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ABSTRACT

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A compilation for the northwest of Europe of over 500 uranium-series speleothem and travertine dates is presented using a cumulative distributed error frequency approach. These secondary carbonate deposits require both significant groundwater supply and a biogenic soil carbon dioxide source for their growth. During glacial periods soil CO_2 production is inhibited and the water supply is ice locked, therefore growth is slowed significantly if not stopped. Thus deposits can be used on a chronological basis to give a signal of glacial and non-glacial periods. However, sensitivity to the groundwater supply modifies this signal, with growth also limited in times of high aridity, the palaeoclimatic signal is thus complex. The compilation presented is used both as a chronology for comparison with the orbitally tuned marine oxygen isotope record, and as a palaeoclimatic indicator. Attention is focused on two periods of the last glacial / interglacial cycle where the record provides a significantly different palaeoclimatic record to other terrestrial and oceanic records. These are the isotope stages 5/4 transition, for which a relatively low cumulative growth frequency indicates an earlier increase in aridity than observed elsewhere; and isotope stage 3, the pleniglacial, where both statistically significant high (49–62 ka) and low (22–35, 44–46 ka) levels of growth are observed, and can be used to constrain the timing of many of the interstadial and stadial events within this period.

Introduction

The growth record of secondary carbonate deposits offers potential as a measure of climatic change on the continents. The deposits investigated here consist of speleothem, which forms as stalagmite, stalactite and flowstone within cave systems, and spring deposited travertine. Although secondary carbonates have the potential to provide many different types of climatic signals, including palaeotemperature proxies from oxygen isotope values and fluid inclusions (Hendy, 1971; Thompson et al., 1976), and measures of the type of plant cover from carbon isotope studies (Brook, 1990), and pollen included within the deposits (Bastin and Gewelt, 1986), the simplest is that signal provided solely by the presence or absence of carbonate deposition at a particular time and place (Gascoyne et al., 1983). This last method is the one developed here, and provides a complex palaeoclimate signal.

The most important mechanism for conventional speleothem and travertine deposition is the degassing of groundwaters which contain elevated carbon dioxide concentrations (Ford and Williams, 1989; Gordon et al., 1989); other growth mechanisms are possible but uncommon, for example common-ion effects (Atkinson, 1983) and evaporation (Harmon et al., 1983). The elevated carbon dioxide is derived from the high partial pressures of CO_2 in the soil atmosphere, generated by microbial processes and root respiration (Dorr and Munnich, 1986). This gives a climate related signal; soil CO_2 production is strongly correlated to temperature (Dorr and Munnich, 1989), and high soil

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 CO_2 concentrations and speleothem and travertine growth are thus associated with warm periods. In regions of cold climate, with lower CO_2 production levels (Poole and Miller, 1982) and thus lower soil CO_2 concentrations (Solomon and Cerling, 1987) and a shorter growing season, growth is thought to be much slower if not completely inhibited, as indicated by sparse speleothem growth in arctic or high alpine regions today. Furthermore, recharge may be reduced or cease in times of permafrost and ice advance (Kane and Stein, 1984), and growth is thus restricted or halted. During warm periods growth may also be limited at times of extreme aridity when little surplus water is available for recharge (Brook et al., 1990).

Secondary carbonates may be dated with considerable accuracy by uranium series dating which can provide ages up to 350 ka (Schwarcz, 1980). Thus despite ambiguities in the exact palaeoclimate signal they may represent, their good age control can be used to obtain precise chronological record of these events. Gordon et al. (1989) employed a cumulative distributed error frequency curve to determine growth frequencies for the time period 20–160 ka using a data set of 340 uranium series analyses derived from the United Kingdom. In this paper we use this technique, with modifications, to analyse over 500 dates from North and West Europe, which are then compared to other terrestrial records of climate for the period 20–160 ka.

The growth record for Northwest Europe

Data sources

The geographical area considered in this compilation is shown in Fig. 1 and is characterised by having been ice-covered or marginal to ice sheets during glacial stages. The present-day climate is temperate with rain at all seasons and cool short summers (temperate oceanic climate, type D; Lydolph, 1985). Any spatial variations in temperature that exist within the region today are small compared to the magnitude of climatic variations in the Quaternary. Mean annual temperatures have been estimated to have varied by approximately 15°C between glacial and interglacial stages (Atkinson et al., 1987).



