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### Intra- and inter-annual growth rate of modern stalagmites

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#### Abstract

We measure the factors that determine growth rate (temperature, drip rate, calcium ion concentration) for 31 waters that feed stalagmites within six cave systems throughout Europe. Water samples were collected at a frequency of at least 1 month, to permit the modelling of both inter- and intra-annual growth rate variations, utilising the theory of Wolfgang Dreybrodt (Chem. Geol. 29 (1980) 89-105; Chem. Geol. 32 (1981) 237-245; Dreybrodt, W., 1988. Processes in Karst Systems. Springer-Verlag, Berlin 288 pp.). Inter-annual growth rates were measured using the stalagmites that were associated with the analysed water samples; growth rate was determined from annual lamina counting, specific time markers within the stalagmites, and location of bomb <sup>14</sup>C. When compared to theoretically predicted values, a good agreement between theoretical and measured stalagmite growth rates is observed ( $R^2 = 0.69$ ). When compared to site climate and geochemical parameters, a good correlation is observed between measured growth rate and mean annual temperature for five sites ( $R^2 = 0.63$ ) and dripwater calcium content ( $R^2 = 0.61$ ), but not drip rate ( $R^2 = 0.09$ ). The good correlation with both calcium and temperature is due to soil CO<sub>2</sub> production being primarily determined by surface temperature and soil moisture. However, when we compare our data to that in the Grotte de Clamouse, a site that has little soil cover, we observe that the growth rate-temperature relationship breaks down due to either the lack of soil CO<sub>2</sub> production or prior calcite precipitation. Intra-annual data demonstrates that maximum growth rate occurs when calcium concentrations are high, and that this occurs under different seasons depending on the hydrology of each site. Our results demonstrate a stronger dependence of intra-annual stalagmite growth rate on dissolved calcium ion concentrations than drip rate for the range of drip rates investigated here  $(0.01 < t < 2 \text{ drip s}^{-1})$ , but for lower drip rates, this factor becomes important in controlling growth rate. We suggest that for well-monitored and -understood sites, stalagmite growth rate variations can provide useful information for palaeoclimate reconstruction. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Speleothem; Stalagmite; Growth rate; Palaeoclimate

#### 1. Introduction

Stalagmites are increasingly being investigated as a potential archive of palaeoenvironmental data, due to the large number of potential climatic, environmental and hydrological proxies that they contain (pollen, isotopes, timing of deposition, etc.; Bastin, 1978; Bar-Matthews et al., 1997; Baker et al., 1995, 1998; Banner et al., 1996; Genty et al., 1997, 1998). It has also become obvious that each individual stalagmite within a cave system may respond differ-

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Mean geochemical and environmental data of the drip water stations used for the calculation of the modelled growth rate (for details, see Dreybrodt, 1980, 1981, 1988; Baker et al., 1998)

