

Paleohydrologic reconstruction based on *n*-alkane distributions in ombrotrophic peat

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Abstract

Proxies such as plant macrofossils, humification indices and testate amoebae have been developed to reconstruct past hydrological variability in ombrotrophic peatlands.

Reconstructions based on these approaches have resulted in records of decadal- to centennial-scale moisture variability in regions of North America and elsewhere. Although the various peatland moisture proxies generally show significant temporal covariance, multi-proxy approaches can refine knowledge of the multivariate nature of climate change and increase confidence in interpretations. Here, we demonstrate that ratios of the abundances of *n*-alkanes provide a new and efficient way of reconstructing past peatland surface moisture change. We found strong correlations among *n*-alkane indices, humification indices and testate amoebae-inferred water table depths from a single core. Biogeochemical proxies can be used in paleohydrological reconstructions for ombrotrophic bogs to provide a new and complementary source of data from these under-utilized paleoclimate archives.

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1. Introduction

The sediments from ombrotrophic peatlands are a well-established source of paleoenvironmental information (Blackford, 2001). Such peatlands are hydrologically independent of groundwater, receiving all moisture and nutrients from precipitation (Charman, 2002). The close relationship between substrate moisture levels and atmospheric conditions, coupled with the high preservation of organic

material that results from low pH and anoxia, make the sediment archives of these peatlands particularly useful for paleoclimate studies.

Previously developed proxies for past substrate-moisture conditions for ombrotrophic peatlands include testate amoebae (Charman, 2001), humification indices (Blackford and Chambers, 1993) and plant macrofossils (Barber et al., 1994). These methods have been extensively validated through comparative studies within the same site (Charman, 1999; Booth and Jackson, 2003), between sites (Booth et al., 2006) and through direct comparison with the instrumental climate record (Schoning

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et al., 2005). Although different proxies generally show coherent patterns of temporal variability, a multi-proxy approach can corroborate, refine, and more realistically capture the potential interactions among different climate variables (Booth and Jackson, 2003; Blundell and Barber, 2005; Langdon and Barber, 2005).

The strong relationship between plant species distribution and surface moisture conditions on ombrotrophic peatlands (Charman, 2002) suggests that the relative abundance of organic biomarkers derived from peat-forming plants can also be used to reconstruct past surface moisture conditions. The distributions of organic biomarkers from plant tissues in peat have been reported (Nott et al., 2000; Pancost et al., 2002; Zhou et al., 2005), but their application as paleoclimate proxies has been underexplored. The method has promise in situations where plant macrofossils are poorly preserved (Nott et al., 2000; Pancost et al., 2002) and as a complementary approach in multi-proxy studies. Different types of plants produce leaf wax *n*-alkanes with differing carbon chain lengths. Leaf wax of vascular plants (grasses, sedges, trees and shrubs) is dominated by long chain (C_{29} – C_{31}) *n*-alkanes, whereas Sphagnum leaf wax is characterized by medium chain length (C_{23} – C_{25}) *n*-alkanes (Nott et al., 2000; Pancost et al., 2002). Ratios of these *n*-alkanes have been compared with

macrofossils of the plants, but with limited success (Pancost et al., 2002). However, ratios of *n*-alkanes in peat have not been directly compared with amoebae-inferred water table depths and humification indices.

In this study we assess the potential of using biomarker *n*-alkanes as surface moisture proxies by directly comparing changes in the ratios of biomarker *n*-alkanes with already-observed changes in testate amoebae-inferred water table depths and humification indices in a peat core from Minden Bog in southeastern Michigan, USA (Fig. 1). We hypothesized that the biomarker ratios from these sediments would covary with these other proxies. The present day vegetation on the bog is dominated by ericaceous shrubs and Sphagnum. We expected temporal changes in biomarker ratios to be consistent with increased relative abundance of Sphagnum mosses during wetter time intervals and increased relative abundance of other plants during dry periods.

