



Compound-specific isotope records of late-quaternary environmental change in southeastern North Carolina

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ARTICLE INFO

Article history:

Received 12 September 2017

Received in revised form

19 December 2017

Accepted 27 December 2017

Keywords:

n-alkane

Hydrogen

Deuterium

Lake sediment

North America

Paleolimnology

Atlantic coastal plain

ABSTRACT

Reconstructions of late Quaternary paleohydrology are rare from the U.S. Atlantic coastal plain (ACP). Here we present compound-specific hydrogen ($\delta^2\text{H}_{\text{alkane}}$) and carbon ($\delta^{13}\text{C}_{\text{alkane}}$) isotope analyses of terrestrially-derived *n*-alkanes from Jones Lake and Singletary Lake in eastern North Carolina spanning the last ~50,000 years. Combined with pollen, charcoal, and bulk geochemical analyses, the $\delta^2\text{H}_{\text{alkane}}$ data indicate arid conditions during the late-Pleistocene, but differing edaphic conditions at the sites perhaps related to differing water table depths. The $\delta^{13}\text{C}_{\text{alkane}}$ data indicate a significant C_4 plant component during the late Pleistocene, but other proxies indicate a sparsely-vegetated landscape. The Pleistocene-Holocene transition is marked by rapid fluctuations in $\delta^2\text{H}_{\text{alkane}}$ values that are similar to the patterns of Bølling Allerød and Younger Dryas isotope data from Greenland indicating sensitivity of the regional climate to short-lived, high-amplitude climatic events. The $\delta^2\text{H}_{\text{alkane}}$ data indicate a mesic early Holocene that supported colonization by *Quercus*-dominated ecosystems. Evidence of middle Holocene aridity in eastern Tennessee and western North Carolina contrasts with evidence of mesic conditions on the ACP, a geographic pattern similar to modern teleconnected precipitation responses to the Pacific Decadal Oscillation. A transition to *Pinus*-dominated ecosystems ~5500 cal yr B.P. is accompanied by a large increase charcoal, but is not coincident with any large changes in $\delta^2\text{H}_{\text{alkane}}$ values, indicating that hydrologic change was likely not responsible for sustained late-Holocene dominance of *Pinus*. The lack of a change in middle Holocene hydrology and the spatiotemporally heterogeneous nature of the *Quercus*-*Pinus* transition on the ACP indicate prehistoric anthropogenic land management practices may represent the most parsimonious explanation for the regionally pervasive ecological change.

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1. Introduction

The southeastern U.S., particularly the Atlantic Coastal Plain (ACP), was the focus of some of the earliest North American paleoclimatology and paleoecology studies between 1940 and 1980 C.E. (Whitehead, 1967; Watts, 1980b). Since that time, paleoenvironmental research spanning the Pleistocene-Holocene transition in the greater southeastern U.S. has slowed considerably, largely as a result of a scarcity of geological archives with sufficient preservation of climate proxies (LaMoreaux, 1999; Liu et al., 2013). The majority of paleoenvironmental studies on the ACP have utilized traditional proxies such as fossil pollen in lacustrine and wetland sediments, which are highly effective proxies of ecological change. Quantitative and semi-quantitative reconstructions of

paleotemperature and paleohydrology based on pollen assemblages have proven highly effective in the western, northern, and northeastern U.S. (Webb et al., 1998; Williams and Shuman, 2008; Bartlein et al., 2011), but the dominance of climatologically ambiguous pollen types such as *Pinus* spp. and *Quercus* spp. that contain species adapted to a wide range of environmental conditions often hinder these approaches on the ACP (Otvos, 2005). Further, the lack of modern analogues and limited chronological control has necessitated the removal of a disproportionate number of southeastern U.S. sites from paleoclimate macroanalyses (Harrison et al., 2003; Bartlein et al., 2011; Liu et al., 2013).

