Oxygen isotope ratios of oak in east England: implications for reconstructing the isotopic composition of precipitation

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Keywords

Abstract

Annual latewood $\delta^{18}O$ values from oak (*Quercus robur* L.) are presented for the period 1895-1994 for four trees growing in eastern England. We demonstrate, using the $\delta^{18}O$ values of precipitation from Wallingford (1982-1994) and the longer time-series from Groningen (1965-1994), that the $\delta^{18}O$ values of winter precipitation had the strongest influence on the $\delta^{18}O$ values of tree-ring cellulose. The influence of winter precipitation on the spring/summer formed latewood may be explained by several factors: most soil water is replenished during the winter, water may take several months to penetrate the unsaturated zone, and oaks may sample water from lower in the profile during times of water stress. The $\delta^{18}O$ values of oak latewood cellulose were also significantly ($p<0.01$) correlated with summer relative humidity. Therefore, although precipitation $\delta^{18}O$ values may be altered by

partial isotopic exchange with xylem water and the influence of stomatal conductance upon evapotranspiration, some of the original $\delta^{18}O$ signature remains.

Introduction

The establishment of the Global Network of Isotopes in Precipitation (GNIP) by the International Atomic Energy Agency (IAEA) and the World Meteorological Organization (WMO) has enabled the spatial and temporal distribution of stable isotopes in precipitation to be investigated [1-5]. In its simplest form, precipitation may be modelled by a Rayleigh-type distillation process, which assumes isotopic equilibrium between water and vapour. Water evaporating from the ocean is enriched in the ^{16}O as $H_2^{16}O$ has a higher saturation vapour pressure than $H_2^{18}O$. Conversely, raindrops formed by the condensation of water vapour are enriched in ^{18}O because $H_2^{18}O$ condenses preferably to $H_2^{16}O$. Consequently, as rain is condensed from an air parcel, it becomes progressively depleted in ^{18}O.

As the present day spatial relationship between isotope ratios of precipitation and temperature is strongly associated [6], it is often assumed that this relationship has held in the past. However, if climate change alters the seasonality of precipitation or the evaporative origin of the source water, the use of this relationship to reconstruct temporal trends may be inappropriate [7,8]. Although Global Circulation Models (GCMs) have been used to evaluate these assumptions [9,10], further calibration work is required to improve the interpretation of palaeoclimatic data. Unfortunately most of this research has been directed towards investigating spatial relationships.

Temporal trends have been investigated using independent proxy measures of past temperature, such as the noble gas content of groundwater [11] or temperatures inferred from bore holes in ice sheets [12]. White *et al.* [13] demonstrated the association between an annually resolved stacked isotope record at Greenland and environmental variables. Similarly, data from the GNIP database have been used to calibrate this relationship [4,5]. However, as the GNIP database is, at best, restricted to 40-years, a proxy measure for the isotopic composition of precipitation is required.

Although the isotopic composition of source water has been inferred from natural archives, such as lacustrine sediments [14], there has been little research into indirect measures of the isotopic composition of past precipitation. Hydrogen or oxygen isotopes in tree-ring cellulose obtained from tree-ring chronologies may offer this potential. Even if it is unlikely that the tree rings will be a direct recorder of the isotopic composition of precipitation [15], the $\delta^{18}O$ values of tree-ring cellulose are still a valuable paleoclimatic archive [16,17]. This study investigates the $\delta^{18}O$ values of oak growing in eastern England as a proxy measure for the $\delta^{18}O$ value of precipitation.

