yool and Tyrrell, 2003), Quaternary variations in DSi output from tropical rivers could potentially have had a significant impact on the drawdown of atmospheric CO₂ by the marine biological pump.

Major uncertainties still surround the impact of Late Quaternary fluctuations in climate and terrestrial ecosystems on the continental weathering flux of DSi and hence on the oceanic carbon

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cycle (De La Rocha and Tréguer, 2012; De La Rocha and Bickle, 2005; Frings et al., 2014a; Froelich et al., 1992; Laruelle et al., 2009; Lupker et al., 2013; von Blanckenburg et al., 2015). Analyses of Si isotopes in diatomaceous lake muds, which are largely immune to recycling by surface processes, have the potential to elucidate the response of the continental Si cycle to past changes in climate, hydrology, vegetation and chemical weathering (Chen et al., 2012; Street-Perrott et al., 2008; Swann et al., 2010). Here, we reconstruct the response of the Si cycle in the headwaters of the White Nile to variations in climate and human activity since the last major desiccation event (~15 ka BP), using sediment cores from Lakes Victoria and Edward. We employ coupled measurements of Si and O isotopes in diatoms ($\delta^{30}\text{Si}_{\text{diatom}}$ and $\delta^{18}\text{O}_{\text{diatom}}$) and lipid biomarkers as tracers for the Si cycle, palaeohydrology and ecosystem changes, respectively. Additional insights are provided by published stratigraphical, geochemical and palaeoecological records from these lakes.

2. Background

2.1. Oxygen isotopes

Studies of O isotopes in diatom silica are relatively well established, for example in temperate latitudes, where it has been shown that the oxygen-isotope values of diatom silica ($\delta^{18}\text{O}_{\text{diatom}}$), like those of carbonates, are controlled primarily by water temperature and/or by the isotope composition of the lake water (Leng and Barker, 2006; Leng and Marshall, 2004). In the tropics, seasonal temperature changes are small (Rozanski et al., 1996, 1993). Instead, the isotopic composition of lake water is influenced by a combination of factors including the amount of precipitation, the $\delta^{18}\text{O}$ value of the water vapour and its trajectory from the source region; although surface processes such as riverine inputs and evaporative enrichment are generally the main controls on $\delta^{18}\text{O}_{\text{diatom}}$ (Barker et al., 2011, 2007, 2001; Hernández et al., 2011; Hernández et al., 2010; Leng and Barker, 2006; Polissar et al., 2006).

2.2. Silicon isotopes

Processes operating at every stage of the Si cycle can be monitored using the stable isotopes of Si as a tracer (Basile-Doelsch, 2006; Cornelis et al., 2011; De La Rocha, 2002; Ding et al., 1996; Douthitt, 1982). Si has three stable isotopes, ^{28}Si , ^{29}Si and ^{30}Si , which can be measured in silicates and natural waters by mass spectrometry. The $^{30}\text{Si}/^{28}\text{Si}$ ratio is reported in standard delta notation relative to an international standard (Leng et al., 2009). Si isotopes are not fractionated by the hydrological cycle. However, uptake of DSi to form biogenic silica (BSi) or secondary minerals, which are isotopically depleted, raises the $\delta^{30}\text{Si}$ value of the residual solution (Basile-Doelsch, 2006). Hence, the instantaneous $\delta^{30}\text{Si}$ value of precipitating silica is dependent on the evolving $\delta^{30}\text{Si}$ of the water and the degree of utilization of the available DSi. Theoretically, in a “closed” system such as the oceanic mixed layer or the epilimnion of a large, stratified lake, Si-isotope fractionation by diatoms will follow a Rayleigh model and the $\delta^{30}\text{Si}$ of the accumulated silica (recorded by $\delta^{30}\text{Si}_{\text{diatom}}$ in sediment cores) will rise towards the starting composition of the source water as the DSi supply is exhausted (Alleman et al., 2005; Basile-Doelsch, 2006; De La Rocha, 2006). By contrast, in an open (steady-state) system with a continuous input of nutrients derived from the same external source, $\delta^{30}\text{Si}_{\text{diatom}}$ values may climb even higher during prolific diatom blooms (Opfergelt et al., 2011; Varela et al., 2004). In the oceans, Si-isotope data have been used as a proxy to reconstruct palaeoproductivity (or more strictly marine silicic-acid use by diatoms relative to initial dissolved silicic-acid concentrations)