The ambiguous nature of southeastern pollen data has confounded efforts to reconstruct regional paleoclimates. As an example, Goman and Leigh (2004) argued for a relatively wet early to middle Holocene (~9900–6000 cal yr B.P.) for the ACP based on increased abundances of mesic taxa pollen such as *Nyssa*, *Quercus*, and *Cyperaceae* at the Little River site in North Carolina. Goman and Leigh hypothesized that this increased moisture resulted from a

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northeastward shift of the Bermuda High, which enhanced moisture transport into the region and also promoted more frequent tropical storm strikes along the ACP. However, [Otvos \(2005\)](#) argued that the Little River site ‘lacks explicit aridity indicators’ and the change in vegetation may only reflect local edaphic conditions or flood-plain aggradation. Further, Otvos argued that dune formation in the nearby Cape Fear River valley and along the Georgia coast indicates drier conditions during the early Holocene. These contrasting interpretations of Holocene paleohydrology are quite common in the southeastern U.S., with multiple studies indicating a relatively arid early to middle Holocene ([Frey, 1951, 1953; Watts, 1969, 1971, 1975, 1980a; Rich and Spackman, 1979; Delcourt, 1980; Hussey, 1993; Otvos and Price, 2001; Otvos, 2004; Grimm et al., 2006; Tanner et al., 2015](#)) whereas other studies indicate the occurrence of relatively arid conditions during the middle to late Holocene ([Whitehead, 1972, 1981; Seielstad, 1994; Brook, 1996; Goman and Leigh, 2004; Leigh et al., 2004; Leigh, 2008](#)).

Improving our understanding of southeastern U.S. paleoclimatology is becoming increasingly important, in part because the region appears climatologically anomalous based on modern climate change patterns and past climate change events. The modern-day ‘warming hole’ centered over the ACP represents an area that has warmed considerably slower than the rest of the continent for the second-half of the twentieth century ([Meehl et al., 2012; Hartmann et al., 2013](#)). Additionally, limited evidence of high-amplitude, global climate change events in the past, such as the Younger Dryas, has led some researchers to question if such events had any impact in the southeastern U.S. ([Meltzer and Holliday, 2010](#)). [Grimm et al. \(2006\)](#) argued for strong hydrologic responses of Floridian precipitation patterns to global climate events with warmer and wetter climate conditions coinciding with Heinrich events and the Younger Dryas event that are typified by cold, arid conditions in most other parts of the Northern Hemisphere, again highlighting the potentially anomalous response of southeastern U.S. climate to global perturbations.

Other lingering questions regarding the paleoecology of the ACP have arisen as a result of fossil pollen and charcoal analyses. For example, [Taylor et al. \(2011\)](#) noted the temporal and spatial heterogeneity of the transition from early Holocene *Quercus* dominance to late Holocene *Pinus* dominance in pollen records from the ACP, which occurs as early as ~9400 cal yr B.P. in some records, but not until ~4000 cal yr B.P. in others. This heterogeneous response of ecosystems with apparently similar species compositions (*Quercus*-dominated woodlands) and, in some cases, located within 150 km of one another, argues against a pervasive change in climate conditions as the dominant forcing mechanism. [Spencer et al. \(2017\)](#) also noted orders of magnitude increases in macroscopic charcoal influx coincident with the transition from *Quercus* to *Pinus* dominance, but it is unclear if the changing fire regimes were responsible for the change in forest composition or if the changing forest composition was the mechanism responsible for the change in fire regime. Considering the preservation and restoration efforts focused on fire-adapted ecosystems such as the endangered long-leaf pine (*Pinus palustris* Mill.) forests of the ACP, we need to understand the origin and history of these unique ecosystems in order to assure proper management and conservation practices ([Gilliam and Platt, 1999; Van Lear et al., 2005](#)).

In this study, we apply geographically-novel compound-specific isotopic proxies to better understand vegetation and climate change during the late Quaternary on the ACP. Specifically, we analyzed the stable hydrogen ($\delta^2\text{H}_{\text{alkane}}$) and carbon ($\delta^{13}\text{C}_{\text{alkane}}$) isotope compositions of terrestrially-derived *n*-alkanes in sediments collected from Jones Lake and Singletary Lake, North Carolina. The Jones Lake and Singletary Lake records have already been

the subject of repeated multi-proxy analyses that span ~50,000 cal yr B.P. Prior analyses of pollen, charcoal, and bulk sediment geochemistry have yielded important insights into vegetation change and fire activity in the region ([Frey, 1951, 1953; Whitehead, 1964, 1967, 1973; Spencer et al., 2017](#)), but application of the $\delta^2\text{H}_{\text{alkane}}$ and $\delta^{13}\text{C}_{\text{alkane}}$ proxies provides the opportunity to more directly assess paleohydrology and its potential role as a forcing mechanism for ecological change.