Fig. 1. Location of karst regions from which samples in this compilation were derived.

Data for this compilation (Table 1) has been derived from published and unpublished sources. The dates were screened for accuracy before being included in the data set; analyses which showed evidence of contamination (activity ratio of $Th^{230}/Th^{232} < 20$) or of analytical problems (thorium or uranium yield of less than 10%) were excluded. Analyses yielding infinite ages were also omitted. The remaining dates were then recalculated from the isotopic analytical ratios using a single ²³⁰Th decay constant of 9.195×10^{-6} a⁻¹, because different laboratories have previously used different values. Failure to do this, as in the previous study of Gordon et al. (1989) has a considerable influence on the timing of growth maxima and minima. The cumulative secondary carbonate growth frequency was then calculated using the method of Gordon and Smart (1984) by summing the distributed error probabilities for all individual analyses using a 500 year time interval (Fig. 2b).

Statistical testing

Rigourous testing of the significance of individual peaks and troughs in growth frequency is necessary if there is to be confidence when interpreting the cumulative frequency growth curve in terms of the timing of glacial, interglacial, stadial and inter-

TABLE 1

Location	Number	Source			
Sutherland, Scotland	15	Ford, D.C. (unpublished)			
		Atkinson et al. (1986)			
Gower, Wales	9	Ford, D.C. (unpublished)			
		Stringer et al. (1986)			
		Sutcliffe and Currant (1984)			
Tawe Valley, Wales	16	Christopher (unpublished)			
Pontnewydd, Wales	39	Schwarcz, Ivanovich et al.,			
		Debenham et al., in Green (1984)			
Mendip Hills, England	57	Ford, D.C. (unpublished)			
		Smart (unpublished)			
		Atkinson et al. (1978, 1984)			
Derbyshire, England	52	Ford, T. et al. (1983)			
		Ford, D.C. (unpublished)			
		Rowe et al. (1989)			
Yorkshire, England	176	Atkinson et al. (1978)			
, C		Gascovne (1979)			
		Latham et al. (1979)			
		Gascoyne et al. (1983)			
		Sutcliffe et al. (1985)			
Devon, England	12	Proctor (unpublished)			
County Clare, Ireland	12	Ford, D.C. (unpublished)			
Ardennes, Belgium	52	Schwarcz (unpublished)			
		Gewelt and Juvigne (1986)			
		Quinif (1986, 1989), Gewelt (1985)			
		Bastin and Gewelt (1986)			
Erfurt region, Germany	34	Brunnacker et al. (1983)			
		Blackwell and Schwarcz (1986)			
		Schwarcz et al (1988)			
		Harmon et al (1980)			
Stuttgart region, Germany	26	Hennig (1979), Grun et al (1982)			
North Rhine, Germany	20	Hennig (1979)			
	Total: 520				

Geographical area, number of analyses and source of uranium seriesages on secondary carbonate deposits included in the compilation for North-West Europe

stadial events. This can be undertaken by generating random data sets having the same range and analytical uncertainty as the actual data set; such an approach was previously used in the evaluation of C¹⁴ dated sea-level data (Geyh, 1980; Shennan, 1979). Data sets of 500 random ages were generated with standard deviations equal to 0.075 of the age (the average value of the one standard deviation uncertainty of the real alpha-spectrometric dates). Results of 6 such runs are shown in Fig. 2a. A set of 40 runs was used to determine the 95% (c. 2σ) probability bounds for individual peaks and troughs being generated solely by chance. Thus in Fig. 2b parts of the curve above and below this probability band are said to be significantly favourable or unfavourable for deposition at a 95% confidence level respectively. Peaks or troughs which are discussed in the text are labelled A–J on Figs. 2b and 3a.

