



Hydrogen isotope fractionation in leaf waxes in the Alaskan Arctic tundra

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Abstract

Leaf wax hydrogen isotopes (δD_{wax}) are increasingly utilized in terrestrial paleoclimate research. Applications of this proxy must be grounded by studies of the modern controls on δD_{wax} , including the ecophysiological controls on isotope fractionation at both the plant and landscape scales. Several calibration studies suggest a considerably smaller apparent fractionation between source water and waxes (ϵ_{app}) at high latitudes relative to temperate or tropical locations, with major implications for paleoclimatic interpretations of sedimentary δD_{wax} . Here we investigate apparent fractionation in the Arctic by tracing the isotopic composition of leaf waxes from production in modern plants to deposition in lake sediments using isotopic observations of precipitation, soil and plant waters, living leaf waxes, and waxes in sediment traps in the Brooks Range foothills of northern Alaska. We also analyze a lake surface sediment transect to compare present-day vegetation assemblages to ϵ_{app} at the watershed scale. Source water and ϵ_{app} were determined for live specimens of *Eriophorum vaginatum* (cottongrass) and *Betula nana* (dwarf birch), two dominant tundra plants in the Brooks Range foothills. The δD of these plants' xylem water closely tracks that of surface soil water, and reflects a summer-biased precipitation source. Leaf water is enriched by $23 \pm 15\%$ relative to xylem water for *E. vaginatum* and by $41 \pm 19\%$ for *B. nana*. Evapotranspiration modeling indicates that this leaf water enrichment is consistent with the evaporative enrichment expected under the climate conditions of northern Alaska, and that 24-h photosynthesis does not cause excessive leaf water isotope enrichment. The ϵ_{app} determined for our study species average $-89 \pm 14\%$ and $-106 \pm 16\%$ for *B. nana* *n*-alkanes and *n*-acids, respectively, and $-182 \pm 10\%$ and $-154 \pm 26\%$ for *E. vaginatum* *n*-alkanes and *n*-acids, which are similar to the ϵ_{app} of related species in temperate and tropical regions, indicating that apparent fractionation is similar in Arctic relative to other regions, and there is no reduced fractionation in the Arctic. Sediment trap data suggest that waxes are primarily transported into lakes from local (watershed-scale) sources by overland flow during the spring freshet, and so δD_{wax} within lakes depends on watershed-scale differences in water isotope compositions and in plant ecophysiology. As such, the large difference between our study species suggests that the relative abundance of graminoids and shrubs is potentially an important control on δD_{wax} in lake sediments. These inferences are supported by δD_{wax} data from surface sediments of 24 lakes where ϵ_{app} , relative to δD_{xylem} , averages $-128 \pm 13\%$ and

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–130 ± 8‰ for *n*-acids and *n*-alkanes, respectively, and co-varies with vegetation type across watersheds. These new determinations of plant source water seasonality and ϵ_{app} for the Arctic will improve the $\delta\text{D}_{\text{wax}}$ paleoclimate proxy at high latitudes. © 2017 Elsevier Ltd. All rights reserved.

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1. INTRODUCTION

Hydrogen and oxygen isotope ratios in meteoric water (δD and $\delta^{18}\text{O}$) are well-established tracers of environmental processes (Dansgaard, 1964; Ehleringer and Dawson, 1992; Vachon et al., 2010; Welker, 2012). When preserved in the geologic record, these isotopes serve as robust tools for paleoclimate reconstructions (Feakins et al., 2012; Jasechko et al., 2015; Klein et al., 2016; Konecky et al., 2016). Hydrogen isotope ratios of plant leaf waxes are an increasingly utilized proxy because they are abundant in many sediments (Huang et al., 2004; Sachse et al., 2004), are stable over long time periods (Yang and Huang, 2003), and their isotopic composition ($\delta\text{D}_{\text{wax}}$) primarily reflects the δD of precipitation ($\delta\text{D}_{\text{precipitation}}$) (Sternberg, 1988; Sauer et al., 2001; Huang et al., 2004; Sachse et al., 2004; Sachse et al., 2010). The $\delta\text{D}_{\text{wax}}$ is depleted by a fractionation factor (ϵ_{app}) relative to $\delta\text{D}_{\text{precipitation}}$ due to several isotope-discriminating processes that occur between precipitation and leaf wax synthesis and deposition (Sessions et al., 1999; Chikaraishi et al., 2004; Sachse et al., 2012; Kahmen et al., 2013b). Accurate estimates of ϵ_{app} are therefore fundamentally important to guide climatic interpretations of ancient $\delta\text{D}_{\text{wax}}$ (Polissar and Freeman, 2010; Yang et al., 2011; Garcin et al., 2012; Feakins, 2013; Niedermeyer et al., 2016), and ideally, to quantitatively determine $\delta\text{D}_{\text{precipitation}}$ and climate variations in geological time.

