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Variations in stalagmite luminescence laminae structure at Poole's Cavern, England, AD 1910–1996: calibration of a palaeoprecipitation proxy

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Abstract: Duplicate records of variations in the structure of stalagmite annual luminescence laminae are investigated for the period AD 1910 to 1996 for Poole's Cavern, Buxton, central England. For the two stalagmites, 88% of the years have luminescence laminae that exhibit a near sinusoidal shape with no structural variations. However 10 laminae (12% of total) exhibit a double band structure; these are demonstrated to occur in years with high monthly or daily mean precipitation. It is suggested that high intensity (>60 mm d^{-1}) and high quantity (>250 mm per month) of precipitation may flush luminescent organic material onto the stalagmites from either the soil or groundwater zones and generate a double lamina. However, not all precipitation events generated double laminae. High-intensity events in summer were ineffective due to a soil moisture deficit and/or interception by the woodland canopy. High-rainfall months (>250 mm) failed to generate double laminae when preceded by two or more months of greater than 150 mm, suggesting exhaustion of the organic acid supply can occur. When compared to monthly precipitation data for Buxton, laminae shape and the percentage of double laminae of the Poole's Cavern stalagmites are best explained by a centre-weighted running mean of the preceding six to seven months' precipitation. The palaeoclimate potential of structural variations in stalagmite luminescence laminae is discussed.

Key words: Speleothems, stalagmite, luminescence, laminae, precipitation, rainfall intensity, palaeoclimate.

Introduction

One recent focus of research in the field of speleothem palaeoclimatology has been the investigation of speleothem luminescence properties. Speleothem luminescence predominantly derives from organic acids that are trapped within the speleothem calcite and which derive from the overlying soil (Baker *et al.*, 1993; Shopov *et al.*, 1994). Research into speleothem luminescence has focused on:

(1) long-term (10^2–10^4 year) variations in speleothem luminescence intensity, that may be related to both long-term climate and environmental changes (Shopov *et al.*, 1994; Baker *et al.*, 1996);
(2) annual luminescence laminae, that in temperate latitudes are deposited in autumn and winter, which can provide both an annual chronology, as well as a potential high-resolution palaeoclimate or palaeoenvironmental record (Baker *et al.*, 1993; Shopov *et al.*, 1994; Genty *et al.*, 1997; Tan *et al.*, 1997).

Annual luminescent laminae have to date, to the authors' knowledge, only been used as a palaeoenvironmental proxy through their determination of annual growth rate variations. For example, Tan *et al.* (1997) demonstrated that, over the historical time period, the growth rate of one Chinese stalagmite correlated with monsoon strength. This correlation was used to extrapolate the monsoon record back for 1000 years. However, a recent study that undertook high-resolution (7–20 day) sampling of groundwaters over the period 1996–1998 at one speleothem-depositing site demonstrated significant variations in groundwater winter luminescence intensity that varied with the timing of periods of hydrologically effective precipitation (Baker *et al.*, 1999). In particular, for the temperate maritime region investigated, luminescence intensity increased:

(1) during the winter when soil moisture deficit was exceeded for several months and the groundwater could be recharged;
(2) potentially at other times of the year after periods of extreme precipitation intensity or totals, which could flush dissolved organic material from the soil to the groundwater via, for

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example, soil macropores or fissure overflow routes in the karst aquifer.

These results suggested that, for some years, subannual variations of dissolved organic matter might occur; if these are subsequently trapped within speleothem calcite then they may be able to provide palaeoclimate information. Qin *et al.* (1998) report seven types of structural variations in speleothem laminations observed in visible and UV illumination of a Chinese stalagmite from a monsoon climate region, and ascribe several structures to surface precipitation variations. They conclude that annual laminations can preserve seasonal climate information; however, no calibration against historical climate proxies was undertaken. Therefore, in order to assess the potential of this palaeoclimate proxy, further historical calibration is needed from stalagmites for which a long surface climate record is available. This is the focus of this paper.