Caves and dripping stations	Ca <sup>2+</sup> (mg/l)	Error $(1\sigma)$	Т (°С)	Error	Flow rate (drip s <sup>-1</sup> )	Error $(1\sigma)$	GR1 (mm/year)	Error	GR2 (mm/year)	Error	Porosity	Corr. GR (mm/year)	Error	Measured GR (mm year)	Error	Method
La Faurie																
Fau14	201.0	30.0	12.9	0.1	0.0159	0.0222	0.88	0.15	0.83	0.15	0.1	0.91	0.15	0.96	0.05	laminae counting $(n = 53)$
Fau13	208.0	28.0	12.9	0.1	0.0917	0.0827	0.92	0.14	0.91	0.14	0.1	1.00	0.14	1	0.1	laminae counting $(n = 7)$
Fau11	152.0	33.0	12.9	0.1	0.0184	0.0160	0.64	0.16	0.61	0.16	0.1	0.67	0.16	1.04	0.03	clay layer mark
Fau10	155.0	38.0	12.9	0.1	0.0098	0.0080	0.65	0.19	0.6	0.19	0.1	0.66	0.19	0.6	0.2	laminae counting $(n = 5)$
Fau8	156	29.0	12.9	0.1	0.0452	0.0250	0.66	0.14	0.64	0.14	0.1	0.70	0.14	0.4	0.2	laminae counting $(n = 8)$
Fau6	153.0	31.0	12.9	0.1	0.0128	0.0060	0.64	0.15	0.6	0.15	0.1	0.66	0.15	0.7	0.2	laminae counting $(n = 10)$
Villars																
Vil1A	138.0	18.0	11.2	0.1	0.0455	0.0225	0.49	0.08	0.48	0.08	0.0	0.48	0.08	0.5	0.1	sample height
Vil1B	176	27.0	11.2	0.1	0.2439	0.1990	0.66	0.12	0.65	0.12	0.0	0.65	0.12	0.8	0.1	sample height
Vil4	144	28.0	11.3	0.1	0.5917	1.3370	0.52	0.12	0.52	0.12	0.0	0.52	0.12			
Vil10A	124	35.0	12.4	0.1	0.2179	0.3131	0.47	0.17	0.47	0.17	0.0	0.47	0.17	0.3	0.1	laminae counting
Vil10B	106	20.0	12.4	0.1	0.0909	0.0761	0.39	0.09	0.39	0.09	0.0	0.39	0.09			
Proumeyssac																
Pro-1	153	31	13.6	0.1			0.68	0.16		0.16			0.16	1.5	0.1	deposition on a plate
Pro-2	146	24	13.6	0.1			0.64	0.13		0.13			0.13			-
Pro-3	105	25	13.6	0.1	0.0268	0.0040	0.43	0.13	0.42	0.13	0.0	0.42	0.13			
Postojna																
PR2	69.3	3.4	8.2	0.1	0.1500	0.0120	0.14	0.01	0.14	0.01	0.0	0.14	0.01			
PR3	71.5	2.6	8.3	0.1	0.1600	0.0115	0.16	0.01	0.16	0.01	0.0	0.16	0.01			
Ent.	79.9	10.5	9.3	0.1	0.2000	0.0526	0.2	0.04	0.2	0.04	0.0	0.20	0.04			
Bri.	55.5	10.0	10.4	0.1	0.1200	0.0387	0.13	0.04	0.13	0.04	0.0	0.13	0.04			

Brown's Folly Mi	ne															
bfm-96-p	103	8.2	10	0.4	14.0000	50.2000	0.31	0.03	0.31	0.03	0.0	0.31	0.03	0.078	0.04	laminae counting and sample height
bfm-96-3	105	11	10	0.4	1.6400	2.9800	0.31	0.04	0.31	0.04	0.0	0.31	0.04	0.071	0.01	laminae counting and sample height
bfm-96-b	101	14	10	0.4	0.2780	0.6590	0.3	0.05	0.3	0.05	0.0	0.3	0.05	0.148	0.05	laminae counting and sample height
bfm-96-f1	101	9.8	10	0.4	0.0356	0.0082	0.3	0.04	0.29	0.04	0.0	0.3	0.04	0.23	0.05	sample height
bfm-96-f4	87.2	8.8	10	0.4	0.0083	0.0010	0.24	0.03	0.23	0.03	0.0	0.24	0.03	0.23	0.05	sample height
bfm-96-f2	104.8	14	10	0.4	0.0147	0.0028	0.31	0.05	0.3	0.05	0.0	0.31	0.05	0.21	0.05	sample height
bfm-96-j	92	14	10	0.4	0.0996	0.1390	0.26	0.05	0.26	0.05	0.0	0.26	0.05	0.25	0.05	sample height
Uamh an Tartair																
SU-96-7	37	4	7.25	1	0.0011	0.0022	0.04	0.02	0.03	0.02	0.0	0.03	0.02	0.022	0.01	laminae counting
Clamouse																
Clam1A	44.4	2	14.5	0.1												
Clam1B	43.8	2	14.5	0.1												
Clam2	35.7	2	15.3	0.1												
Clam3	38.4	2	15.3	0.1												
Clam4	32.4	2	15.5	0.1												
GB Cave (Stenner, 1973)	63.6	15.6	10	0.5												
Poole's Cavern (Pitty, 1966)	97.2	6.8	9	0.5												
Clamouse (Fairchild, 1999)	47	11	14.5	0.1												
Ernesto (Fairchild, 1999)	53.1	5.2	6.5	0.1												

Our data represent mean values of monthly sampling (or less) during 1 year, except for Clamouse, which has been sampled 1 day (spring 1999). Modelled growth rates have been calculated using: (i) calcium concentration and temperature as the principal determinants (GR1); (ii) calcium concentration, temperature and flow rate (GR2). For all calculation, we have used a mean water film thickness = 0.01 cm and a  $pCO_2$  air pressure =  $3 \times 10^{-4}$  atm. Note the similarity between the two calculated growth rates principally due to the choice of fast drip stations.

ently to surface climate and environmental changes, due to, for example, local variations in soil and vegetation, different flow routes through the karst aquifer and differing responses to surface precipitation events, etc. (Smart et al., 1996; Smart and Friederich, 1987; Baker et al., 1997; 1999a; Genty and Deflandre, 1998). Hence, research by the authors in recent years has focused on high frequency and spatial monitoring of cave systems throughout Europe, with the intention of understanding individual stalagmite responses to surface environmental and climate variations, before proceeding with palaeoclimate investigations.