The low pH and resulting lack of carbonate preservation in nearly all southeastern wetland environments necessitate the application of an organic proxy for reconstructing paleohydrology. The $\delta^2\text{H}_{\text{alkane}}$ record has proven to be an effective proxy of paleohydrology in a wide variety of environmental settings, particularly when paired with other environmental proxies ([Sachse et al., 2012](#)). [Sachse et al. \(2004\)](#) documented a strong correlation between the $\delta^2\text{H}$ of terrestrially-derived *n*-alkanes (long-chain, odd-numbered) and the $\delta^2\text{H}$ of meteoric water. The terrestrially-derived *n*-alkanes displayed systematically higher $\delta^2\text{H}$ values than algal-derived alkanes, an offset attributed to transpiration in terrestrial plants. Subsequent studies indicated that this offset is likely created during evaporation of soil moisture ([McInerney et al., 2011](#)) and is locked in at the time of leaf formation ([Sachse et al., 2010](#)). In either case, terrestrial alkane $\delta^2\text{H}$ values are particularly sensitive to meteoric water $\delta^2\text{H}$ values and subsequent evapotranspiration processes, particularly during the growing season ([Shuman et al., 2006](#)). Studies since that of [Sachse et al. \(2004\)](#) have confirmed the sensitivity of terrestrial alkane $\delta^2\text{H}$ to meteoric $\delta^2\text{H}$ and subsequent evapotranspiration ([Liu and Huang, 2005; Xia et al., 2008; Douglas et al., 2012](#)), and this biomarker proxy for drought stress in terrestrial vegetation has been applied successfully to sedimentary records of Quaternary climate change ([Tierney et al., 2008; Niedermeyer et al., 2010; Schefuß et al., 2011; Douglas et al., 2014, 2015; Lane et al., 2014](#)).

Terrestrially-derived $\delta^{13}\text{C}_{\text{alkane}}$ records have proven to be useful for paleoecological and paleoclimate reconstructions in a variety of lacustrine settings as they are often sensitive to changing abundances of C_3 - vs. C_4 -photosynthetic plants on the landscape. Plants using the C_3 photosynthetic pathway typically produce odd-numbered, long-chain *n*-alkanes with $\delta^{13}\text{C}_{\text{alkane}}$ values averaging around -31‰ , and C_4 plants producing $\delta^{13}\text{C}_{\text{alkane}}$ values around -22‰ ([Diefendorf and Freimuth, 2017](#)). The compound-specific approach allows for separation of autochthonous vs. allochthonous carbon contributions, which can confound bulk sedimentary $\delta^{13}\text{C}$ records of terrestrial vegetation change ([Brinca et al., 2000](#)). Additionally, the $\delta^{13}\text{C}_{\text{alkane}}$ record can be sensitive to changing hydrology and terrestrial water stress as increased (decreased) moisture availability typically leads to increased (decreased) water use efficiency, decreased (increased) stomatal conductance, and more negative (more positive) $\delta^{13}\text{C}_{\text{alkane}}$ values in plant tissues, thus providing further insight into paleoclimate change ([Diefendorf et al., 2010; Diefendorf and Freimuth, 2017](#)).

Application of the $\delta^2\text{H}_{\text{alkane}}$ and $\delta^{13}\text{C}_{\text{alkane}}$ proxies facilitates new assessments of late Quaternary paleohydrology and paleoecology on the ACP that were not previously possible due to proxy restraints or ambiguity. This approach also permits the assessment of some of the outstanding questions regarding climate change and ecological responses on the ACP that have arisen based on prior analyses of regional sedimentary records. We specifically aim to address paleohydrological responses of the ACP to long-term and short-lived global climate changes, particularly during the Pleistocene-Holocene transition, and the role these changes may have played in paleoecological processes such as the regionally-prominent *Quercus-Pinus* transition and in fire regimes that appear to have changed dramatically during the middle-Holocene.

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