Site description

Poole's Cavern, Buxton, Derbyshire, England (OS Grid Reference SK/049725; altitude 340 m) was chosen to undertake a calibration of luminescence lamination structure variations. The cavern, formed in Carboniferous limestone, has undergone detailed investigation of both its dripwater and stream hydrology over 25 years (Pitty, 1966; Baker *et al*., 1998; Gunn, unpublished data). The hydrology of the site is known to be typical of karst groundwater. In addition, the site is part of a nature reserve and designated Site of Special Scientific Interest; the cave is overlain by sycamore, beech and elm secondary woodland and has not been significantly affected by anthropogenic influences over the last 100 years. Furthermore, prior to the development of secondary woodland, the site was utilized for lime-kilning. This has resulted in significant lime deposits in the soil, and many of the stalagmites therefore have accelerated growth rate (1–20 mm yr^{-1}), with deposition from hyperalkaline waters ($pH > 10.0$) by the reaction $CaO+CO_2 \rightarrow CaCO_3$ (Pinsent *et al*., 1956; Barnes *et al.*, 1982; Baker *et al*., 1998). Although the presence of these stalagmites is atypical and unlikely to be commonly observed elsewhere, their high growth rate is beneficial in the reconstruction of high-resolution palaeoclimate proxies as it improves the achievable temporal resolution. Although the rate kinetics of the $CaO+CO_2 \rightarrow CaCO_3$ reaction are poorly understood, and thus growth-rate variations of stalagmites at the site cannot be interpreted, variations in luminescence lamination structure are independent of the deposition mechanism and can therefore be investigated as typical of karst groundwater. Therefore structural variations in luminescence at the site can be calibrated in order to investigate its utility as a palaeoclimate proxy, dependent on the surface climate, soil and vegetation characteristics, and the groundwater hydrology of the site.

Climate data for the site was obtained from British Meteorological Office returns from Buxton (within 2 km of the caverns). Daily precipitation totals have been collected at Buxton since 1906 and returns in the form of monthly means and daily extreme values were compiled from the journal *British Rainfall*. No returns were available for April–October 1978, and for this period data from nearby stations in Derbyshire were used. Temperature and snowcover data were collected from returns collated in the Monthly Weather Report of the British Meteorological Office.

Analytical methods

Three stalagmite samples were collected from Poole's Cavern. One 4 cm tall sample, PC-97-2, was collected from the Roman Chamber, 50 m from the cave entrance and a site of archaeological excavations between AD 1981 and 1984. The stalagmite sample was depositing on matting left in the cavern by the archaeologists, and must therefore postdate AD 1984. The stalagmite was primarily utilized to confirm the annual nature of the luminescence laminations at the site, although it was also used in the calibration with climate.

Two larger stalagmites were also sampled. PC-97-1, sampled in 1997 from behind 'The Cat' in Poached Egg Chamber, 130 m from the cave entrance, was a 30 cm tall minimum diameter stalagmite. Dripwater samples and drip rates were also obtained at monthly intervals at this site to confirm a winter luminescence maximum. In 1998, a second sample (PC-98-1; 36 cm tall) was also sampled after being accidentally broken by cavern staff. Due to the opportunistic sampling of this stalagmite, no water chemistry or drip rate data were obtained.

All stalagmites were sectioned down the long axis of growth and polished on one half (see Figure 1). Chronologies were constructed from the annual laminae that were visible in both UV and visible light, starting at 1996, as the top lamina (1996–97 growth) was badly fractured during the sectioning of the stalagmites. Luminescence intensity variations were measured using a Perkin-Elmer LS-50B luminescence spectrophotometer with fibre-optic extension and moving stage. The excitation wavelength was set to 325 nm, and the emission wavelength to 410 nm, this being the excitation-emission pair that excited the maximum luminescence intensity for the samples. Spectrophotometer slit widths were set to 10 nm to ensure a high signal:noise ratio. Fibre-optic spot size was set to 1 mm width, and the stalagmites scanned at 0.1 s intervals with a translation rate of 3 mm/min. This scan combination gave a 5 μm sampling interval with 200-point data overlap. Samples were scanned in short, overlapping sections, in