One such potential palaeoclimate proxy is that provided by annual growth laminae. Laminae are frequently found in speleothems in both VIS and UV light; in some instances these laminae can be demon-

strated to be annual. Annual laminations within stalagmites can provide a high-resolution record of growth rate simply by measuring the distance between laminae; the use of laminae is a more precise measure of growth rate than that used by early researchers, who were limited to the use of alphaspectrometric U–Th dating.  $^{14}$ C and the use of known time markers (Harmon and Curl, 1978; Gewelt, 1986; and references in Shaw, 1992; Ford and Williams, 1989; Genty and Quinif, 1996; Hill and Forti, 1997). Through the work of Wolfgang Dreybrodt (Drevbrodt, 1980; 1981; 1988; Baker et al., 1998). the factors affecting stalagmite growth rate are well understood: the three significant determinants being the calcium ion concentration and the temperature of the dripwater, and the water supply rate. Baker et al. (1998) demonstrate, for three cave systems in N and



Fig. 1. Comparison of measured and modelled growth rates (a) using just temperature and calcium as determinants ( $R^2 = 0.81$ ), together with best fit lines: (1) dash line, all sites; (2) thick solid line, excluding Proumeyssac; (3) thin solid line, modelled growth rate = measured growth rate (b) using temperature, calcium and drip rate as determinants ( $R^2 = 0.79$ ).



W Europe that exhibited similar climatic characteristics, that there is a good correlation between observed recent stalagmite growth rate and that theoretically predicted, with the principal determinant of growth rate being the calcium ion concentration. In this study, we expand this investigation to determine the following.

(1) The relationship between theoretical and observed growth rates for six sites that cover a wide range of climates, in order to determine whether the theoretical predictions apply for most variations in temperature, calcium and drip hydrology, which might be expected to be observed over interglacial and interstadial Quaternary climates in European caves.

(2) The intra- and inter-site relationship between growth rate and climate and environmental factors. By measuring the geochemistry and hydrology of individual drip water-stalagmite pairs at four of the six sites, we can assess the intra-site variations of stalagmite growth rate and, therefore, the utility of the site for regional palaeoclimate reconstruction. In addition, inter-site comparisons can also be undertaken in order to compare observed intra-site variability of growth rate with inter-site variations, for sites over a wide range of climate zones, using both our data presented here as well as other published drip geochemistry results from Derbyshire (England; Pitty, 1966), GB Cave (England; Stenner, 1973), Clamouse (South France) and Ernesto (North Italy) (Fairchild et al., 2000).

(3) The intra-annual variation in growth rate for stalagmites at sites where we can model intra-annual growth rate from the seasonal variation in drip hydrology and geochemistry. If the theoretical growth rate model is applicable to all sites investigated here, then with dripwater sampling at 1 month or better frequency, we can utilise theoretical growth rate to predict intra-annual growth rate variations. Such investigations will provide insight into the intra-annual structure of stalagmite laminae (for example, the ratio of porous to compact calcite in visible laminae, or the ratio of luminescent to non-luminescent laminae in laminae observed under UV light).

#### 2. Research sites and methodology

2.1. Six cave systems were utilised in this investigation

2.1.1. La Faurie, Dordogne, France (45°08'N, 1°11'E, elevation 225 m)

This cave is a fossil, horizontal linear phreatic tube that is found at a depth of 10–20 m below the surface in Bajocian limestone (Genty and Massault, 1997); overlying vegetation comprises a natural mix

of dispersed oak woodland and grasses with a thin brown rendzina soil cover. Six dripwater sites were monitored (paired samples at three locations within the cave) for their hydrology and geochemistry at monthly intervals over the period 1997–1998, and at the end of the study period, the associated stalagmites were removed and sectioned.