Materials and methods

Sites

Two sites in eastern England (52°50'N, 0°30'E) with different edaphic conditions were selected for study. Sandringham Park is a relatively dry, free draining site, compared to Babingley Osier Carr which was selected as a wet, poorly drained site [18]. The sites are located approximately 3 km apart. The mean monthly precipitation at Sandringham (1904-1994) is illustrated (fig 1).
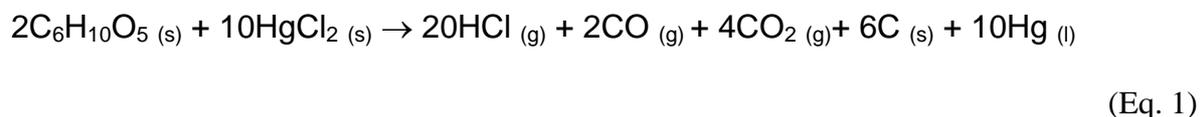
Sampling

Both sites were surveyed accurately and twenty oak (*Quercus robur* L.) trees were cored at each site. Rather than counting back rings, absolutely-dated ring-width chronologies were established. A mean ring-width site chronology for Sandringham (SAND) was developed that spanned the period 1847-1994 and similarly, at Babingley (BABS), for 1843-1994. The chronologies correlate well with other independent chronologies. t-Values of 4.4 (SAND) and 6.5 (BABS) were obtained against the English Midlands oak chronology [19] and t-values of 4.6 (SAND) and 7.5 (BABS) were obtained against the British Isles oak chronology (Baillie, *personal communication, 1997*). These values exceed the minimum acceptable value for an oak time-series of this length (t>3.5) and, as the chronologies were confirmed visually [20], the dating was accepted. Two 12mm diameter cores were obtained at each site for oxygen isotope analysis. The resulting oxygen isotope time-series were pooled by taking the simple unweighted mean values to form the respective site isotope chronologies.

As earlywood cellulose may be formed from carbohydrates assimilated during the previous year [21] and the initial water utilised in earlywood synthesis may be isotopically enriched [22], latewood samples were selected for analysis. Under magnification, annual latewood samples were removed with a razor blade to produce slivers approximately 60µm thick. These slivers were placed into individual Soxhlet thimbles to enable the oxidation of

lignin with acidified sodium chlorite solution. The hydrolysis of hemicelluloses was achieved with the addition of concentrated alkali to yield α-cellulose [23].

The α-cellulose (*ca.* 3-4 mg) was loaded into 125 mm long x 7 mm bore pre-heated quartz tubes (>550°C, 1h) together with 130 mg of anhydrous mercury(II) chloride. A breakseal was made in the loaded tubes which were evacuated to <10^{-3} mbar overnight (>8h) to ensure that all traces of absorbed water were removed. The tubes were sealed with an oxy-gas torch at a length of *ca.* 100mm and heated in a muffle furnace for *ca.* six hours at 550°C.

The overall reaction is given by equation 1. It is believed that the *in situ* generation of hydrogen chloride enables the oxidation of α-cellulose with mercury acting as the catalyst (equation 1):

$$2C_6H_{10}O_{5\ (s)} + 10HgCl_{2\ (s)} \rightarrow 20HCl_{\ (g)} + 2CO_{\ (g)} + 4CO_{2\ (g)} + 6C_{\ (s)} + 10Hg_{\ (l)}$$

(Eq. 1)

The hydrogen chloride must removed before $\delta^{18}O$ determination. This was achieved using a pre-dried (*ca.* 50°C, 48h) macroreticular ion exchange resin, *Amberlyst A-21* [24]. The uncondensable CO was quantitatively converted to CO_2 by a high voltage electric discharge [24, 25]. Cryogenic distillation of CO_2 removed any hydrocarbon impurities [26]. The pyrolysis of α-cellulose, with an excess of mercury(II) chloride, and the subsequent conversion of CO to CO_2 using an electric discharge, is a quantitative reaction that is represented by equation 2:

$$2C_6H_{10}O_5{}_{(s)} + 10HgCl_2{}_{(s)} \rightarrow 20HCl{}_{(g)} + 5CO_2{}_{(g)} + 7C{}_{(s)} + 10Hg{}_{(l)}$$

(Eq. 2)
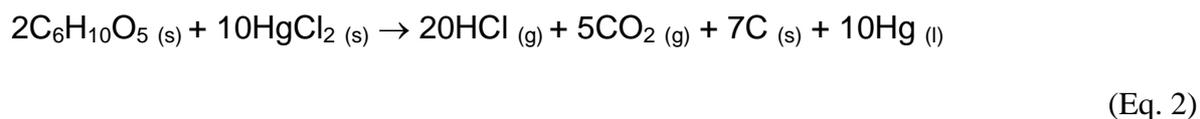
$\delta^{18}O$ values were determined on CO_2 using a VG Isogas SIRA II mass spectrometer and are expressed relative to the VSMOW Standard [27]. The overall precision for $\delta^{18}O$ analysis was typically 0.2‰ (e.g. IAEA-C3 cellulose standard; $\bar{x}$=32.14, n=6, σ_{n-1}=0.20‰).