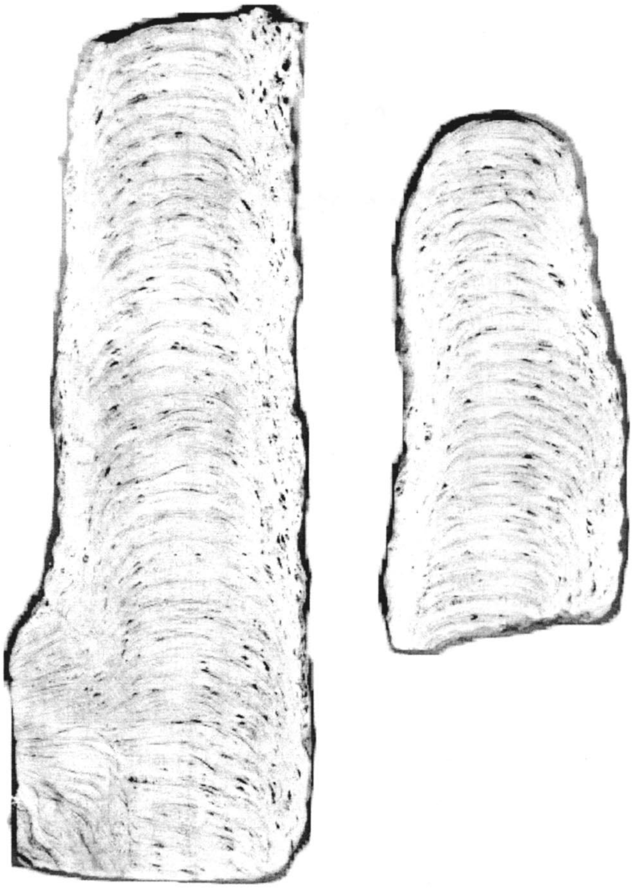

Figure 1 Stalagmite PC-97-1 under visible light illumination. Sample height is 30 cm; visible laminae correspond to laminae visible under UV excitation with the dark visible laminae corresponding to high luminescence laminae.

an attempt to avoid sections of porous calcite, which were located by visual observation and with a hand-lens. Highly porous calcite interferes with the luminescence intensity signal by generating spurious troughs in luminescence intensity caused by holes and pits in the calcite surface that cause the luminescent light to be scattered at the surface and have a lower detection rate by the fibre-optic. All scans were performed in triplicate and a mean and standard deviation luminescence intensity calculated.

Water samples of the dripwater for sample PC-97-1 were collected at the end of each month in 10 ml glass bottles and analysed on the luminescence spectrophotometer within four days of sampling. Excitation wavelengths were scanned from 280 to 340 nm wavelengths at 2 nm steps; for each excitation wavelength, luminescence emission was determined from 370 to 450 nm at 0.5 nm steps. The excitation:emission wavelength pair of the maximum luminescence was recorded, together with the luminescence intensity. Blanks were run as detailed in Baker *et al*. (1999). The drip rate at PC-97-1 was also recorded to the nearest second.

Results and discussion

Confirmation of annual lamination and construction of stalagmite chronologies

Lamination counting in both visible and UV illumination revealed 14 laminations in PC-97-2. Given the sampling date of AD 1997 and the expected start of stalagmite deposition between AD 1981 and 1984, the lamina count agreed well with that expected for annual laminations. Given the similarity in lamina appearance for all three sectioned samples, it was considered a reasonable assumption that all laminations were annual. By counting laminations from the top of the sample to the base, the length of deposition of each sample was determined to be AD 1927–1997 (PC-97-1), AD 1983–1997 (PC-97-2) and AD 1905–1997 (PC-98-1). The growth rate for the three samples averaged 5.0 mm yr^{-1} (PC-97-1), 2.1 mm yr^{-1} (PC-97-2) and 3.8 mm yr^{-1} (PC-98-1). For the latter sample, growth rate decreased around a threshold year of 1967; prior to this date the growth rate averaged 4.8 mm yr^{-1} and after AD 1969 averaged 1.7 mm yr^{-1}. The cause of this growth rate change is not known, but may reflect a flushing of CaO from the soil during consecutive years of above average (131% and 126% above the 30 year mean) precipitation in AD 1965 and 1966.

Luminescence intensity variations for the dripwater feeding PC-97-1 are presented in Figure 2. The data confirms a winter luminescence intensity maximum, although the monthly sampling interval is inadequate to resolve any structure in the luminescence signal. Drip rate for the site varied between 20 and 25 s per drip and exhibited no seasonal variations in drip rate. This suggests that groundwater at this site has a significant component of long residence time, storage flow. As seasonal luminescence variations are observed, however, a shorter storage time component must also be present.