# 2.1.2. Villars, Dordogne, France $(45^{\circ}30'N, 0^{\circ}50'E, elevation 175 m)$

This cave comprises both active and relict vadose and phreatic passages ranging from depths of 10 to 40 m from the surface in Bajocian limestone (Genty and Massault, 1997; Baker et al., 1998). It is covered by a mixture of deciduous woodland (oak, hazel) and grassland, and has a thin brown rendzina soil cover. Hydrological and geochemical data was collected from five sampling locations at monthly intervals over the period 1997–1998; and at the end of the study period stalagmites at three of the sites were removed and sectioned.



Fig. 2. Comparison of measured growth rate with mean annual temperature at each site (GR measured = 0.193T - 1.67;  $R^2 = 0.63$ ).

# 2.1.3. Proumeyssac, Dordogne, France (44°55'N, 0°56'E, elevation 155 m)

This cave primarily comprises a large chamber with a height 50 m; the overlying geology comprises Maastrichtian limestone (Genty and Massault, 1997) and the overlying vegetation is grass and agricultural with an improved brown rendzina soil. Three water samples whose sources were drip or flows from the roof of the chamber and that were forming large stalagmites bosses were collected at monthly periods over the period 1997–1998, and actual growth rates were observed at one of the sites by analysing the calcite deposited on a glass plate that was placed on top of a stalagmite boss over the period of 1 year.

# 2.1.4. Brown's Folly Mine, Wiltshire, England (51°23'N, 2°22'W, elevation 180 m)

This site is an abandoned mine cut horizontally through ooltic Jurassic limestone at a depth of 10–15

m; overlying vegetation is secondary deciduous woodland (Baker et al., 1998) with a thin brown rendzina soil cover. Sixteen sites were monitored for hydrology and geochemical variations at 10- to 20day frequency between November 1996 and April 1998 (Baker et al., 1999a,b,c), stalagmites from seven of these were subsequently sampled and sectioned.

# 2.1.5. Uamh an Tartair, Assynt, Scotland (58°15'N, 4°57'W, 220 m)

This cave is an active vadose system found at 5–15 m depth below the surface in Cambro–Ordovician dolomite. The cave is overlain by a thin peat deposit with typical blanket bog vegetation cover; further details can be found in Baker et al. (1993, 1999a,b,c). Water samples were collected at six sites from 1997 to 1998 at monthly periods, except where sampling was prohibited due to flooding of the en-



Fig. 3. Comparison of dripwater Ca<sup>2+</sup> concentration with mean annual temperature for our study sites, plus previously published literature (Pitty, 1966; Fairchild, 1999). Note that Clamouse data set breaks down the correlation observed on the other sites (calcium concentration =  $17.66 \times T - 78.02$ ;  $R^2 = 0.63$ ).

trance passages in winter. Subsequent to the water sampling, one stalagmite was collected which was in the middle of the sampling area although not actually sampled for drip water.

### 2.1.6. Postojna, Slovenia (45°46'N, 12°12'E, elevation 529 m)

This cave was developed in upper Cretaceous limestone at a depth of between 10 and 120 m from the surface. It is covered by a pine tree forest and grass fields and thick (25-30 cm) brown earth and rendzina soils. Four sites were monitored for hydrol-

ogy and hydrochemistry over the period 1996–1997, and one stalagmite was sampled that was unassociated with the drip sites.

At each site, water samples were collected in glass bottles; on return to the laboratory the water samples were filtered and acidified with concentrated  $HNO_3$ . Calcium ion concentrations were determined by atomic absorption spectrophotometry at the University of Exeter (BFM, Villars), University of Newcastle (UAT), at the Moulis Laboratory (La Faurie, Proumeyssac) and at the Joseph Stefan Institute, Postojna (Postojna Cave). Interlaboratory compar-

Table 2

Comparison of modelled monthly growth rates using calcium and temperature as determinants (GR1) and using calcium temperature and drip rate (GR2)