Results

Relationship with environmental variables

Figure 2 shows the annually resolved mean Sandringham and mean Babingley $\delta^{18}O$ values over the period 1895-1994. An east England oxygen isotope chronology was also calculated, by taking the unweighted mean of the four trees used to construct the site oxygen isotope chronologies. The relationship between annual $\delta^{18}O$ values from latewood cellulose and averaged monthly environmental variables was investigated over the period 1895-1994 for all three chronologies. Latewood $\delta^{18}O$ values had the strongest association with averaged monthly relative humidity recorded at 1 p.m. GMT over the period of available data (1920-1994). The June to September relative humidity had a significant ($p<0.01$) influence on cellulose $\delta^{18}O$ values (figure 3). The combined July/August relative humidity values were usually more influential than relative humidity values of any other month, or combination of months, in determining the $\delta^{18}O$ values of latewood cellulose. Significant correlations were observed between combined July/August relative humidity and the oxygen isotope chronologies the sites of Babingley (*r=0.50; p<0.01*), Sandringham

($r=0.48; p<0.01$) and the east England composite $\delta^{18}O$ chronology ($r=0.57; p<0.01$). This relationship is supported by growth measurements on oaks growing *ca.* 60km from the site, where approximately two-thirds of latewood was formed during these months (Hemming, *personal communication, 1997*). Hence, the relationship between $\delta^{18}O$ values and combined July/August environmental variables was investigated (table 1).

The difference in the response of oak $\delta^{18}O$ values to environmental variables under the different hydrological conditions of Babingley and Sandringham (table 1) was not significant ($p>0.01$). The association between tree-ring $\delta^{18}O$ values and environmental variables is similar to the few studies that have investigated temporal variations in tree-ring $\delta^{18}O$ values at an annual resolution [16, 17, 35-38]. The relationship with environmental variables is not as strong as that reported for $\delta^{13}C$ indices developed at the same sites [18]. However, this result is not surprising, as several studies [4, 5, 10] have reported that, in many regions, the temporal variations in the isotopic composition of precipitation are not strongly correlated with temperature or the amount of precipitation. At Babingley, the standardised ring-widths display an extremely weak link with environmental variables, whereas the association of $\delta^{18}O$ values was significantly greater ($p<0.01$) despite the $\delta^{18}O$ chronology being constructed from far fewer trees.

Influence of $\delta^{18}O$ precipitation

The relationship between oxygen isotope chronologies and the $\delta^{18}O$ value of monthly-averaged precipitation at Wallingford, England (51°36'N, 1°6'W) obtained by taking an unweighted mean, was calculated for the period of available data (1982-1994)

(figure 4). At Babingley, the δ^{18}O values of tree-ring cellulose had the strongest association with the δ^{18}O values of precipitation from January (*r=0.71; p<0.01*) and December of the previous year (*r=0.69; p<0.01*). Whereas at Sandringham, the relationship was more complex, with the δ^{18}O values of tree-ring cellulose being significantly correlated with the δ^{18}O values of precipitation from January *(r=0.56; p<0.05)*, March (*r=0.68; p<0.05*) and October (*r=-0.56; p<0.05*). The δ^{18}O values of tree-ring cellulose from the unweighted east England isotope chronology were influenced predominantly by the δ^{18}O values of precipitation from January (*r=0.79; p<0.01*) and December of the previous year (*r=0.65; p<0.05*).