2. Samples and methods

We used two complementary approaches. We examined the distributions of *n*-alkanes in the extractable lipids of a variety of bog plants to identify systematic variation in the abundance of *n*-alkanes of different carbon number. We then analyzed

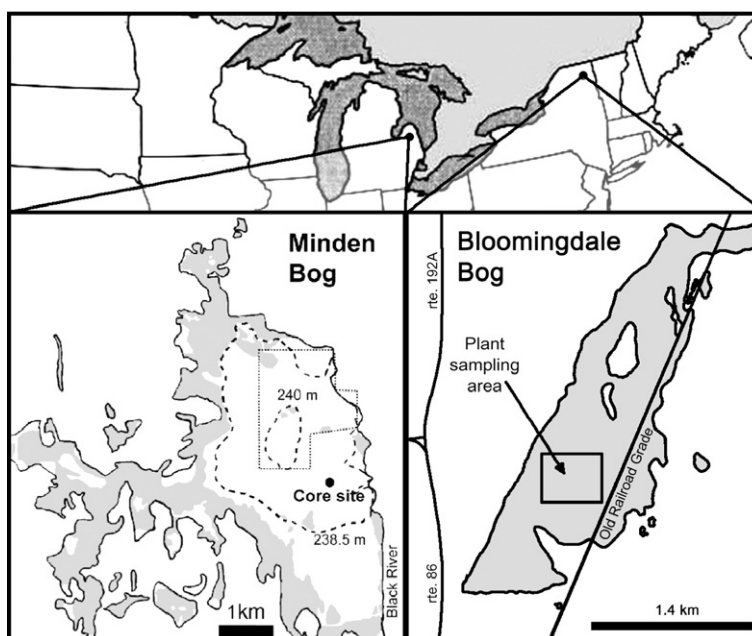


Fig. 1. Top: map of the northern USA showing locations of Minden and Bloomingdale Bogs. Bottom: site maps of Minden (after Booth and Jackson, 2003) and Bloomingdale Bogs.

the abundance of *n*-alkanes in 107 peat samples, representing ~3000 yr, from a 184 cm sediment core from Minden Bog, Michigan, USA (43°36'43"N, 82°50'10"W). Each sample represented 1 cm of peat depth and each had a corresponding testate-amoeba assemblage, testate-amoeba-inferred water table depth and humification measurement (Booth and Jackson, 2003). The age model used was published by Booth and Jackson (2003). We compared the *n*-alkane results with paleohydrological variation inferred from other proxies from the same core (Booth and Jackson, 2003).

Seven modern bog-forming plant species were collected from Bloomingdale Bog in Bloomingdale, NY, USA (44°22'58"N, 74°8'18"W) (Fig. 1) to identify potential taxa-specific biomarkers. Samples of two mosses (*Sphagnum* sp., *Polytrichum formosum*), four shrubs (*Rhododendron groenlandicum*, *Kalmia polifolia*, *Kalmia angustifolia*, *Chamaedaphne calyculata*) and one sedge (*Carex*) were extracted by soaking for 24 h in a 1:1 solution of CH₂Cl₂ and MeOH. Peat samples were extracted for 30 min using ultrasonic agitation and 24 h of soaking with a 9:1 solution of CH₂Cl₂ and MeOH. The polarity of the extraction solvent for peat samples was lowered to avoid extracting large amounts of (unwanted) polar compounds. Total lipid extracts from both plants and peats were separated into neutral and acid fractions using a solid phase extraction column (Amino-propyl Bond Elute; Huang et al., 1999a). Straight chain alkanes were separated from the rest of the neutral fraction by eluting them from a silica gel flash column using hexane. No aliquot was taken. The *n*-alkane abundances were calculated as % of total *n*-alkanes using the response data from gas chromatography-flame ionization detection (GC-FID). Compounds were identified using gas chromatography-mass spectrometry (GC-MS; Huang et al., 1999a).

We explored various ratios of *n*-alkanes extracted from both modern plants and peat and used principal components analysis (PCA) to identify patterns of variability within the distribution data for both modern and downcore samples. Non-linear methods such as non-metric multidimensional scaling and correspondence analysis were also explored, and the dominant gradient of variability was similar among all methods. PCA was performed on *n*-alkane data from peat and modern plant samples after first transforming the biomarker data into % values based on the total amount of C₂₁, C₂₃, C₂₅, C₂₇, C₂₉, C₃₁, and C₃₃ *n*-alkanes. Data used for this

analysis were compiled for the six plant species collected and analyzed for this study with additional data from Nott et al. (2000), Baas et al. (2000) and Pancost et al. (2002). In an effort to include as many different species as possible, we included specimens collected from Europe as well as North America. All the species in the analysis are common bog plants, but we assumed that a particular species produces a similar distribution of *n*-alkanes regardless of location. MATLAB 7.0.1.24704 software was used for the analyses and correlation coefficients were used for the cross products matrix.