Stalagmite	$R^2$	Gradient, intercept of linear fit	Months where $GR2 \ll GR1$				
La Faurie							
Fau14	0.789	0.758, +0.118	July–December 1997				
Fau13	0.989	0.974, -0.007	-				
Fau13	0.989	0.974, -0.007	-				
Fau11	0.927	1.077, -1.041	May 1997, October 1997				
Fau10	0.947	1.017, -0.035	May 1997				
Fau8	0.994	1.028, -0.035	_				
Fau6	0.988	0.973, -0.027	-				
Villars							
Vil1A	0.999	0.995, -0.007	_				
Vil1B	0.999	1.007, -0.009	-				
Vil4	1.000	1.000, 0.003	-				
Vil10A	0.997	0.985, -0.002	-				
Vil10B	0.950	0.933, +0.017	November 1997				
Proumeyssac							
Prou-3	1.000	0.968, -0.001	-				
Postojna							
PR2	1.000	1.003, -0.002	_				
PR3	1.000	1.003, -0.002	_				
Ent	0.998	1.039, -0.010	_				
Bri	0.999	1.036, -0.006	-				
Brown's Folly Mine							
BFM-96-P	1.000	1.003, -0.003	_				
BFM-96-3	1.000	1.022, -0.007	_				
BFM-96-B	0.981	1.111, -0.040	July 1997				
BFM-96-f1	0.998	0.9643, +0.005	_				
BFM-96-f4	0.992	0.917, -0.009	_				
BFM-96-f2	0.998	0.957, -0.001	_				
BFM-96-J	0.967	0.931, +0.011	May 1997				

isons were performed on blind samples between Newcastle, Exeter, Moulis and Joseph Stephan Institute. Reproducibility was determined to be +7%. Hydrology of the sample sites was determined by either the timing between individual drips or, for fast drip rates, the time taken to fill a bottle of known volume. At the Grotte de Proumevssac, where the two of the drip sources were diffuse and 10 m above the stalagmite bosses, a discharge measurement was not possible, although we observe discharge to be continuous throughout the hydrological year with some visible seasonal variations. The temperature at each site was measured in the cave atmosphere and occasionally in the dripwater. Cave air CO<sub>2</sub> was also measured at the sites and demonstrated to be in the range of 340-3000 ppm (Baker and Genty, 1998 and unpublished data). This range of concentrations do not significantly affect stalagmite growth rate (explains  $\sim 10\%$  of growth rate variability at a dripwater calcium ion concentration of 2 mmol  $1^{-1}$ ) and has decreasing significance with increasing dripwater calcium ion concentration (Dreybrodt, 1988; Baker et al., 1998); no correction for  $pCO_2$  was applied in this study.

Stalagmite samples were collected, sectioned and polished at each site, with the exception of Proumeyssac, where a glass plate was placed on the surface of the stalagmite boss and removed after 1 year and then sectioned. Growth rate determinations were undertaken in a variety of ways. (1) Through the measurement of the width of visible laminae (Villars, La Faurie, Proumeyssac) (2) Through the measurement of the width of luminescent laminae (Uamh an Tartair, Brown's Folly Mine) (3) through the measurement of total growth since a known time marker (clav layers placed on the stalagmite by the authors (La Faurie), the time of first possible stalagmite growth (Brown's Folly Mine). the location of a carbon laver after a bomb explosion in 1944 (Postiona)). In several cases, we have been able to confirm the annual nature of the laminae through either the comparison of two of the above methods at any one site (La Faurie, Brown's Folly Mine, Villars: Baker et al., 1998), as well as using the location of bomb derived <sup>14</sup>C as a control (Villars, LaFaurie Cave, Postiona: Genty et al., 1998: Genty et al., 1998), or TIMS U-Th (Uamh an Tartair; unpublished data).



Fig. 4. Mean annual drip rate vs. correlation coefficient (*R*) between GR1 (modelled growth rate with calcium and temperature) and GR2 (modelled growth rate with calcium, temperature and drip rate). Note that when mean annual drip rate decreases under 0.1 drip s<sup>-1</sup> and is very variable, the correlation becomes weaker.

### 3. Results and interpretation

# 3.1. Comparison between theoretical and observed stalagmite growth rates

Geochemical and hydrological data, as well as theoretical and observed growth rates are presented in Table 1. Mean annual temperature for all sites varied from a minimum of 7.2°C (Uamh an Tartair) to 13.6°C (Proumeyssac). Theoretical growth rates for each stalagmite (or for the sites as a whole where we do not have paired dripwater–stalagmite data) are calculated using the theory of Dreybrodt (Eqs. (1) and (3), Baker et al., 1998). We include a correction



Fig. 5. Monthly (a) drip rate and (b) calcium ion concentrations for La Faurie Cave. (c) Modelled intra-annual growth rate variations using temperature and  $Ca^{2+}$  concentrations as determinants. For Fau14, which has a slow drip rate, we present data for both GR1 and GR2; both growth rates have the same seasonal trends but GR2 is significantly lower at low drip rate.



for porosity of 10% at La Faurie cave where the stalagmites comprise porous calcite (see Baker et al., 1998 for method) and we assume that dripwater temperature is equal to mean annual air temperature as observed in most caves (Wigley and Brown, 1977). Fig. 1a presents the theoretically predicted growth rate using just calcium and temperature as growth rate determinants, in comparison to that actually observed; the best-fit line has an equation:

$$GR_{mod} = 0.477GR_{mes} + 0.248 \quad R^2 = 69\%$$
(1)

where  $GR_{mod}$ . and  $GR_{mes}$ . are modelled and measured growth rates, respectively.