The east England isotope chronology was selected to investigate the association between the δ^{18}O values of tree-ring cellulose and monthly δ^{18}O values of precipitation. This time-series, calculated as an unweighted mean, had a higher degree of replication than the chronologies from the individual sites and should reduce non-environmental variability. The combined δ^{18}O time-series is more likely to represent trees obtained from long oak chronologies [41] whose precise edaphic environments are unknown. The δ^{18}O values of tree-ring cellulose from the east England isotope chronology and the δ^{18}O values of precipitation from the months of January and the previous December, combined by taking the simple unweighted mean, were strongly associated (*r=0.88; p<0.01*) (figure 5). These results demonstrate that the δ^{18}O values of winter precipitation had the strongest influence on latewood cellulose δ^{18}O values.

As the significant (*p<0.01*) influence of winter precipitation on latewood cellulose δ^{18}O values was based upon the restricted dataset at Wallingford (1982-1994), the

relationship was also investigated for the longer time-series (1965-1994) of $\delta^{18}O$ values of monthly precipitation from Groningen, The Netherlands (53°13'N, 6°34'E) (figure 6). Although the meteorological station at Groningen is situated approximately 450km from the sites in eastern England, the $\delta^{18}O$ values of precipitation should record similar storm trajectories to Wallingford. The association between sites is reflected in the correlation between the monthly $\delta^{18}O$ values of precipitation at Wallingford and Groningen for the period 1982-1994 (*r=0.51; p<0.01*). However, any relationship with east England tree-ring $\delta^{18}O$ values will be diminished owing to evaporation and condensation occurring in the storm. The $\delta^{18}O$ values of tree-ring cellulose from the east England isotope chronology and the $\delta^{18}O$ values of precipitation at Groningen for the simple unweighted averages for the combined months of August, January and the previous December had the strongest association (*r=0.59; p<0.01*). Although the $\delta^{18}O$ values of precipitation from several months may influence the $\delta^{18}O$ values of oak tree-ring cellulose, it is the $\delta^{18}O$ values of predominantly winter precipitation that had the strongest influence at these sites.

Discussion

The association of cellulose $\delta^{18}O$ values with environmental variables (table 1), especially relative humidity (figure 3), and the unweighted $\delta^{18}O$ values of precipitation (figure 4) is supported by field and greenhouse observations [15]. Although the relative humidity signal is a direct result of the influence of stomatal conductance on evaporative enrichment, there is subsequent partial isotopic exchange of carbohydrates with xylem water at the time of cellulose synthesis subsequently modifying these $\delta^{18}O$ values.

The δ^{18}O values of latewood cellulose are clearly influenced by the δ^{18}O values of precipitation from January and the previous December, however this relationship was not reflected in the influence of monthly environmental variables (figure 3). Only the associations with relative humidity values from the months of June to September were significant (*p<0.01*). Initially, these results appear to contradict each other. Latewood oak cellulose δ^{18}O values, formed in the spring/summer, are influenced strongly by the relative humidity values throughout the summer and yet, it is the δ^{18}O of precipitation values from the winter months that exert the strongest influence.

The influence of summer environmental parameters on tree-ring cellulose δ^{18}O values may be explained by the effects of climate upon stomatal conductance and hence, evaporative enrichment during these months. However, the δ^{18}O values of summer precipitation only have a minor influence on tree-ring cellulose δ^{18}O values, as evaporative losses are high in the summer [42].

The strong influence of winter precipitation δ^{18}O values on tree-ring cellulose δ^{18}O values may be explained by the fact that most of the soil water reserves in east England are replenished during the winter [43]. The increased precipitation in autumn (September - November) and winter (December – February) is supported by precipitation data from Sandringham (figure 1). Infiltration of this water is predominantly through the soil matrix and conduction rates may vary considerably depending upon the physical characteristics of the soil. In general, the coarser the soil particles, the larger the intervening voids and faster conduction of water. Conduction rates of 4.5m year^{-1} have been reported for coarse soils, rich in sand and gravels and 1m year^{-1} for finer soils such as clays [44]. Although it is difficult to estimate infiltration rates, it would be reasonable to assume that, at Babingley,

water may take several months to move through the unsaturated zone and reach the rhizosphere. However, at Sandringham, with its rapidly draining soils, the trees rely heavily on regular replenishment from meteoric sources and hence, the reliance upon meteoric water supplies throughout the year.