The growth frequency record as a chronology

The cumulative growth frequency record presented here can be used to define the timing of warm (enhanced growth) and cold (limited growth) periods by taking the timing of the peaks and troughs as a simple binary signal. The record is compared with the orbitally tuned oxygen-isotope



Fig. 2a. Cumulative frequency distribution of 500 randomly generated uranium series ages, using a standard deviation of 0.075 of the age. b. Cumulative frequency distribution of 520 uranium series analyses from Northwest Europe; 95% error bars generated from random data runs shown by the hashed lines. Parts of the curve above the lines have significantly high levels of growth, those below significantly low. Peaks and troughs discussed in the text are labelled A-J.

chronology of Martinson et al. (1987), Fig. 3b. For isotope stage 5 we employ the generally used substages as defined by Shackleton (1969), while in stages 2–4 the comparisons are made with isotope events as defined by Pisias et al. (1987).

Initial very high levels of growth frequency (A on Fig. 3a) correlate with the Holocene interglacial, the large number of dates under this peak create a tail, adversely affecting the timing of the isotope stage 2 maxima (event 2.2; trough B). Peak C, although not statistically significant, provides evidence of an improvement in climate and probably correlates with event 3.1. Peaks D, FI and F2give a good correlation with events 3.13, 3.3 and 3.31. The double peak structure of peak F is significant; the runs of random dates generated double peaks in fewer than 10% of all cases. The presence of these in the cumulative frequency curve

is therefore unlikely to be due to random fluctuations, and the F1/F2 doublet is probably real. The significantly low level of growth at trough E does not correlate with any recognisable event in the orbitally tuned oxygen isotope chronology, but does correlate with a cool and arid period in the Grande Pile pollen record (Fig. 3c), a point discussed later. In stage 5, peak H and trough I could correlate with the isotope sub-stages 5a and 5b, or alternatively be fluctuations within stage 4. The multiple peaks J1-J3 thus could either contain the whole of stage 5, or alternatively just substages 5c-5e, a debate developed in the next section. Peaks J1–J3 can not be adequately differentiated; this may be due to favourable palaeoclimatic conditions throughout this period; more probably it is due to the large counting errors associated with analyses of this age (typically 5-10% at 110 ka), which obscures any possible reductions in growth frequency.

The growth frequency record has a reliable and internally consistent radiometric time-base, and thus provides a useful alternative to the orbitally tuned oxygen isotope record which is widely used as a general Quaternary timescale. However, precise interpretation of the palaeoclimatic signal provided by secondary carbonate deposits is difficult due to the dependence of growth on both warm and/or wet conditions for growth. To obtain a better understanding of the signal they provide, correlations need to be sought with other terrestrial palaeoclimate records which are equally well dated or which are stratigraphically unambiguous. In this respect, two areas will be investigated further; the stage 5/4 transition, where growth frequency levels are significantly different from the oxygen isotope record, and the pleniglacial (stage 3), where statistically significant oscillations in climate are much better defined than in the oxygen isotope record, and better dated than in other terrestrial palaeoclimate records.

The last interglacial

Study of the period of climatic decline from the last interglacial is important as it may provide an analogue for changes in the present climate (Bowen, 1990). Two possible palaeoclimatic interFrequency (dates per 500 years)

(dates per 500 years)

Frequency

Annual temperature (°C)

Annual precipitation (mm)

0

-200

-400

-600

-800 ό

20

40

60

1.5



Fig. 3. Comparison of palaeoclimatic records for the period 0-160 ka. a. The secondary carbonate growth record. b. Oxygen isotope record (Martinson et al., 1987; events from Pisias et al., 1984) and coral growth frequency (Smart and Richards, 1992). c. Grande Pile pollen record of temperature and precipitation deviations from present (Guiot et al., 1989).

80

Age (ka)

100

120

160

140

pretations can be made from the growth frequency record for this period. The first is that the statistically significant peaks J1-J3 correlate with substages 5a-5e, and that H and I are oscillations within the stage 4 glacial. This interpretation, however, is unlikely on purely chronological grounds. The period of significant growth at peak J1 terminates at 87 ka, much earlier than the recognised termination of this sub-stage from orbital tuning $(79.3 \pm 3.6 \text{ ka}; \text{ Table 2})$. The record of the periods of active coral reef formation (Smart and Richards, 1992; Fig. 3b), which reaches a maximum when global ice volumes are at a minima, and which is also based on uranium series dates, agrees with this timing $(81.5 \pm 5.5 \text{ ka})$. Thus termination of significantly favourable climatic conditions in substage 5a at 87 ka is not supported by these global climate records. Other terrestrial records of a sub-stage 5a event are chronologically not precisely constrained. For instance, recent uranium series estimates on peat from the Chelford interstadial site, recognised by some authors as correlated with stage 5a, have considerable uncertainties (86 ± 24 ka; 1σ), permit-

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ting correlation anywhere between stage 3 and substage 5c (Heijinis and Van der Plick, 1992).