One stalagmite from the Proumeyssac site is an outlier to the dataset, that of Prou-1. For this stalagmite, measured growth rate is higher than predicted; this may be due to increased deposition due to turbulent flow under high discharge conditions and higher energy upon drip impact due to the high fall height (10 m), both of which will increase growth rate (Buhmann and Dreybrodt, 1987). Also, the water feeding Prou-1 was observed to be heavily contaminated with nitrates (more than 100 mg  $1^{-1}$ ) and so growth rate may also be influenced by foreign-ion effects which should have an opposite effect on the growth rate (Buhmann and Dreybrodt, 1987). Given both of these factors, we removed Prou-1 from the dataset. The relationship between modelled and observed growth rate becomes:

$$GR_{mod} = 0.634GR_{mes} + 0.195 \quad R^2 = 81\%$$
 (2)

Eq. (2) demonstrates a very good fit between modelled and measured growth rate, although a perfect fit would result in a gradient of 1, we observe that forcing a regression line with this gradient maintains an  $R^2$  of 81%.

Fig. 1b shows the relationship between the measured growth rate and modelled growth rate where the latter also includes drip rate as a determinant. For this model, with Prou-1 sample removed from the dataset as above, the equation is almost identical to Eq. (2):

$$GR_{mod} = 0.608GR_{mes} + 0.191 \quad R^2 = 79\%$$
 (3)

This result suggests that no improvement is obtained with the inclusion of drip rate. The critical step in determining growth rate that is of relevance here is the time taken for the degassing of  $CO_2$ ; if the drip time is significantly longer than that required for complete degassing then growth rate will slow as there will be insufficient water to replace each drip. However, for our sites, the drip rate is typically more than 0.01 drip s<sup>-1</sup>, and therefore drip rate is not a significant control on growth rate (Baker, 1993; Dreybrodt, 1988); we suggest that Ca and T

are the dominant variables in determining inter-site stalagmite growth rate for the range of growth rate determining variables studied here (7.2°C < *T* < 13.6°C, 0.9 mmol  $1^{-1}$  < Ca < 5.2 mmol  $1^{-1}$ , 0.01 drips s<sup>-1</sup> < discharge < 2 drips s<sup>-1</sup>).



Fig. 6. Monthly (a) drip rate and (b) calcium ion concentrations for Brown's Folly Mine. (c) Modelled intra-annual growth rate variations using temperature and  $Ca^{2+}$  concentrations as determinants.



# 3.2. Comparison between intra- and inter-site growth rates and climatic and environmental parameters

Within four of the six cave systems, we have been able to measure the drip chemistry of waters that are directly forming the sampled stalagmite (Table 1). For these samples, we can investigate the relative importance of intra- and inter-site geochemical and hydrological variations on the stalagmite growth rate at these sites. Results demonstrate that intra-site measured growth rate variations may be considerable. For example, La Faurie Cave has growth rates that vary from 0.40 to 1.04 mm/year, with intra-site growth rates correlating with the spatial location of the paired samples (Fau6 and Fau8, Fau10 and Fau11, Fau13 and Fau14 being adjacent pairs). However, these intra-site variations are not as significant as inter-site variations: La Faurie and Villars are not statistically different, (different at the 75% confidence level), but they are from the same region (within 50 km of each other). La Faurie and Villars together have a higher growth rate than Brown's Folly Mine, which has a higher growth rate than Uamh an Tartair at a 99% confidence (ANOVA test). In order to determine the cause of this inter-site variability, we have undertaken comparisons of measured growth rate with temperature,  $Ca^{2+}$  concentration and drip rate. A strong correlation is observed between mean annual temperature (*T*) and measured growth rate (Fig. 2):

$$GR_{mes} = 0.1927T - 1.6706 \quad R^2 = 63\% \tag{4}$$

and also with  $Ca^{2+}$  (in mg  $l^{-1}$ ):

$$GR_{mes} = 0.0075Ca^{2+} - 0.4785 \quad R^2 = 61\%$$
 (5)

No correlation is observed with drip rate ( $R^2 =$  9%). The good correlation with both calcium and cave air temperature is due to soil CO<sub>2</sub> production being primarily determined by surface temperature and soil moisture (Drake, 1980, 1983); high soil CO<sub>2</sub> leading to a greater amount of dissolved Ca<sup>2+</sup> being generated. Despite possible inter-site differences in water flow path between open and closed system endmembers, which will also influence dissolved Ca<sup>2+</sup> load, it appears that stalagmite growth rate may be determined by temperature changes.

