



Stable-isotope (H, O, and Si) evidence for seasonal variations in hydrology and Si cycling from modern waters in the Nile Basin: implications for interpreting the Quaternary record

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ABSTRACT

Seasonal variations in hydrology and Si cycling in the Nile Basin were investigated using stable-isotope (H, O, and Si) compositions and dissolved Si (DSi) concentrations of surface waters, as a basis for interpreting lacustrine diatom sequences. $\delta^{18}\text{O}$ ranged from -4.7 to $+8.0\text{‰}$ in the wet season and $+0.6$ to $+8.8\text{‰}$ in the dry season (through 2009–2011). Higher $\delta^{18}\text{O}$ values during the dry season reflected increased evapotranspiration and open water evaporation under conditions of lower humidity. Progressive downstream enrichment in the heavy isotope ^{18}O also occurred in response to cumulative evaporative losses from open water bodies and swamps. $\delta^{30}\text{Si}$ values of DSi ranged from $+0.48$ to $+3.45\text{‰}$ during the wet season and $+1.54$ to $+4.66\text{‰}$ during the dry season, increasing the previously reported global upper limit for $\delta^{30}\text{Si}$ values in natural waters by 1‰ . Si-isotope fractionation was most intense during the dry season when demand for DSi by aquatic ecosystems exceeded supply. Progressive downstream enrichment in the heavy isotope ^{30}Si , coupled with decreasing DSi concentrations, represented cumulative Si uptake by diatoms, macrophytes and other Si-accumulating aquatic organisms. The pronounced seasonal variations in DSi concentrations and Si-isotope compositions in the River Nile suggest that its DSi flux to the ocean may have varied significantly on a glacial/interglacial time scale, with important consequences for the marine Si budget and consequently the global C cycle. Anthropogenic impacts were evident in both the water- and Si-isotope datasets, especially during the dry season and along the Main Nile, where water management is most intensive.

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1. Introduction

Variations in the stable-isotope composition of surface waters are valuable tracers of the hydrological cycle (O and H isotopes) and the Si cycle (Si isotopes), both for the modern system and for palaeoenvironmental studies (Kendall and Coplen, 2001; Georg et al., 2006a; Street-Perrott and Barker, 2008; Street-Perrott et al., 2008). When interpreting stable-isotope data from the sediment record it is essential to understand the isotope systematics of the modern system in order to identify the factors that influenced the isotopic composition of the sediment archive (Leng and Marshall, 2004). Diatoms, photosynthetic microalgae found in most aquatic environments, are typically well preserved and abundant in

lacustrine sediments (Round et al., 1990). Diatom silica is formed of biogenic opal ($\text{SiO}_2 \cdot n\text{H}_2\text{O}$) containing oxygen and silicon isotopes that can be used in palaeoenvironmental studies (Leng and Barker, 2006; Leng and Swann, 2010). The isotopic composition of the frustules reflects the aqueous environment in which they formed. The oxygen-isotope composition of diatoms ($\delta^{18}\text{O}_{\text{diatom}}$) is controlled primarily by water temperature and/or by the isotope composition of the water, whereas their silicon isotope composition ($\delta^{30}\text{Si}_{\text{diatom}}$) is related to the availability of this nutrient, which in turn is connected to local factors such as catchment geology and vegetation, chemical weathering, river and groundwater inputs, water-residence time and the occurrence of seasonal diatom blooms (Leng and Marshall, 2004; Leng and Barker, 2006; Leng et al., 2009).