(Brzezinski et al., 2002; De La Rocha et al., 1998). However, in continental environments, there have been fewer Si-isotope investigations. Si in rivers comprises both dissolved and particulate matter; measurement of both permits assessment of both weathering- and productivity-related fractionation. Modern studies of Si isotopes in large fluvial systems have documented intense biotic Si cycling in the humid tropics (Frings et al., 2014b; Hughes et al., 2012, 2011, 2013), which may progressively elevate $\delta^{30}\text{Si}$ values downstream in rivers like the Nile that traverse extensive lakes and wetlands, in which DSi is efficiently stripped from the water by both biotic uptake and neof ormation of clays (Cockerton et al., 2013; Ding et al., 2004; Fontorbe et al., 2013).

Modern Si-isotope investigations of lakes have focused on the relationships between climate, hydrogeology, diatom productivity and lake-mixing regimes (Alleman et al., 2005; Opfergelt et al., 2011). These studies, while highlighting the complexity of lake systems (which potentially receive influxes of several weathering-derived components), confirm that Si-isotope ratios are consistent with DSi concentrations (Alleman et al., 2005; De La Rocha et al., 2000; Opfergelt et al., 2011). They also suggest that Si-isotope fractionation by diatoms is independent of species and temperature, offering potential information on past changes in nutrient supply and diatom production. However, few previous studies have analysed Si isotopes in biogenic components of Quaternary lake sediments (Chen et al., 2012; Stephens, 2011; Street-Perrott et al., 2008; Swann et al., 2010). For this reason, we compare our Si-isotope data in this paper with new measurements of lipid biomarkers and published palaeoenvironmental data in order to validate our interpretations.

2.3. Lipid biomarkers

The organic fraction of lacustrine sediments comprises a mixture of autochthonous compounds synthesized by living organisms that lived within the lake, such as algae, bacteria and submerged/floating macrophytes, and allochthonous inputs from the catchment, notably from vascular land plants. *n*-Alkanes, which are least susceptible to degradation, are most commonly used in palaeoenvironmental reconstructions (Meyers, 1997; Meyers and Ishiwatari, 1993). Suites of *n*-alkanes characteristic of certain plant groups can be used to investigate contributions from specific sources (i.e. terrestrial vs. aquatic). Odd-numbered, long-chain homologues (C_{27} – C_{35}) are generally characteristic of terrestrial higher-plant leaf waxes (Eglinton and Hamilton, 1967), whereas short-chain homologues (C_{17} – C_{21}) are characteristic of aquatic algae (Cranwell et al., 1987) but vulnerable to diagenesis. Ficken et al. (2000) demonstrated that mid-chain-length (C_{23} – C_{25}) *n*-alkanes formed the main component of leaf waxes produced by submerged and floating (non-emergent) aquatic macrophytes.

Biomarker ratios of long- and mid-chain *n*-alkanes are used here to assess the relative contributions from vascular-plant sources. The P_{wax} ratio is given by the abundance of long-chain *n*-alkanes over the sum of mid- and long-chain *n*-alkanes ($P_{\text{wax}} = (\text{C}_{27} + \text{C}_{29} + \text{C}_{31}) / (\text{C}_{23} + \text{C}_{25} + \text{C}_{27} + \text{C}_{29} + \text{C}_{31})$), and reflects the proportions of leaf waxes derived from terrestrial plants and emergent aquatic macrophytes such as reeds, relative to those derived from submerged/floating aquatic macrophytes (Zheng et al., 2007). Ficken et al. (2000) proposed the *n*-alkane P_{aq} proxy ($P_{\text{aq}} = (\text{C}_{23} + \text{C}_{25}) / (\text{C}_{23} + \text{C}_{25} + \text{C}_{29} + \text{C}_{31})$) to distinguish the relative contribution of submerged/floating aquatic macrophytes from that of emergent aquatics and terrestrial plants. A P_{aq} value of >0.4 signifies that an important fraction of the *n*-alkanes originated from submerged/floating plants (Ficken et al., 2000).

Odd-numbered, mid- to long-chain *n*-alkenes in lake sediments are widely used as algal indicators (de Mesmay et al., 2007;

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