The Babingley (and east England) $\delta^{18}O$ results contrast with previous studies [16, 17], where the $\delta^{18}O$ values of beech and spruce were influenced by the $\delta^{18}O$ values of growing season precipitation. The lack of response to summer precipitation has also been reported for woody perennial plants growing in semi-arid environments [22]. This difference in response to monthly precipitation $\delta^{18}O$ values can be partly attributed to the contrasting hydrological conditions at the sites and also by variations in root structure. Whereas oak has a deep root system [45], beech and spruce are generally shallow rooted and therefore, utilise precipitation in the unsaturated zone more rapidly than oaks. In all these studies, the amplitude of the $\delta^{18}O$ signal in tree ring cellulose is damped compared to the $\delta^{18}O$ value of precipitation (figure 5). The reduction in signal strength may be attributed to several processes including the use of groundwater, isotopic fractionation during evapotranspiration and post-photosynthetic exchanges [15, 46].

The influence of the $\delta^{18}O$ values of winter precipitation on the $\delta^{18}O$ values of oak cellulose can also be used to explain the previously reported relationship between oak earlywood and latewood $\delta^{18}O$ values of the same year [21, 36]. Throughout the period of earlywood and latewood formation the trees must sample water, which primarily reflects the $\delta^{18}O$ value of winter precipitation. The slight enrichment in latewood $\delta^{18}O$ values reported by Field [36] can be explained by the fact that, during the summer months, there is additional water loss through evapotranspiration leading to enrichment in ^{18}O of the

remaining water. This latter observation supports the influence of summer climate on latewood $\delta^{18}O$ values (figure 3). However, these results contrast to those reported for Gambel's Oak where enrichment in early season xylem water $\delta^{18}O$ values was found [22].

Conclusions

Although the $\delta^{18}O$ values of oak latewood cellulose are influenced by the $\delta^{18}O$ value of precipitation from several months, it is primarily the $\delta^{18}O$ value of winter precipitation that has a significant (*p<0.01*) influence. Summer environmental variables, reflected in the association with monthly relative humidity values, will alter this value through changes in stomatal conductance. Subsequent isotopic exchange that occurs during biochemical reactions and post-photosynthetic processes will also have an influence [15, 46]. Despite the complexity of these reactions, some of the original $\delta^{18}O$ signature of winter precipitation remains and hence, $\delta^{18}O$ value of tree-ring cellulose may be used as a proxy measure for the isotopic composition of precipitation.

This preliminary study was based on a limited number of time-series, owing to the time required to measure $\delta^{18}O$ values; therefore the results do not allow us to determine the degree of replication required to construct oxygen isotope chronologies representative of the macro-environmental changes. Fortunately, recently developed on-line techniques for the determination of $\delta^{18}O$ from organic matter [47-50] now make this possible.

Acknowledgements

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Figure 1. *Mean monthly precipitation at Sandringham (1904-1994).*

Figure 2. *Annual mean Sandringham (trees SP10 and SP19) and mean Babingley (trees BW04 and BW05) oxygen isotope values (1895-1994). The inter-site correlation (r=0.53) was significant (p<0.01). The mean Sandringham $\delta^{18}O$ value was 29.38‰ (n=196; σ_{n-1}=1.13; standard error=0.08) and the mean Babingley site $\delta^{18}O$ value was 29.40‰ (n=200; σ_{n-1}=0.92; standard error=0.07). The overall precision was ca. ±0.2‰.*

Figure 3. *Relationship between unweighted oxygen isotope chronologies and averaged monthly relative humidity values recorded at 1 p.m. GMT (1920-1994). The east England $\delta^{18}O$ chronology (n=4) consists of the combined annual Sandringham (n=2) and Babingley (n=2) $\delta^{18}O$ chronologies. The monthly relative humidity values of the current year are labelled with capital letters; lower case represents the monthly relative humidity of the previous year. Correlations are represented by the Pearson product moment correlation coefficient (r). Solid bars indicate the correlation is significant at p<0.01.*