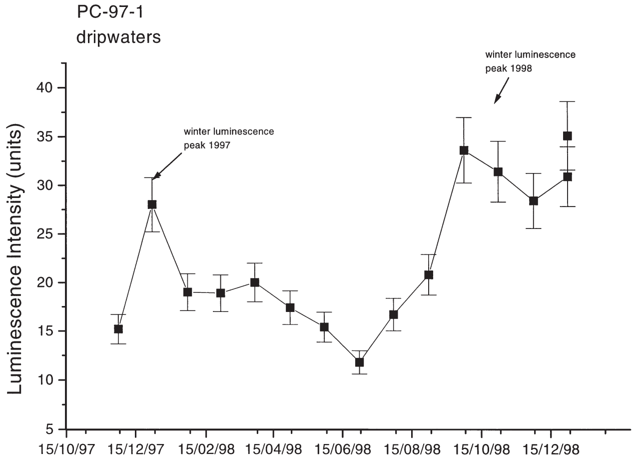

Figure 2 Luminescence intensity data for the dripwater feeding PC-97-1 for the period 1997 to 1999.

Structure of luminescence laminae

The principal structure observed in the stalagmites was that of a double lamination, defined as the occurrence of two distinct maxima within the top 25% of any luminescence peak, rather than the presence of an inflection point within a luminescence maximum.

PC-97-2

Only the top ten laminations (AD 1986–1996) were scanned for structural variations; prior to 1986 the growth layers were not parallel, caused by settling of the matting upon which the stalagmite was forming. For the period 1986–1996, no variations in the structure of the luminescence intensity peaks was observed (Figure 3).

PC-97-1

The complete luminescence intensity record for this sample is presented in Figure 4. For the majority of years, the luminescence laminae exhibit no variations in structure and appear sinusoidal

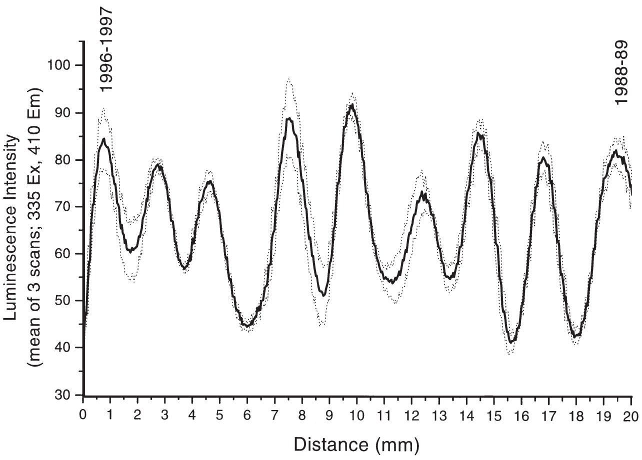

Figure 3 Mean (solid line) and standard deviation (dotted) line variations in lamina luminescence for PC-97-2. Note the lack of structure in the signal and the almost perfect sinusoidal shape.

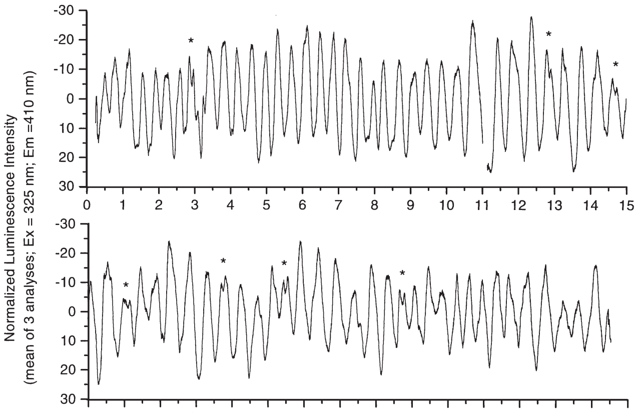

Figure 4 PC-97-1 luminescence laminae. Only the mean of three analyses is shown for clarity. Years with a double lamina structure are asterisked. Note also the low intensity of luminescence in the drought year of 1975 and high intensity in the wet year of 1965.