A second hypothesis, that either one or both of peak H and through I are within sub-stage 5a, must thus be considered. However, it is then necessary to consider palaeoclimatic explanations for the low growth frequency at this time, compared with earlier stage 5 warm periods (Fig. 3a). Such an explanation could either be related to a decrease in temperature or an increase in aridity. The palaeotemperature record from pollen in Northwest Europe suggests that isotope stage 5 had three periods of interglacial or interstadial character; the British Ipswichian and European Eemian (substage 5e), followed by the European Brørup (substage 5c) and Odderade (sub-stage 5a). The Brørup is actually bipartite, being divided into the Danish and Dutch Brørup and Amersfoort interstadials, separated by a cooler period (Zagwijn, 1990). The record at Grande Pile (Guiot et al., 1989; Fig. 3c) does not show the Eemian to be significantly warmer than the subsequent interstadials. Conversely, Zagwijn (1990) observes a more northerly

TABLE 2

Timing of isotope stages from different chronological records. Oxygen isotope dates from Martinson et al. (1987), coral reef uranium series ages from Smart and Richards (1992). Statistical significance of speleothem growth record shown, \pm and - indicating significantly low and high

Statistical significance of speleothem	growth record shown,	+ and -	indicating	significantly	low	and	high
levels, respectively. For reasoning beh	ind correlations see text	•					

Isotope	Stage	Orbital tuned	Coral reef	Secondary carbonate growth periods			
	event	oxygen isotope		Peak	Timing	95% significant?	
2	2.2	17.9 <u>+</u> 1.4		В	22-27	Yes (-)	
	3.1	25.4 ± 5.9	33.0 ± 2.5	С	28-31	Yes (-)	
	3.13	43.9 ± 4.7	40.5 ± 6.5	D	35-42	No	
3	_	_		E	44-46	Yes (-)	
	3.3	50.2 + 3.9	50.0 + 2.0	Fl	49-56	Yes $(+)$	
	3.31	55.5 ± 5.0	62.5 ± 6.0	F2	56-62	Yes (+)	
4	4.22	65.2+6.1					
•	4.23	68.8+4.2		G	63-71	Yes(-)	
	4.24	70.8 ± 4.0		-			
		-)		(H	72-78	No	
	5.1	79.3 ± 3.6	81.5 ± 5.5	1 a	79-81	Yes (-)	
		_	_	(J1	87-98	Yes (+)	
5	5.33	103.3 ± 3.4	102.5 ± 2.5	J2	98-115	Yes (+)	
	5.5	123.8 ± 2.6	122 ± 15.0	J3	115-133	Yes (+)	

limit of *Abies* in the Eemian in Northwest Europe than in the Brørup or Odderade, and the evidence from British pollen records supports this suggestion with the presence of *Acer monsperssulanum* at several Ipswichian sites including Stone and Trafalgar Square (Godwin, 1975, p. 171). Cooling of the sub-stage 5a palaeoclimate compared to that in 5c is not evident either in the German pollen record (Gruger, 1990; Behre, 1989), where a mean July temperature of perhaps $13-15^{\circ}$ C for sub-stages 5a and 5c is postulated, or in the record from Grande Pile (Guiot et al., 1989).

The oxygen isotope record, Fig. 3b from Martinson et al. (1987), suggests from ice volume evidence that the Eemian was warmer than the other two sub-stages. This is also supported by the presence of a distinctive warm "hippopotamus fauna" in Victoria Cave, Yorkshire, associated speleothem dates range from 135 ± 8.5 to 114 ± 5 ka (Gascoyne et al., 1981, 1983). Some evidence of a deterioration of climate in England at the end of stage 5 is available however, with the presence of a cold fauna including wolverine (*Gulo gulo*) at Stump Cross Caverns, Yorkshire, overlain by speleothem dated to 83 ± 6 ka (Sutcliffe et al., 1985).

