

Supplementary Material

S1. Analysis of stable carbon isotopes of HMW n-alkanoic acids

The stable carbon isotopic composition ($\delta^{13}\text{C}$) of HMW *n*-alkanoic acids was performed using gas chromatography coupled to isotope ratio mass spectrometry (GC-IRMS). The samples were measured on a Thermo Trace gas chromatograph coupled via a combustion reactor to a MAT252 mass spectrometer. Isotope values were measured against a calibrated reference gases (CO_2) and are reported in per mil relative to the VPDB standards. In order to check the performance of the system a standard mixture consisting of 16 *n*-alkanes was run every sixth sample. The accuracy and precision (mean deviation from offline values and the respective relative standard deviation, RSD) were 0.18 ‰ and 0.25 ‰, respectively. The machine was operated only when the average absolute deviation from offline values was <0.3 ‰. Replicate measurements of the samples yielded a standard deviation of 0.0-0.3 ‰. We report the $\delta^{13}\text{C}$ of the *n*- $\text{C}_{26:0}$ and *n*- $\text{C}_{28:0}$ alkanolic acids as they turned out to be the most abundant homologues in our samples. Since the fatty acids were methylated to facilitate GC-IRMS analysis (see section 3 of the main manuscript), the $\delta^{13}\text{C}$ -signature of the respective fatty acid methyl esters were corrected for the added methyl group using isotope mass balance ($\delta^{13}\text{C}_{\text{wax } n\text{-alkanoic acid}}$).

S2. Calculating δD of precipitation (δD_p)

Correcting for isotopic fractionation that takes place during biosynthesis (apparent fractionation, referred to as ϵ_{app}) makes possible to reconstruct the δD_p from $\delta\text{D}_{\text{wax}}$ (Sachse et al., 2012, Collins et al., 2013; Tierney et al., 2017). This, in turn depends on hydrological processes including rainfall amount, evapotranspiration or the moisture source. Moreover, thermodynamic fractionation associated with temperature changes can affect the isotopic composition of the source water (Sachse et al., 2012). In tropical regions temperature changes are negligible and the amount effect dominates δD of precipitation (δD_p) (Sachse et al., 2012). Therefore, the $\delta\text{D}_{\text{wax } n\text{-alkanoic acids}}$ (this study) and $\delta\text{D}_{\text{wax } n\text{-alkanes}}$ (Castañeda et al., 2016) from core GeoB7702-3 can be used to reconstruct δD_p once ϵ_{app} is known. ϵ_{app} varies across plant-species, particularly associated with different photosynthetic pathways. C4 grasses tend to have more negative ϵ_{app} than C3 trees and shrubs (Sachse et al., 2012). In order to correct for past changes in C4 versus C3 plants ($\delta\text{D}_{p\text{-vc}}$, where “vc” refers to vegetation corrected) we inferred the relative abundance of C4 plants (%C4) from the $\delta^{13}\text{C}_{\text{wax } n\text{-alkanoic acids}}$ (this study) and $\delta^{13}\text{C}_{\text{wax } n\text{-alkanes}}$ (Castañeda et al., 2016) according to Collins et al. (2013). We used the endmember values for C4 and C3 plants given in the “all Africa” compilation by Garcin et al. (2014) (C4: -19.8 ‰ and C3: -33.4 ‰). Next, ϵ_{app} was calculated using -113 ‰ and -126 ‰ as ϵ_{app} endmembers for C3 and C4 plants, respectively (Sachse et al., 2012; Tierney et al., 2017). We note that these values correspond to the *n*- C_{29} alkane homologue. As there are no comparable endmember data for the *n*- $\text{C}_{26:0}$ and *n*- $\text{C}_{28:0}$ alkanolic acids, we assume that alkanes and alkanolic acids have similar ϵ_{app} values. We forwent to correct for temperature changes since the effect of temperature is considered negligible in low latitudes (Sachse et al., 2012). $\delta\text{D}_{p\text{-vc}}$ were corrected for glacial-to-Holocene changes in global ice-volume ($\delta\text{D}_{p\text{-vc-ic}}$, where “ic” means ice volume corrected) applying stacked data of oxygen isotopic compositions ($\delta^{18}\text{O}$) of benthic foraminifera (L04-stack; Lisiecki and Raymo, 2005). The downcore results of $\delta\text{D}_{p\text{-vc-ic}}$ are illustrated in Figure S1.