3. Results and discussion

3.1. Model *n*-alkane distributions in peat-forming plants

The average distribution of *n*-alkanes in *Sphagnum* species and other bog-forming species is shown in Fig. 2. On average, the most abundant *n*-alkane in *Sphagnum* is C₂₃, whereas the most abundant in other plant species is C₃₁. A similar pattern has been observed (Nott et al., 2000; Pancost et al.,

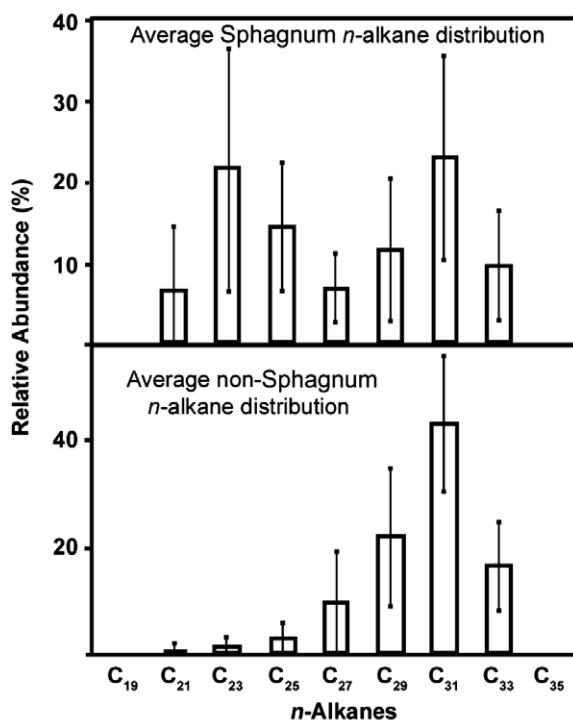


Fig. 2. *n*-Alkane distribution averages for *Sphagnum* and non-*Sphagnum* plants. The distribution includes data from this study and published data (Nott et al., 2000; Pancost et al., 2002).

2002). PCA analysis of the *n*-alkane distribution data is shown in Fig. 3. The patterns indicate substantial differences among species, particularly between Sphagnum and other plants. The dominant gradient (axis 1) revealed by PCA correlates best with variation in the amount of C₂₃ and C₃₁ *n*-alkanes ($r = 0.61$ and $r = -0.57$, respectively). This also corresponds with the differences in *n*-alkane composition of Sphagnum and other plant species,

with medium chain length *n*-alkanes more characteristic of Sphagnum and long chain length *n*-alkanes characteristic of other plants (Fig. 3). Axis 2 corresponds to variation in the amount of C₂₉, and C₃₁ ($r = 0.78$ and $r = -0.42$, respectively). There are, however, variations in *n*-alkane distribution among Sphagnum species, with those found typically in hollows habitats generally producing greater amounts of C₂₃ relative to C₃₁, and those