Surface waters can be valuable indicators of the average isotopic composition of rainfall, especially in situations in which limited evaporative enrichment has occurred since precipitation (Fritz,

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1981). The linear relationship between $\delta^2\text{H}$ and $\delta^{18}\text{O}$ in meteoric waters was established by Craig (1961) as $\delta^2\text{H} = 8 \cdot \delta^{18}\text{O} + 10\text{‰}$, and is known as the Global Meteoric Water Line (GMWL), although local deviations occur due to differing climatic and geographical factors (Clark and Fritz, 1997). Hence, in regional or site-specific studies, a Local Meteoric Water Line (MWL) may be preferred. Globally, variations in the $\delta^2\text{H}$ and $\delta^{18}\text{O}$ ratios of precipitation are controlled by climatic (temperature, rainfall amount, humidity, evaporation, wind regime) and geographical parameters (latitude, altitude, distance from moisture source) as described by Dansgaard (1964). At low latitudes, the spatial distribution of isotopes in precipitation is primarily controlled by the source of the water, subsequently modified by continental, altitude and amount effects that are explained by the Rayleigh distillation process. Along the trajectory of an air mass, isotopically heavy water molecules preferentially fall from a diminishing vapour mass, leaving the residual vapour to become progressively depleted (leading to lower $\delta^2\text{H}$ and $\delta^{18}\text{O}$). Subsequent rainfall becomes increasingly lower in isotopic composition (Dansgaard, 1964; Gat, 1996, 2000; Clark and Fritz, 1997). This rainout effect occurs during the transport of an air mass from an oceanic moisture source to the interior of a landmass (continental effect), during orographic uplift (altitude effect) and during heavy convective rainstorms such as those associated with the passage of the Intertropical Convergence Zone (ITCZ) (amount effect).

Deviation from the GMWL/MWL indicates that kinetic effects have modified the original isotopic composition of the precipitation since it formed. Several processes can cause this effect. Surface water or rainfall that has undergone evaporation will typically plot below the MWL on independent Local Evaporation Lines (LELs) (Craig, 1961; Gat et al., 1994). Low humidity leads to slopes very different from the MWL as water-vapour exchange is minimized, and evaporation becomes an increasingly non-equilibrium (kinetic) process, leaving the residual water enriched in the heavier isotopes ^{18}O and ^2H (Craig and Gordon, 1965). Rain condensed from this evaporated vapour will plot above the MWL (i.e. with a greater y-intercept, or deuterium excess). The concept of deuterium excess (or d-excess), defined as $d(\text{‰}) = \delta^2\text{H} - 8 \cdot \delta^{18}\text{O}$, was introduced by Dansgaard (1964). It measures the degree of evaporation at the moisture source or the amount of evaporative enrichment in ^{18}O after the water has condensed. The most important control on d-excess is thought to be humidity (Merlivat and Jouzel, 1979). Information about the fractionating processes in convective systems can be obtained from d-excess values, which may have been modified from their original source composition during their transportation to the precipitation site (Fröhlich et al., 2002). Values lower than 10‰ may indicate secondary evaporation processes, such as the evaporation of falling raindrops in a warm, dry atmosphere (Stewart, 1975; Araguás-Araguás et al., 2000). Recycling of water-vapour in continental basins may be responsible for large d-excess values, as identified in the Amazon Basin (Gat and Matsui, 1991) and the Great Lakes region of North America (Gat et al., 1994).

Growing interest in the role of continental biota in the global Si cycle reflects the close coupling between the global biogeochemical cycles of Si and C (see Street-Perrott and Barker, 2008; Struyf et al., 2009 for reviews). Previously, the main focus was on long-term geological processes of silicate-rock weathering and the draw-down of CO_2 in the marine realm (Berner et al., 1983; Berner, 1994, 1995; Smetacek, 1998; Dugdale and Wilkerson, 2001; Yool and Tyrrell, 2003; Ragueneau et al., 2006; Hilley and Porder, 2008). However, growing evidence shows that certain plants and aquatic organisms have the ability to modify the Si cycle by taking up, recycling and storing significant amounts of Si in their cells before it reaches the ocean (Conley, 1997, 2002; Street-Perrott and Barker, 2008; Struyf and Conley, 2009). Although Si is not classified as an

essential nutrient for plants, amongst other benefits, it can enhance structural rigidity and growth, and reduce abiotic and biotic stresses (Jones and Handreck, 1967; Raven, 1983; Epstein, 1999; Ma et al., 2001). Certain plants that contain $>1\%$ dry weight of silica are known as Si accumulators. These are abundant in a variety of terrestrial and aquatic ecosystems (e.g. grasslands, tropical rainforests, temperate deciduous forests and wetlands). They have the potential to retain large amounts of Si (Bartoli, 1983; Alexandre et al., 1997; Struyf et al., 2005; Blecker et al., 2006; Struyf et al., 2007; Street-Perrott and Barker, 2008; Struyf and Conley, 2009; Schoelynck et al., 2010; Alexandre et al., 2011).