When mean site geochemical data and temperature are plotted for all six cave sites, together with other European sites where dripwater calcium and temperature have been determined (Poole's Cavern, Derbyshire; Pitty, 1966; GB Cave, Somerset; Stenner, 1973, and Grotte de Clamouse and Grotta di Ernesto; Fairchild et al., 2000 and our unpublished data) (Table 1 and Fig. 3), it is apparent that the temperature–growth rate correlation breaks down due to the inclusion of the Grotte de Clamouse ( $R^2 = 0.0\%$  with Grotte de Clamouse included in the dataset;  $R^2 = 61\%$  if it is excluded). Several reasons can explain the observed low calcium ion concentration in Clamouse seepage water, as follows.

(1) This site has a Mediterranean climate and is overlain by dolomite with only sparse vegetation



Fig. 7. Monthly (a) drip rate and (b) calcium ion concentrations for Gouffre de Proumeyssac. (c) Modelled intra-annual growth rate variations using temperature and  $Ca^{2+}$  concentrations as determinants.



cover, with soil and vegetation limited to bushes growing in thin soils within depressions in the dolomite; hence soil  $CO_2$  production is limited by a lack of soil cover and this in turn limits dissolved  $Ca^{2+}$  in the groundwater. Therefore, growth rates are low at this site when compared to all the other sites that have an mid-latitude maritime or continental climates and have complete soil cover and more abundant vegetation. Stalagmite growth rate may therefore determined by the limited vegetation cover at the site. Experiments involving the incubation of Clamouse soils gave very high  $pCO_2$  values comparable to those observed in other sites (Fairchild et al., 2000; however a mix of high  $pCO_2$  waters from vegetated patches and low pCO<sub>2</sub> from unvegetated bedrock can explain our observed dripwater calcium ion concentrations.

(2) The high Magnesium concentrations  $(0.94 \pm 0.37 \text{ mmol } 1^{-1}$ ; Fairchild et al., 2000 and unpublished measurements) could be the consequence of prior calcite precipitation. Consequently, the calcium concentration of the seepage water is less than that if prior precipitation had not taken place. Fairchild et al. (2000) demonstrate that the observed cave water Mg/Ca and Sr/Ca ratios can only be reached by prior calcite precipitation, therefore this process may

also be a significant factor affecting the observed growth rates.

### 3.3. Intra-annual dripwater hydrology and geochemistry and modelled intra – annual growth rates variations

Given the good agreement between modelled and observed growth rates over a wide range of growth rates and environmental factors (Section 3.1), we have confidence in using the modelled growth rate at an intra-annual level of resolution. For our sites, where we have continuous monthly (or higher resolution) sampling of the growth rate determining variables (La Faurie, Villars, Proumeyssac, and Postojna caves and Brown's Folly Mine), we use this data to model intra-annual growth rate variations. Given the known mean annual temperature at each site, the intra-annual growth rate of the stalagmites can be investigated given that growth rate depends on temperature, calcium ion concentration and drip rate. For all sites, we compared the modelled growth rate by using (1) just monthly Ca<sup>2+</sup> and temperature as determinants (GR1) and (2) using monthly  $Ca^{2+}$ , temperature and drip rate (GR2). GR1 and GR2 are well correlated for all the studied stalagmites (Table 2 and Fig. 4). However, the correlation becomes weaker for stalagmites that have a highly variable drip rate and a mean annual drip rate below 0.1 drip  $s^{-1}$ ; when monthly drip rate decreases below 0.05 drip  $s^{-1}$  at these sites (in summer and/or autumn) the growth rate is limited by insufficient replacement of water on the stalagmite cap as theorised by Drey-

brodt (1988) (Fig. 4). For the majority of the stalagmites, there is no difference between GR1 and GR2 as drip rates are maintained at between 2.0 and 0.1 drip  $s^{-1}$  throughout the year, and seasonal variations in Ca<sup>2+</sup> are the primary determinant of intra-annual growth rate variations (Table 1). Figs. 5–9 present



Fig. 8. Monthly (a) drip rate and (b) calcium ion concentrations for Villars. (c) Modelled intra-annual growth rate variations using temperature and  $Ca^{2+}$  concentrations as determinants.



calcium (a), drip rate (b) and modelled growth rate (GR1) (c) for all the sites.