Table 1 Years of observed double laminations (✓ = yes; × = no)

Stalagmite sample	PC-97-1	PC-97-2	PC-98-1	Precipitation extreme
Deposition/data period	1927–1997	1983–1997	1905–1967 excluding 1959–62, 1940–42, 1933	1908–1994 excluding April–Oct 78
1988–1989	✓	×	No data	none
1964–1965	✓	No data	×	12/12 71 mm
1960–1961	✓	No data	No data	3/12 65 mm
1957–1958	✓	No data	✓	Sept 256 mm
1956–1957	×	No data	✓	Aug 271 mm
1951–1952	✓	No data	✓	Nov 289 mm
1947–1948	✓	No data	✓	Jan 308 mm
1940–1941	✓	No data	No data	Nov 250 mm
1940–1941	✓	No data	No data	22/6 64 mm
1927–1928	No data	No data	✓	Jan 273 mm
1914–1915	No data	No data	✓	Dec 207 min*

*Text entry in *British Rainfall* states a 50-year maximum monthly value in this month, with 250% of normal precipitation. This would suggest a misreturned figure with a true value of >350 mm.

in shape and identical in appearance to PC-97-2. However, for seven years a double structure is visible (Figure 4); these years are listed in Table 1.

PC-98-1

A complete luminescence intensity record was not obtained from this sample, due to three regions of high porosity that were present across growth layers and not avoidable during data collection. Thus no luminescence structure information was obtained from AD 1933–1934, 1940–1943 and 1959–1962. In addition, no data was obtained for the period 1969–1997, as the growth rate was too low to adequately resolve subtle variations in luminescence intensity. For the remainder of the sample, six years exhibited a double luminescence lamina structure, listed in Table 1.

Comparison with climate data

The abundance of sinusoidal luminescence peaks in all stalagmite samples at Poole's Cavern suggests that the effect of surface precipitation variations on groundwater luminescence fluxes is considerably smoothed. For PC-97-1, where the near constant drip rate of the water feeding this sample suggests the presence of a significant groundwater storage component, the smoothed luminescence signal may be explained by the dominance of this component. In order to assess the precipitation-luminescence relationship, the Buxton mean monthly precipitation record was analysed using 3–9 month unweighted and centre-weighted running means and compared to the number of lamination structures observed in PC-97-1, PC-97-2 and PC-98-1 (Table 3). The unweighted smoothing functions all produced time-series with structures visible in most years and a gradual loss of the annual signal as the running mean increased. Weighting the running mean precipitation produced a better fit with the observed speleothem lamination shape, with the best agreement observed with a six- or seven-month centre-weighted running mean of the preceding precipitation. The necessity to apply a weighting function to the running mean data suggests that there is a groundwater residence time of several months, with some mixing of components of different residence times, feeding the stalagmites at the site.

The only lamina structure visible in the stalagmites at Poole's Cavern is the occasional presence of double laminae. Several climate parameters were considered that might influence fluxes of luminescent organic matter from the soil zone, and thus generate double laminae. Previous studies have suggested that periods of intense or high quantities of precipitation may flush dissolved organic matter into the groundwater (Baker *et al*., 1999). Hence Buxton monthly mean precipitation data was analysed, with the

Table 2 Extreme climate events not recorded in the Poole's Cavern stalagmites

Year	Climate event	Comments
1986	261 mm in December	preceded by 2 months >150 mm
1987	25/8 67 mm in the day	
1988	264 mm in February	preceded by 2 months >150 mm
1975	1/12 61 mm in the day	
1973	15/7 87 mm in the day	
1965	324 mm in December	preceded by 2 months >150 mm; features as an intense peak in both samples
1962	59 days snowcover and mean monthly T <0°C in January and February	coldest winter during deposition period; no effect on stalagmite laminae
1946	7/2 66 mm in the day	
1947	258 mm in September	preceded by 2 months >150 mm

Table 3 Comparison of smoothing function and number of years with structural variations present in the monthly precipitation record for Buxton