S3. Supplementary Figures

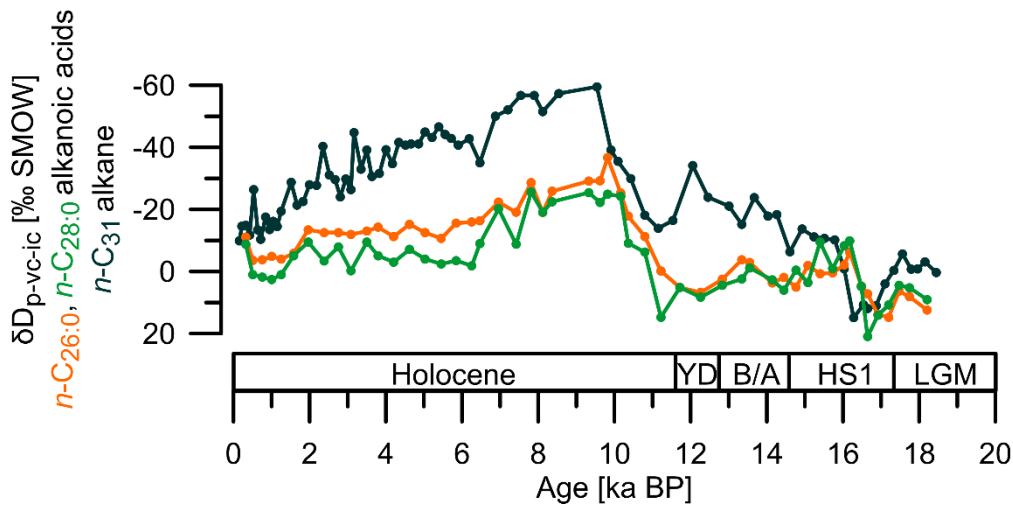


Figure S1: $\delta D_{p-vc-ic}$ of HMW n -alkanoic acids and HMW n -alkanes calculated from $\delta D_{wax\ n-alkanoic\ acids}$ (this study) and $\delta D_{wax\ n-alkanes}$ (Castañeda et al., 2016) in core GeoB7702-3.