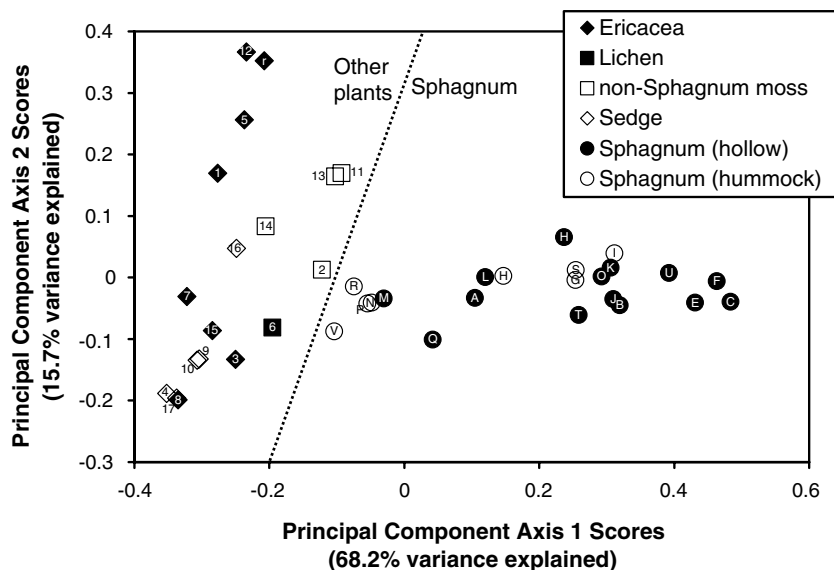


Fig. 3. Results of PCA analysis of modern plant *n*-alkane distributions. Non-Sphagnum plants and long chain *n*-alkanes group together and are circumscribed with a thin line. Sphagnum species and medium chain length *n*-alkanes group together and are also circumscribed with a thin line. *n*-Alkanes are labeled with carbon numbers. Plant species are labeled as follows (* this study; ** Nott et al., 2000; † Baas et al., 2000; ‡ Pancost et al., 2002):

1. *Andromeda polifolia***
2. *Aulacomnium palustre***
3. *Calluna vulgaris***
4. *Carex* sp.*
5. *Chamydaphne calyculata***
6. *Cladonia* sp.**
7. *Empetrum nigrum***
8. *Erica tetralix***
9. *Eriophorum angustifolium***
10. *Eriophorum vaginatum***
11. *Hypnum cupressiforme***
12. *Kalmia polifolia***
13. *Polytrichum formosum***
14. *Polytrichum* sp.**
15. *Rhododendron groenlandicum***
16. *Rhynchospora alba***
17. *Trichophorum cespitosum***
18. *Vaccinium oxycoccus***

- A. *Sphagnum compactum*†
- B. *Sphagnum cuspidatum*†
- C. *Sphagnum cuspidatum*†
- D. *Sphagnum fimbriatum*†
- E. *Sphagnum imbricatum*†
- F. *Sphagnum imbricatum*†
- G. *Sphagnum magellanicum*†
- H. *Sphagnum molle*†
- I. *Sphagnum palustre*†
- J. *Sphagnum papillosum*†
- K. *Sphagnum pulchrum*†
- L. *Sphagnum recurvum*†
- M. *Sphagnum rubellum*†
- N. *Sphagnum rubellum*†
- O. *Sphagnum tenellum*†
- P. *Sphagnum capillifolium***
- Q. *Sphagnum cuspidatum***
- R. *Sphagnum magellanicum***
- S. *Sphagnum palustre***
- T. *Sphagnum papillosum***
- U. *Sphagnum recurvum***
- V. *Sphagnum* sp.*

characteristic of hummock top habitats producing greater amounts of C_{31} . For example, *Sphagnum magellanicum*, and *Sphagnum capillifolium*, are common on drier hummock tops (Andrus, 1980; Crum, 1988; Hájková and Hájek, 2002). Both produce a greater abundance of C_{31} (33.5% and 33.6% of total n -alkanes, respectively) than C_{23} (7.9% and 11.3%; cf. Nott et al., 2000). *Sphagnum recurvum*, *Sphagnum papillosum* and *Sphagnum cuspidatum* typically grow in hollow areas of a bog (Andrus, 1980; Crum, 1988; Hájková and Hájek, 2002) and produce a distribution similar to the average, with C_{23} making up >30% of the total n -alkanes and C_{31} making up 16.1% or less (Nott et al., 2000). Though they occupy different micro-habitats, hummock-loving and hollow-loving species of *Sphagnum* both thrive when conditions in the bog are wetter and both are distinct from vascular plants in terms of n -alkane distributions. Such patterns are not unexpected, given that one of the functions of leaf wax n -alkanes is preservation of water (Eglinton and Hamilton, 1967). Longer chain waxes on leaf surfaces should be more effective in reducing evaporative water loss.