Smoothing function on preceding monthly mean precipitation	Years with double/complex winter luminescence intensity peak (1910–1996)	
	Number	%
3-month running mean (unweighted)	38	44
5-month running mean (unweighted)	21	24
7-month running mean (unweighted)	22	26
9-month running mean (unweighted)	no laminae	
5-month running mean (centre-weighted)	20	23
6-month running mean (centre-weighted)	10	12
7-month running mean (centre-weighted)	5	6
8-month running mean (centre-weighted)	0	0

upper 5, 1 and 0.1% precipitation months (Figure 5) compared to years with double laminae. The upper 5% of all monthly precipitation events represent monthly mean precipitation greater than 200 mm; 49 such occurrences occurred during the study period, significantly more than the number of double laminae structures present in the speleothems. The upper 1% of all events represents precipitation greater than 250 mm per month (11 occurrences) and the upper 0.1% represents precipitation greater than 350 mm per event (one occurrence). A comparison between the years where double laminae are present, and climate in those years is presented in Table 1. For 7 of the 10 laminae present in PC-97-1 and PC-98-1, double laminae occur in years where mean monthly precipitation totalled greater than 250 mm (upper 1% of all monthly precipitation over the period), for example January 1948 (308 mm), November 1951 (289 mm) and September 1957 (256 mm).

Not all months where precipitation exceeded 250 mm generated double laminae, with four such events leaving no imprint on the stalagmites. Further analysis of the Buxton precipitation record demonstrated that double laminae were only recorded in years where there had been less than two months of greater than 150 mm of precipitation preceding the month of extreme (>250 mm) precipitation. Where no double laminae were recorded in years with an extreme rainfall month, it was preceded by two or more months of monthly mean precipitation greater than 150 mm. Comparison of double lamina years and total precipitation preceding the month of extreme (>250 mm) precipitation, or the timing of the extreme precipitation months within the hydrological year, did not provide a better correlation. Therefore it seems likely that exhaustion of the supply of luminescent dissolved organic acids occurs within individual hydrological years, limiting the preservation of double laminae structures within the stalagmites.

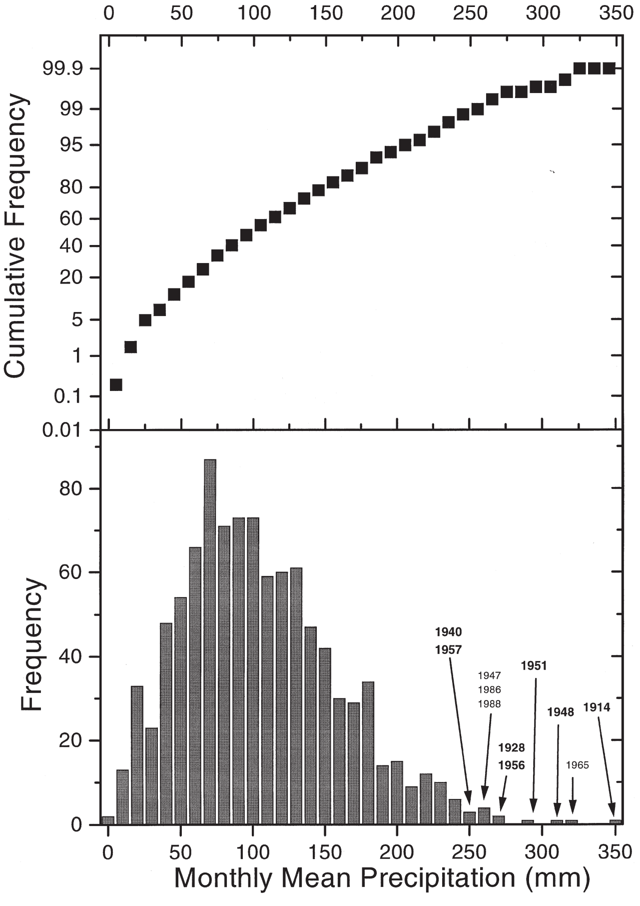

Figure 5 Monthly precipitation statistics for Buxton reporting station. Top: cumulative frequency of monthly precipitation (mm). Bottom: frequency of monthly mean precipitation. For the high-precipitation months, the corresponding years are labelled: those where stalagmite double laminae are present are in large, bold font and those where laminae are absent in small, standard font.