Thus the pollen record provides no evidence of a significant difference in temperature between substages 5a and 5c. The faunal record at Stump Cross may signify a cooling of the climate at the end of stage 5a (an earlier timing would be in contradiction with the pollen records presented above), but its age constraint is insufficient to be certain as to its relevance here. Thus we must consider if the decrease in growth frequency during stage 5a could be due to a decrease in precipitation. Unfortunately palaeoprecipitation indicators are not well developed for the fossil record. Kashiwaya et al. (1991), in a spectral analysis of spelothem abundance in Britain, suggested that in the period 70-100 ka variation in water abundance was as important for speleothem development as that of temperature. Wind blown sands are evident in many cave sites on the south coast of Britain and could possibly provide an indication of increased aridity although they may simply be a function of sediment availability due to lowering sea-levels. At present they can only be constrained to a post-Ipswichian age at Bacon Hole (Stringer et al., 1986) and to between 110 and 55 ka at Minchin Hole (Proctor, unpublished data). Loess deposits are widespread through the region considered in this compilation, although the ages of many have been determined using thermoluminescence dating, there is still considerable uncertainty as to their accuracy when applied to sediments over 50 ka (Wintle, 1990). A palynological palaeoprecipitaion record has been developed by Guiot et al. (1989) at Grande Pile. The average annual precipitation estimated for stage 5e is similar to that for stage 5c (c. 200 mm below present levels), with a relatively minor decrease associated with the intervening cooler stage (Fig. 3c). Stage 5a is on average slightly drier (c. 300 mm below present levels) and is preceded and followed by very arid intervals (although there is less certainty about the 5b aridity, Seret, pers. comm., 1992). This evidence thus supports our suggestion that decreased speleothem and travertine growth in stage 5a is caused by a reduction in effective precipitation.

The increase in aridity in substage 5a could be associated with the preliminary stages of the glacial advance which culminated in stage 4. If the Fennoscandian ice advance commenced during stage 5, this could lead to the development of a semipermanent high pressure systems over the continent (Spaulding, 1991), which would result in a drier, more continental climate. The very low level of secondary carbonate growth evident in isotope stage 4 (trough G) is certainly an important indication of the subsequent severity of climate. There is also more direct terrestrial evidence for the timing and extent of this glacial advance. Recent thermoluminescence dates on glacial deposits from Denmark and Poland signify ice advances around 60-70 ka (no error bars quoted) (Kronberg, 1988, quoted in Olsen, 1990); and the most recent TL dates from Danish meltwater deposits have yielded ages in the range $73-75\pm7$ ka (Kronberg and Mejdahl, 1990), confirming an early glaciation. There is also renewed debate over an Early Devensian (stage 4) glaciation in Scotland and North England (Bowen, 1990); although at this time Worsley (1991) contends that the evidence for such an advance is inconclusive.

We therefore conclude from the limited evidence available that substage 5a in Europe, although as warm as the earlier substages 5c and 5e, was significantly drier, probably due to the initial stages of growth of the Fennoscandian ice sheet which caused development of a more continental climate over much of Northwest Europe. Further work including the analysis of the stable isotopic composition of speleothem calcite and fluid inclusions (Schwarz, 1986) may be instructive in testing this hypothesis.

The isotope stage 3 (Pleniglacial) record

The secondary carbonate growth frequency record provides a detailed record of climate change in isotope stage 3; other deposits from this period often provide good palaeoclimatic records but have poor chronological control (for example, the coleoptera record of the "Upton Warren" interstadial gives an excellant temperature record but is constrained only by a minimum radiocarbon ages of c. 42 ka; Coope, 1975). The growth frequency record shows periods of significant climate change which are precisely dated, and can thus be used as a temporal framework into which these other records may be fitted. The record shows two distinct periods of enhanced deposition in the pleniglacial period, from approximately 35 to 42 ka (peak D; event 3.13), and from 49 to 62 ka (peaks Fl and F2; events 3.3 and 3.31), with an intervening period of below average growth (trough E; an undefined event in Pisias et al., 1987 and Martinson et al., 1987). This is in agreement with the division of isotope stage 3 into three substages suggested by Pujol and Turon (1986). Statistically, this record indicates significantly warm (wet) 3.3 and 3.31 events, followed by the significantly cool (dry) and short duration undefined event at trough E. This trough corresponds with a cold, dry period in the Grande Pile record, although the very wet period immediately preceeding this and corresponding with peak F1 has similar growth frequency to peak F2 which correlates with a drier interval. Further, chronologically less well constrained, evidence of a stage 3 cool and dry period has been found by Ran et al. (1990) in the Netherlands, where ice-wedge casts were dated to a minimum age of 36.6 ka, and overly sediments containing pollen of a chionophilous dwarf shrub tundra. If this correlation with