The characteristically different n -alkane chain length distributions between *Sphagnum* and other peat-forming plants may be used estimate the relative contributions of these plants to peat. We adopt the $C_{23}:C_{29}$ ratio for computing the relative abundance of *Sphagnum* and other plants in this study. We chose C_{29} instead of C_{31} n -alkane to represent input from non-*Sphagnum* plants, because C_{29} is relatively rare in *Sphagnum*, but is abundant in other plants. Although C_{31} n -alkane is the most abundant n -alkane in the non-*Sphagnum* species examined, attempts to use $C_{23}:C_{31}$ n -alkane ratio as a paleohydrological indicator in the sediment core (see below) yielded uninterpretable results. We attribute the failure of $C_{23}:C_{31}$ n -alkane ratio to the presence of relatively high amounts of C_{31}

n -alkane in *Sphagnum* species. Fig. 2 shows the bimodality of the average *Sphagnum* n -alkane distribution.

3.2. Model correction to reduce interferences

Previous studies have directly used n -alkane ratios for comparison with other paleoclimate proxies (Pancost et al., 2002; Meyers, 2003; Zhou et al., 2005). However, because *Sphagnum* also produces a small amount of C_{29} and other peat formers produce a small amount of C_{23} (Fig. 2), the direct ratio of the two does not represent two pure end members. We applied a correction to the $C_{23}:C_{29}$ ratio based on the modeled peat n -alkane distributions developed from the averaged n -alkane distributions of both sets of plants (Fig. 2). By assuming a mean n -alkane distribution in *Sphagnum* species and non-*Sphagnum* species, we can potentially eliminate or reduce the portion of C_{23} n -alkane present in peat that is not derived from *Sphagnum*. Likewise, the *Sphagnum*-derived C_{29} n -alkane can also be removed from the total C_{29} n -alkane. This model correction allows us to better estimate the true ratio of *Sphagnum* species to other species in the peat.

We computed the model n -alkane distributions in peat by progressively increasing the fractional *Sphagnum* n -alkane input (relative to other plants) in nine steps from 0.1 to 0.9, based on average n -alkane distributions (Table 1). We multiplied the % abundance for each n -alkane in the average *Sphagnum* distribution by the *Sphagnum* input (f), and multiplied each n -alkane abundance value in the average non-*Sphagnum* n -alkane distribution by the fractional non-*Sphagnum* input ($1 - f$). The sum of the resultant value for each n -alkane constituted a total peat n -alkane distribution (see Table 2).

The modeled n -alkane distributions in the nine different scenarios allow us to compute the $C_{23}:C_{29}$

Table 1
Model distribution of n -alkanes in peat with varying abundance of *Sphagnum*

f	C_{19}	C_{21}	C_{23}	C_{25}	C_{27}	C_{29}	C_{31}	C_{33}	C_{35}	$C_{23}:C_{29}$
0.1	0.0	1.3	3.6	4.1	8.9	20.4	40.7	15.5	0.1	0.2
0.2	0.1	2.0	6.0	5.5	8.6	19.1	38.5	14.7	0.1	0.3
0.3	0.1	2.7	8.3	6.9	8.3	17.9	36.4	13.9	0.1	0.5
0.4	0.1	3.4	10.7	8.2	8.0	16.6	34.3	13.1	0.1	0.6
0.5	0.1	4.1	13.1	9.6	7.7	15.4	32.2	12.3	0.1	0.9
0.6	0.1	4.8	15.5	11.0	7.4	14.1	30.1	11.5	0.1	1.1
0.7	0.1	5.5	17.9	12.3	7.1	12.9	28.0	10.6	0.0	1.4
0.8	0.1	6.3	20.3	13.7	6.8	11.6	25.8	9.8	0.0	1.7
0.9	0.1	7.0	22.7	15.1	6.5	10.4	23.7	9.0	0.0	2.2

f = *Sphagnum* input to model peat, changed from 0.1 to 0.9 in 0.1 increments.

Table 2
Distribution of *n*-alkanes in plants from Bloomingdale Bog

<i>n</i> -Alkane carbon number	<i>Carex</i> sp.	<i>Chamydaphne calyculata</i>	<i>Kalmia polifolia</i>	<i>Polytrichum formosum</i>	<i>Rhododendron groenlandicum</i>	<i>Sphagnum</i> sp. (hummock top)
21	0.000	0.000	0.000	0.011	0.000	0.028
22	0.000	0.000	0.000	0.016	0.000	0.013
23	0.002	0.012	0.004	0.035	0.001	0.071
24	0.000	0.004	0.002	0.014	0.001	0.011
25	0.002	0.017	0.014	0.087	0.008	0.085
36	0.000	0.006	0.009	0.021	0.004	0.015
27	0.010	0.062	0.055	0.135	0.063	0.075
28	0.002	0.019	0.022	0.027	0.008	0.012
29	0.125	0.451	0.549	0.290	0.169	0.103
30	0.009	0.030	0.035	0.023	0.019	0.013
31	0.600	0.339	0.289	0.230	0.463	0.303
32	0.016	0.013	0.007	0.018	0.034	0.029
33	0.233	0.046	0.015	0.093	0.230	0.242