In addition to monthly mean precipitation, daily precipitation totals were also investigated, as these may reflect short-duration, high-precipitation intensity events that may flush organic material into the groundwater, or activate overflow routes within the karst aquifer. Due to the absence of hourly observations at Buxton, daily precipitation totals were used. Three of the 10 double laminae present in PC-97-1 and PC-98-1 occurred in years with >60 mm daily precipitation totals (one winter, 1940–41, also experienced a month of precipitation greater than 250 mm). Conversely, >60 mm of daily precipitation occurred on four other occasions and were not recorded in the stalagmites. Two such events occurred in midsummer in periods of high soil moisture deficit and so may not have recharged the groundwater. Also, daily precipitation totals provide a relatively crude measure of rainfall intensity. Comparison with hourly rainfall data may demonstrate that this variable is a better correlant with double laminae years; unfortunately such data is not available for the Poole's Cavern site.

Years where double laminae are present in the Poole's Cavern stalagmites thus correlate with months of extreme precipitation totals or intensity. This result is similar to that observed for groundwater luminescence fluxes over an 18-month period at a site in SW England (Baker *et al*., 1999). It is suggested that these events flush luminescent organic material onto the stalagmites, possibly through the activation of overflow routes in the aquifer, or the increased transport capacity at high flow to transport otherwise immobile high molecular weight organic acids. For one lamina, no obvious climatic explanation can be attributed to the lamina structure (1988–89, only visible in one sample); unless the stalagmite had a section of porous calcite at this location that was not observed, no explanation can be given for the double structure. In addition, for two years, only one of stalagmites PC-97-1 and PC-98-1 responded to the extreme precipitation event. This could be explained by the differential response of the two stalagmites to climate variations, with PC-98-1 not responding to high intensity precipitation events, and PC-97-1 less sensitive to months of high mean precipitation. For the short period of its deposition, PC-97-2 exhibited no variations in laminae structure; no extreme precipitation events occurred during its deposition period.

Conclusions

For three stalagmites that deposited within the period 1910–1996, 10 years (~12% of total) exhibited a double annual lamination structure. 90% of these laminae occurred in years of either high monthly or daily precipitation totals. 63% of all monthly precipitation >250 mm events are preserved in the stalagmites, missing years can be explained by the exhaustion of the supply of luminescent organic matter in years where preceding high rainfall months have occurred. Only 33% of daily precipitation >60 mm events correlated with double laminae in the stalagmites, and stalagmites seem less sensitive to this climate parameter. Analysis of hourly rainfall data in future studies may demonstrate this to be a better correlant. Finally, individual stalagmites from the cave demonstrated variations in their response to precipitation variations, PC-98-1 appearing more responsive to daily rather than monthly rainfall extremes. Such variability between samples is to be expected given the heterogeneity of karst groundwater flows.

The results from Poole's Cavern suggest that there is considerable potential in the analysis of stalagmite lamina structure as a palaeoprecipitation signal, especially one of precipitation intensity, and that stalagmites can successfully preserve an intra-annual climate signal. However, the use of individual samples must be treated with caution, as the two stalagmites investigated here had different responses to surface precipitation, and future investigations should include a calibration against historical or other proxy climate records. In addition, a limitation of the scanning method used in this study is the coarse resolution obtained (approximately 1 mm), which means that records of annual luminescent bands can only be obtained from stalagmites with a growth rate exceeding 1 mm/yr. Thus the method is not applicable for most stalagmites, which typically show growth rates of less than 100 microns/yr. However, by using UV microscope technology it is possible to discern annual bands and their structure down to micron scale. By using such methods one can examine a much wider sample of stalagmites to obtain long records of annual bands extending back thousands of years from the present, offering the potential for long-term palaeoclimate studies. Stalagmites offer a number of advantages over existing palaeoclimate proxies such as ice cores (narrow distribution) and tree-rings (strongly seasonal signal). In particular they offer the potential to provide long-term records of annual precipitation, in contrast to tree-ring data, which is dominated by a seasonal temperature signal. This study also demonstrates that it is possible to obtain an intra-annual climate record of extreme precipitation events from luminescent bands in stalagmites. By providing a record of precipitation intensity, it may be possible to obtain a long record of the frequency of cyclonic storms, and hence the strength of westerly circulation over the region. In addition, changes in the structure of annual luminescent bands may provide evidence of changes in seasonality of precipitation over long periods. This suggests that the use of multiple stalagmites from sites of more typical stalagmite growth rate, and averaged records of lamina structure (e.g., decadal averages) over longer time periods, may provide evidence of variations in the strength of the westerly circulation over the region. Work currently in progress in NW Scotland is directed to constructing such a record.

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