the speleothem trough is correct it indicates a mean annual temperature of -2.4 to 6°C, discontinuous permafrost and relatively dry conditions. Van Vliet-Lanoe (1990) also suggests discontinuous permafrost in Northwest Europe during this time, although exact timing is unknown. This cool period can not however be found in two more southerly climate records; the pollen record at Les Echets (Guiot et al., 1989) and the mollusc record in loesses at Alsace (Rousseau and Puisségur, 1990). This may indicate a strong N-S climatic gradient, as suggested by Pons et al. (1990), from the French pollen record, or perhaps a limited duration further south.

The most northern European stage 3 interstadial site with a good temporal constraint is probably the "Oerel" peat deposit in Germany (Behre, 1989), which directly overlays three peat beds correlated with isotope stage 5. This probably correlates with the significant F1/F2 peaks (events 3.3/3.31), its pollen of Betula nana, Salix, Juniperus, Calluna and *Empetrum* indicating a treeless shrub tundra with moist conditions. Placing other interstadial sites is more problematic; Behre (1989) notes that the Dutch sites of Moershoofd, Hengelo and Denekamp may not even be interstadial in character since the presence of peat bogs in itself is not a climatic indicator because they can form solely due to geomorphic factors. Their chronological positioning based on radiocarbon dates is also not justified as the technique is at the limit of its dating range and the published dates from the three sites form a continuous scatter rather than individual peaks (see graph in Zagwijn, 1983). The Upton Warren interstadial possibly correlates with the period 49-59 ka (peaks F1/F2), but again a better time constraint for this site is necessary. The growth frequency record also shows a very limited growth period at about 29 ka (peak C), possibly correlating with a stage 3.1 event. This is also recorded terrestrially by a weak arctic brownearth soil in Germany dated to approximately 28-30 ka by TL analysis of overlying loess and by C14 dating (Zöller et al., 1988; Zöller and Wagner, 1989).

In conclusion, over this time period the climate of Northwest Europe was cool, with frequent periods of climatic oscillation. Three phases of improved (warm and/or wet) conditions, events 3.31, 3.3 and 3.13 are here dated at 59-62, 49-56 and 35-42 ka (with perhaps also a very slight climatic improvement occurring in event 3.1 (28-31 ka); the many European "interstadial" sites probably fall within these events but detailed correlations are not presently possible. A significant cool and/ or arid phase (trough *E*) occurs at 45 ka, but is not represented in the oceanic record, possibly due to it's brief duration and the smoothing of the ocean core record by bioturbation.

Conclusions

It is argued here that secondary carbonate growth phases are precisely dated but provide a complex palaeoclimatic indicator sensitive to both temperature and aridity. Independent changes in either one or both of these can determine periods of enhanced or reduced growth. Therefore these can only be interpreted in combination with evidence from other deposits giving less ambiguous palaeoclimatic information; the latter are however often poorly dated. The correlations presented here show good agreement with the oxygen isotope record, variations from which are explained as being palaeoclimatically significant. During substage 5a, significantly low levels of secondary carbonate growth are shown to be associated with a deterioration in climate preceding the stage 4 maximum, whilst in stage 3 the growth frequency record provides a good framework into which "interstadial" sites can be fitted. In the future, the growth frequency record can be refined both in terms of improved precision in the chronology, through recent advances in thermal ionisation mass spectrometric uranium series dating (Edwards et al., 1987; Li et al., 1989), and in the interpretation of the palaeoclimatic signal, through a better understanding of the mechanisms of growth (Buhmann and Dreybrodt, 1985). This is the focus of current research.

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