Amounts are reported as fraction of total *n*-alkanes detected.

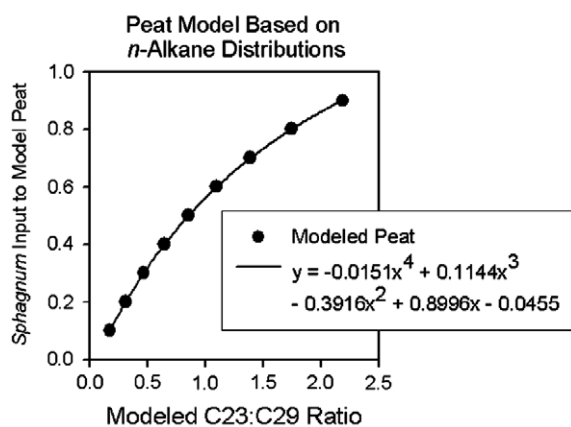


Fig. 4. Sphagnum input to model peat (f) vs. $C_{23}:C_{29}$ ratio based on the model peat's *n*-alkane distribution.

ratio of peats with different % Sphagnum inputs. A fourth order polynomial equation fitted to the resulting plot of Sphagnum input vs. $C_{23}:C_{29}$ ratio (Fig. 4) was subsequently applied to correct the raw $C_{23}:C_{29}$ ratio data for the Minden Bog core.

3.3. Ratios of aquatic to terrestrial plant input

Ratios of *n*-alkanes of different chain lengths have been used as indicators of the relative contributions of aquatic to terrestrial plants to sediments, particularly in lacustrine systems (Ficken et al., 2000; Zhou et al., 2005). For example, $C_{17}-C_{31}$ ratios have been extensively used as an indicator of algae to higher plant input ratios in lake sediments (Meyers, 2003). Submerged vascular macrophytes are characterized by medium chain length *n*-alkanes (Ficken et al., 2000). Their abundance rel-

ative to terrestrial plants (characterized by long chain *n*-alkanes) has been quantified by the “*P*-aqueous” (P_{aq}) ratio as described by Ficken et al. (2000). The ratio (below) compares the abundance of medium chain to the sum of the abundance of medium and long chain *n*-alkanes.

$$\frac{(C_{23} + C_{25})}{(C_{23} + C_{25} + C_{29} + C_{31})} = P_{aq}$$

Because the distribution of *n*-alkanes in aquatic macrophytes is similar to that of Sphagnum (Ficken et al., 2000; Nott et al., 2000; Pancost et al., 2002), the P_{aq} ratio may be a useful index of the relative abundance of Sphagnum to other bog-forming plants. In our subsequent discussion, we compute the P_{aq} ratio values for the *n*-alkanes measured in our core from Minden Bog.

We tested the applicability of a correction to the P_{aq} ratios based on the model distribution of *n*-alkanes as we did for $C_{23}:C_{29}$ ratio (Figs. 2 and 4). We do not, however, report these data, because the difference in correlation with the other proxies between the original values and the corrected values was insignificant.