Numerous analyses of $\delta\text{D}_{\text{wax}}$ from lake sediments and living plants in temperate and tropical regions have begun to converge on average ϵ_{app} values of –100 to –130‰ (Sauer et al., 2001; Sachse et al., 2004; Smith and Freeman, 2006; Hou et al., 2008; Garcin et al., 2012; Kahmen et al., 2013a; Liu et al., 2016), with *n*-alkanes displaying slightly greater isotope discrimination than *n*-alkanoic acids (Chikaraishi and Naraoka, 2007). Recent estimates of ϵ_{app} at high-latitude sites, however, are dramatically different. Shanahan et al. (2013) estimated ϵ_{app} of –61‰ for C_{26} and C_{28} alkanolic acids using lake surface sediment samples from Baffin Island in the High Arctic (latitude: 63–73 °N) compared against mean annual precipitation isotopes compositions for source water estimated from the Online Isotopes in Precipitation Calculator (OIPC) geospatial model (Bowen and Revenaugh, 2003). Porter et al. (2016) produced similar ϵ_{app} values for both long-chain *n*-acids and long-chain *n*-alkanes by comparing fossil waxes to adjacent fossil water (interpreted as mean annual precipitation formed simultaneously with the waxes) in loess sections in the Canadian sub-Arctic (latitude: 63.5 °N). Based on growth chamber experiments, these low ϵ_{app} values in high-latitude, continuous light environments have been suggested to result from plant stomata remaining open throughout the

24-hour sunlit period, thus driving high daily rates of evapotranspiration and high leaf water isotope enrichment (Yang et al., 2009).

In contrast, Wilkie et al. (2012) studied lake sediment waxes (*n*-acids) in northern Siberia (latitude: 67°N) and reported ϵ_{app} of –101‰ with respect to estimates of mean annual precipitation isotope composition, and ϵ_{app} of –110‰ with respect to the measured isotopic composition of spring streamflow. Sachse et al. (2004) report ϵ_{app} of –100 to –135‰ for long chain *n*-alkanes from Arctic Europe using similar methods. These contrasting results raise the following questions: (1) is ϵ_{app} latitude-dependent? (2) is ϵ_{app} highly variable across high latitude biomes? and (3) are observations of small ϵ_{app} an artifact of relying on estimated, rather than measured, source water isotope compositions?

The apparent fractionation of Arctic $\delta\text{D}_{\text{wax}}$ is extremely important to understanding past and current polar climate change. $\delta\text{D}_{\text{wax}}$ records in polar regions have been interpreted as both summer and mean annual temperature change on time-scales from the Holocene to the Paleocene (Pagani et al., 2006; Feakins et al., 2012; Thomas et al., 2012; Pautler et al., 2014; Porter et al., 2016), with implications for the Earth's equilibrium climate sensitivity and future response to rising greenhouse gases. For example, calculations of Paleocene/Eocene $\delta\text{D}_{\text{precipitation}}$ from ancient wax δD and an ϵ_{app} of –100‰ to –130‰ reveal extreme warmth and moisture convergence in the Arctic during the Paleocene/Eocene thermal maximum (PETM) (Pagani et al., 2006). If a smaller ϵ_{app} of –60‰ is used, however, the estimated $\delta\text{D}_{\text{precipitation}}$ during this time period was similar to modern $\delta\text{D}_{\text{precipitation}}$, and not strongly enriched, casting doubt on our understanding of Arctic climate during the PETM. Paleoclimate inferences for Antarctica during the mid-Miocene (Feakins et al., 2012) are likewise sensitive to whether an ϵ_{app} value of –100‰ or –60‰ is used to calculate $\delta\text{D}_{\text{precipitation}}$. Similarly, two temperature anomaly estimates for the last glacial maximum in western Canada (Pautler et al., 2014; Porter et al., 2016), which rely on the same $\delta\text{D}_{\text{wax}}$ data but different values of ϵ_{app} , differ by 14 °C. Clearly, large deviations of ϵ_{app} , caused either by inaccurate assessment of plant source water δD values, by enhanced leaf water isotope enrichment during 24-h transpiration, or by large changes in vegetation assemblages, would complicate interpretations of polar $\delta\text{D}_{\text{wax}}$.

With the exception of the study by Wilkie et al. (2012), investigations of ϵ_{app} in the Arctic have thus far relied on estimated $\delta\text{D}_{\text{precipitation}}$ values from the OIPC model (Bowen and Revenaugh, 2003; Yang et al., 2011; Shanahan et al., 2013) or measurements of relict (frozen) water in permafrost (Porter et al., 2016). Both of these methods could be insufficient for determining ϵ_{app} consider-

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