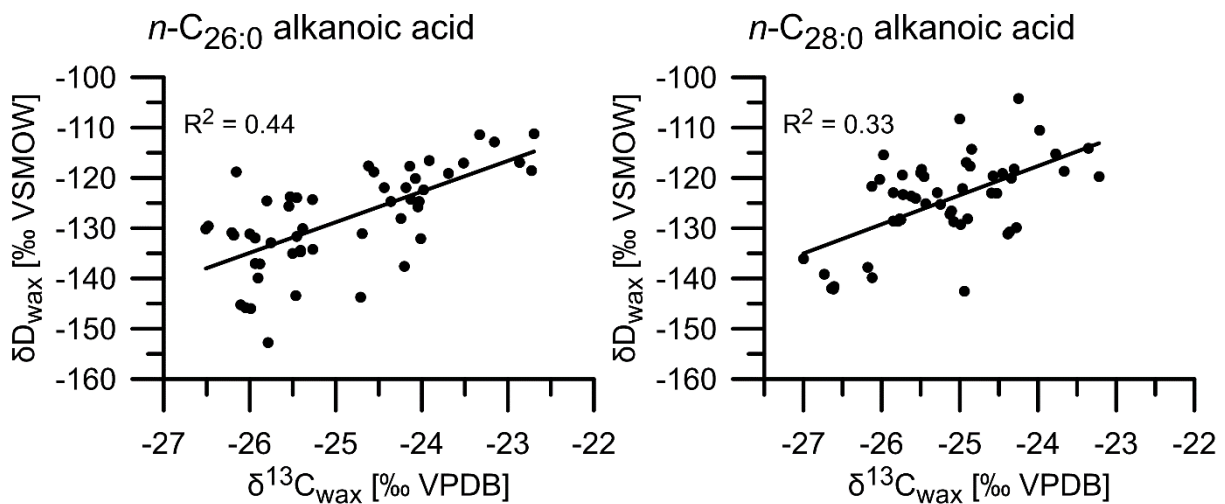


Figure S2) Correlation between $\delta D_{wax\ n-alkanoic\ acids}$ and $\delta^{13}C_{wax\ n-alkanoic\ acids}$ for the $n-C_{26:0}$ and $n-C_{28:0}$ homologues. The respective correlation coefficients (R^2) are indicated and reveal poor linear correlations. Thus, a significant impact of changes in the abundance of C3 versus C4 plants on $\delta D_{wax\ n-alkanoic\ acids}$ throughout the past 18 ka is unlikely.

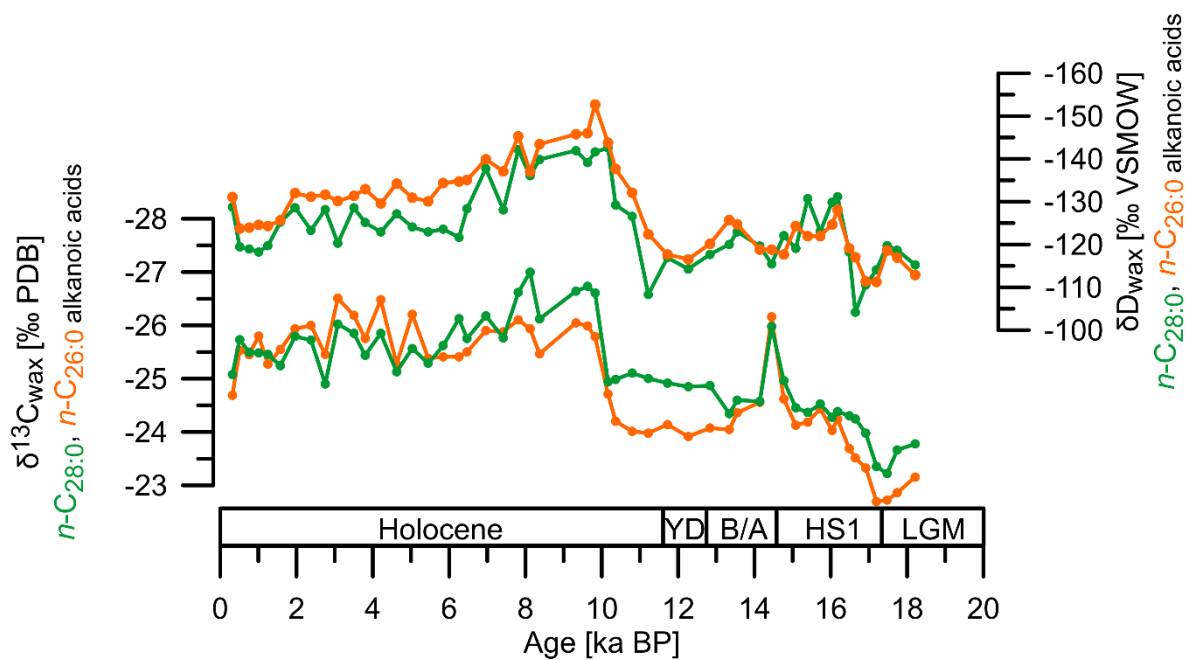


Figure S3: $\delta^{13}\text{C}_{\text{wax}}$ *n*-alkanoic acids along with $\delta\text{D}_{\text{wax}}$ *n*-alkanoic acids in core GeoB7702-3.

References

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