3.4. *n*-Alkane ratios as paleohydrologic indicators: records from Minden Bog

Proxies used for the reconstruction of paleoclimate from peatlands are typically based on the important role of hydrology in controlling the species distribution patterns and decomposition rates. For example, humification is a measure of relative peat decay and is primarily a function of water balance, with more decomposition occurring during

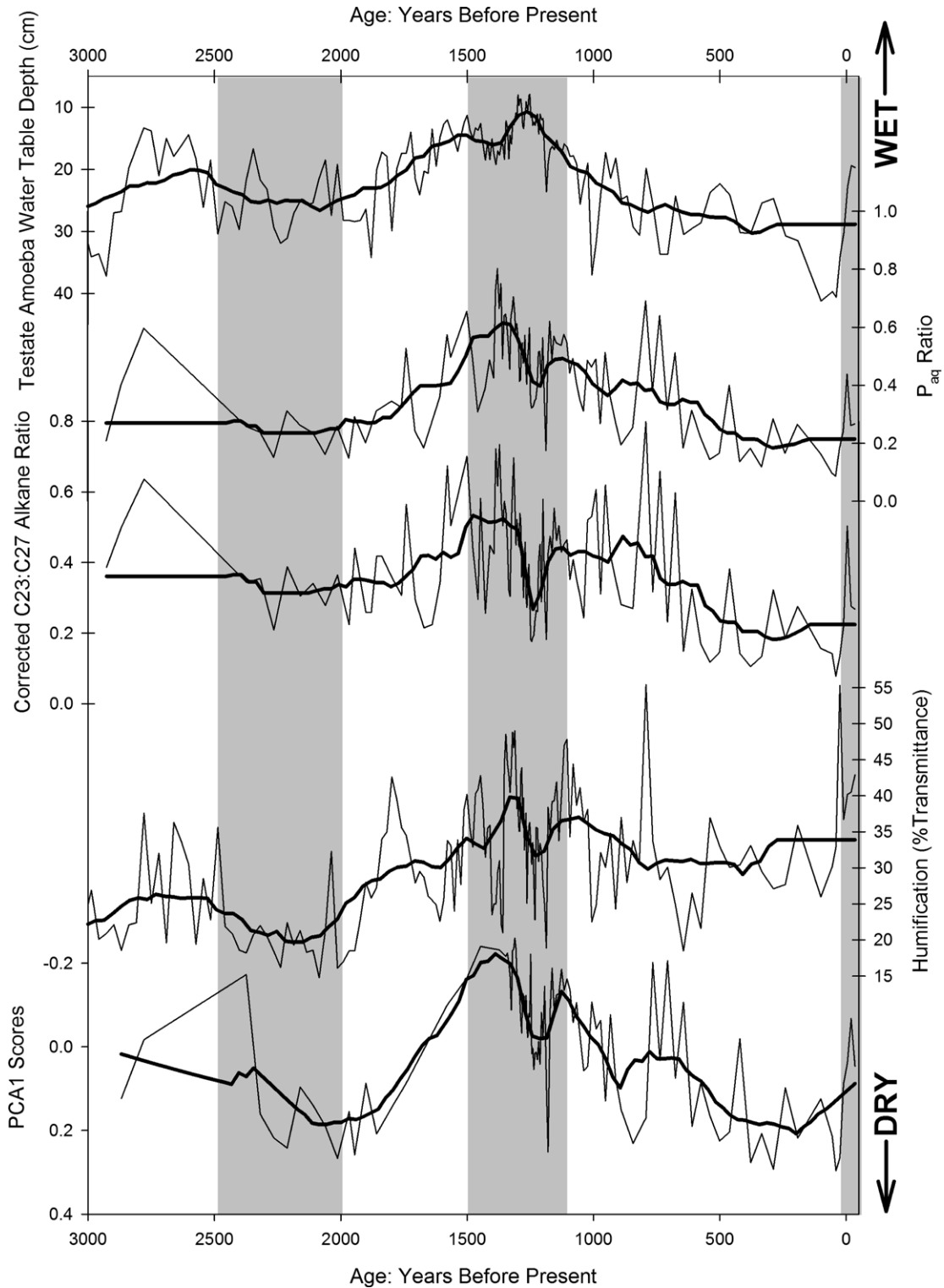


Fig. 5. Paleohydrologic reconstructions from Minden Bog. Testate amoebae water table depth is reported in cm below bog surface and humification index as % transmittance (less light is transmitted through sample as humification increases). *n*-Alkane ratios and PCA axis 1 scores of *n*-alkane distributions in peat are unitless. In the five line plots, values towards the top represent wetter conditions, while values towards the bottom represent drier conditions.

drier periods and less during wet periods. Reconstructions derived from testate amoebae analysis are based on the observation that species distribution is controlled by substrate moisture conditions, allowing the development of transfer functions to infer past changes in water table depth (Woodland et al., 1998; Booth, 2002). Plant macrofossils have been used in a similar way, although they have been less quantitatively calibrated against modern distribution patterns. For example, changes in the relative abundance of various plant taxa, particularly species of Sphagnum, sedges, and ericaceous shrubs, are strongly linked to changes in surface hydrology (Mauquoy and Barber, 2002; McMullen et al., 2004).