Si is ultimately derived from silicate-rock weathering and is released in dissolved form (dissolved Si; DSi) as orthosilicic acid ($\text{Si}(\text{OH})_4$). Globally, weathering rates are high in tropical headwaters where high relief, high annual-mean temperatures and monsoonal rainfall facilitate rapid physical weathering and erosion, creating freshly weathered surfaces and thereby enhancing the rate of chemical weathering (Brady and Carroll, 1994; White and Blum, 1995; Cochran and Berner, 1996; Gaillardet et al., 1999). In addition to the effects of bedrock composition, topography and climate, it has been shown that higher plants accelerate the rate of silicate weathering by improving the moisture and organic-matter status of soils (Hinsinger et al., 2001). Interactions between plant roots and soil microbes in the rhizosphere also expedite chemical weathering (Kelly et al., 1998; Lucas, 2001).

DSi present in soil solution may be taken up by terrestrial vegetation and precipitated as hydrated amorphous silica (phytoliths), or transported into rivers and lakes, where Si-accumulating aquatic organisms, such as diatoms, sponges and aquatic macrophytes, progressively extract DSi. The residual DSi is transported via rivers, eventually reaching the oceans where it is an essential nutrient for the siliceous phytoplankton that dominate the marine biological pump (Harrison, 2000; Tréguer and Pondaven, 2000). On glacial to interglacial time scales, the Si flux to the oceans can therefore be expected to vary as a result of changes in climate, vegetation type and distribution, hydrology and limnology (Georg et al., 2006a; Street-Perrott and Barker, 2008; Engström et al., 2010).

Although measurements of Si isotopes in natural samples are still relatively scarce, previously reported $\delta^{30}\text{Si}$ values for fresh waters ranged from -0.17 to $+3.4\text{‰}$ (De La Rocha et al., 2000; Ding et al., 2004; Alleman et al., 2005; Georg et al., 2006a, 2007, 2009; Cardinal et al., 2010; Engström et al., 2010; Ding et al., 2011; Opfergelt et al., 2011; Hughes et al., 2012), showing that DSi in rivers and lakes is isotopically heavy compared with primary minerals (felsic magmatic rocks: $\delta^{30}\text{Si} = -0.07 \pm 0.05\text{‰}$; gneisses, granulites and migmatites: $\delta^{30}\text{Si} = -0.10 \pm 0.15\text{‰}$ (André et al., 2006); and mafic magmatic rocks: $\delta^{30}\text{Si} = -0.29 \pm 0.08\text{‰}$ (Savage et al., 2011)). During formation of secondary products (e.g. phytoliths, diatoms and clays), the light isotope of Si (^{28}Si) is preferentially incorporated into the product (De La Rocha et al., 2000), thereby enriching the residual aqueous solution in the heavier isotopes ^{29}Si and ^{30}Si . Hence, Si isotopes offer great potential as tracers of the continental Si cycle (Street-Perrott and Barker, 2008).

Although several studies have used Si isotopes to trace Si cycling in a few river basins across the globe (Ding et al., 2004; Georg et al., 2006a, 2007, 2009; Cardinal et al., 2010; Engström et al., 2010; Ding et al., 2011; Hughes et al., 2012), no-one has so far investigated downstream Si cycling under different climatic regimes in a basin-wide study. Here, we present an overview of the first downstream dataset of coupled $\delta^2\text{H}$, $\delta^{18}\text{O}$ and $\delta^{30}\text{Si}$ measurements on surface waters, in order to determine the impact of seasonal hydrological changes on Si cycling in the Nile Basin. Understanding the processes controlling the isotope systematics of surface waters will facilitate interpretation of the lacustrine diatom record from river-fed lakes along the River Nile.

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