Figure 4. *Relationship between unweighted oxygen isotope chronologies and the oxygen isotope value of monthly precipitation at Wallingford (1982-1994). $\delta^{18}O$ values of monthly precipitation data were obtained from the British Geological Survey, Wallingford [39, 40]. The monthly $\delta^{18}O$ values of precipitation of the current year are labelled with capital letters; lower case represents the monthly $\delta^{18}O$ values of precipitation of the previous year. Correlations are represented by the Pearson*

product moment correlation coefficient (r). Solid bars indicate the correlation is significant at p<0.01; diagonal lines indicate the correlation is significant at p<0.05.

Figure 5. *Relationship between east England tree-ring cellulose $\delta^{18}O$ values and the $\delta^{18}O$ values of precipitation from the previous December/January at Wallingford [39, 40] over the period of combined data (1982-1994).*

Figure 6. *East England tree-ring cellulose $\delta^{18}O$ values for the period 1965-1994; error bars represent the standard error of the mean. Site precipitation $\delta^{18}O$ time-series, calculated from the individual months with significant (p<0.01) correlations with the tree-ring cellulose $\delta^{18}O$ values are illustrated. For Wallingford, the combined $\delta^{18}O$ values of precipitation were calculated for the previous December/January for 1982-1994 [39, 40], whereas at Groningen, the $\delta^{18}O$ values of precipitation were calculated for the previous December, January and August for 1965-1994 ([40]; Meijer, personal communication, 1997).*

Table 1. *Correlation between site oxygen isotope chronologies and July/August environmental variables.*

	Sandringham		Babingley	
	$\delta^{18}O$	Standardised Ring-widths	$\delta^{18}O$	Standardised Ring-widths
Raw Environmental Variables				
Temperature	0.34 †	-0.20 *	0.38 †	0.00
Relative humidity	-0.48 †	0.27 *	-0.50 †	0.05
Vapour pressure deficit	0.47 †	-0.28 *	0.51 †	-0.05
Rainfall	-0.37 †	0.13	-0.41 †	0.06
PDSI	-0.40 †	0.20	-0.35 †	0.06
Reconstructed river flow	-0.45 †	0.43 †	-0.30 †	0.25 *
High-Frequency Environmental Variables				
Temperature	0.33 †	-0.18	0.43 †	0.01
Relative humidity	-0.53 †	0.36 †	-0.49 †	0.08
Vapour pressure deficit	0.48 †	-0.28 *	0.50 †	-0.04
Rainfall	-0.32 †	0.11	-0.37 †	0.06
PDSI	-0.40 †	0.22 *	-0.38 †	0.07
Reconstructed river flow	-0.40 †	0.22 *	-0.38 †	0.07

Averaged monthly temperature values were calculated using the mean 24 hour maxima and minima daily values read at 9 a.m. GMT from the CET record for 1895-1994 ([28, 29] P.D. Jones and E.B. Horton, *peronal communication, 1997*). Relative humidity values are given for 1 p.m. GMT from the combined Cranwell-Waddington record for the period 1920-1994 [30]. Vapour pressure deficit values were calculated using relative humidity values at 1 p.m. GMT from the combined Cranwell-Waddington record and CET values for 1920-1994 [31]. Rainfall was measured at Sandringham for the period 1904-1994. Palmer Drought Severity Index (PDSI) values, representing various degrees of water shortage or water excess, for a 5° x 5° box grid centered upon 52°30'N, 2°30'E were calculated for 1895-1991 [32] and Great Ouse reconstructed river flow was calculated for 1895-1992 [33]. Standardised ring-with data based upon 25 cores at Sandringham and 20 cores at Babingley [18, 34]. Here, correlations are represented by the Pearson product moment correlation coefficient.

- Significance level *p*<0.05; † Significance level *p*<0.01.