The time series of *n*-alkane indicator records and the published testate amoeba and humification reconstructions (Booth and Jackson, 2003) are reported in Fig. 5. Overall, there is excellent agreement among the various proxies. From about 2500 years before present (ybp) to 2000 ybp, all three proxies show values representing drier conditions. There is a trend from drier to wetter conditions between 2000 and 1500 ybp and a reversal to drier conditions from 1700 to 100 ybp. These general climate trends are consistent among the *n*-alkane data sets (Fig. 5) and also between the *n*-alkane and the humification and testate amoebae data (Fig. 5). The patterns of climate variations are discussed in more detail by Booth and Jackson (2003) and Booth et al., (2006). Our results indicate that both model-corrected $C_{23}:C_{29}$ and P_{aq} *n*-alkane ratios are useful paleohydrologic indicators.

To further test whether the total distribution of *n*-alkanes in peat is determined by surface moisture conditions, we performed a PCA on the *n*-alkane distributions for peat samples from Minden Bog (Fig. 5). We included samples where all the odd carbon numbered *n*-alkanes from C_{21} through C_{33} were detected and did not coelute with contaminant substances. All samples had been analyzed previously for humification and testate amoebae (Booth and Jackson, 2003). Axis 1 scores are strongly correlated with both humification ($r = -0.67$) and testate amoeba-inferred water table depths ($r = 0.64$), indicating that PCA1 of *n*-alkane distributions is another robust proxy for past surface moisture.

While correlations among the various proxies are good, there are subtle differences. At approximately 1300 ybp, the testate amoebae record shows a trend towards values representing wetter conditions, whereas the other three proxies show a trend

towards values representing drier conditions. Humification index responds not only to wetness and dryness, but also to peat composition (Caseldine et al., 2000). Sphagnum in peat decays less rapidly than other plants, so humification will increase with a decrease in Sphagnum content (Frolking et al., 2001). Protozoa and plants may respond somewhat differently to changes in the seasonality of moisture or to relative changes in precipitation and evapotranspiration. There are many possible reasons for the subtle differences in the proxies and replicated records from multiple sites are necessary to determine whether the different proxies are responding to different aspects of climate variability or if they are due to local autogenic factors.

The *n*-alkane ratio method of reconstructing paleohydrology is advantageous because it is simple to perform and requires only small samples ($<1\text{ cm}^3$ of peat). Because *n*-alkanes, like other leaf waxes, are produced primarily in the leaves of plants and not the roots (Pancost et al., 2002), compounds produced at a particular time are found in a single horizon and are not spread vertically in a peat section. The immobility of *n*-alkanes in sediment (due to their strong hydrophobic nature and affinity for clay; Huang et al., 1999b), the small necessary sample size, and the ease of rapid analysis can allow high resolution study of peat bog paleohydrology. Further analysis of individual plants from peatlands will allow us to refine our model distributions of *n*-alkanes, thereby improving the correction of *n*-alkane ratios.

4. Conclusions

In peatland systems, the distributions of *n*-alkanes can be used as paleohydrologic indicators. The model-corrected $C_{23}:C_{29}$ *n*-alkane ratio, the P_{aq} ratio and the first principle component of *n*-alkane distributions are all indicative of the abundance of Sphagnum relative to other plants in peat and, to a lesser extent, Sphagnum species variations. The *n*-alkane indicators show excellent agreement with hydrologic reconstructions in a peatland system from humification and testate amoebae-inferred water table depth. Leaf wax *n*-alkanes are deposited sequentially on the peat surface and are virtually immobile, allowing minimal disturbance in stratigraphy. The amount of sample required for *n*-alkane analysis is small. Therefore, the *n*-alkane indicators can be used to create high resolution records of peatland paleohydrology and can be added to the

toolbox of multi-proxy-based peatland paleoclimate reconstruction.

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