Intra-annual geochemical and hydrological data from the five sites show the following:

•All the monitored drip sites demonstrate seasonal variations in recharge: it increases between September and December, stays high until the end of spring, and then decreases slowly during Summer and Autumn; we assume, for the interpretation of calcium and growth rate variations, that the seasonality of the flow rate is similar from one year to the next;

•For La Faurie, Villars and Proumeyssac, the major calcium concentration increases in June and July, which is 5–6 months after the winter drip rate increase; however we observed a second, smaller increase in autumn. For Postojna (Bri and Ent) and for Brown's Folly Mine, a single calcium increase occurs less than 2 months after the winter drip rate increase, in October and November 1997 for Postojna and March 1997 and January 1998 for Brown's Folly Mine. For Postojna PR2 and PR3 stations, calcium concentration does not vary significantly due to a high storage source;

•Because modelled growth rates are controlled primarily by calcium concentration, maximum growth rate occurs in spring and summer for La Faurie, Villars and Proumeyssac caves, with a second small increase in autumn, while they are maximum in autumn and winter for BFM and Postojna cave (Fig. 10).

•All the studied stalagmites have a modelled intra-annual growth rate that shows seasonal variations, with a typical intra-annual growth rate variation of  $\times 2$ ;

The difference observed in the timing of maximum growth rate can be explained by differences in sampling depth below the surface, karstification of the limestone or dolomite, and climatic regime. Postojna and Brown's Folly Mine (Figs. 6 and 9) both show an increase in discharge in winter and an associated increase in dissolved  $Ca^{2+}$ , reflecting the transport of soil water resident in the summer during the period of highest soil CO<sub>2</sub> production (for detailed discussion of Brown's Folly Mine, see Baker et al., 1999b). These results are in agreement with previous observations at Postojna by Gams (1965). Postojna Cave exhibits different timing of discharge peaks between winter and spring, associated with the large range of stalagmite sampling depths at this site (Bri: 120 m, PR2 and PR3: 37 m, Ent: 10m). (Fig. 9). In La Faurie, Villars and Proumeyssac (Figs. 5, 7 and 8) two growth rate peaks occur. We hypothesize

that the autumn increase is due to the flushing of high calcium concentration water, which is subsequently replaced by low calcium water once this source is flushed. We assume that the summer increase is due to an increased contribution of high calcium concentration stored water from the groundwater at low flow conditions. However, our interpretation of all these results does not consideration of



Fig. 9. Monthly (a) drip rate and (b) calcium ion concentrations for Postojna Cave. (c) Modelled intra-annual growth rate variations using temperature and  $Ca^{2+}$  concentrations as determinants.



the possible occurrence of prior calcite precipitation. If this phenomenon is significant in all of the studied caves (as demonstrated for Clamouse cave by Fairchild et al., 2000) then the calcium concentration of the seepage waters will be controlled also by this process. Consequently, the  $[Ca^{2+}]$  decrease observed in summer and fall for Villars, La Faurie and Proumeyssac caves might be due to this process, favoured by the dryness of these seasons. This would not change the modelled growth rates, but it will



Fig. 10. Synthetic graph showing periods of the year where modelled growth rates are maximum.

change their environmental significance. We are currently undertaking further studies to confirm the cause(s) of these two peaks.

### 4. Conclusions

We demonstrate that the mean annual growth rate at six cave sites throughout Europe is primarily dependent on the dripwater calcium ion concentration. In addition, there is a correlation between measured growth rate and mean annual temperature due to the correlation between calcium ion concentration. soil  $pCO_2$  and surface temperature. However, this is not a general rule because one of our sites. Grotte de Clamouse, has a high mean annual temperature but low dissolved  $Ca^{2+}$  concentration, due to either a prior calcite precipitation and /or a lack of soil cover diminishing soil CO<sub>2</sub> production. These results are of interest in the interpretation of stalagmite growth rates in Quaternary samples; natural vegetation changes in limestone areas that typically have a thin (< 20 cm) soil cover may lead to temporary soil loss and a reduction in groundwater  $CO_2$ , which will, in turn, generate a period of very slow stalagmite deposition or maybe even corrosion. But for time periods of constant vegetation, inter-annual growth rate variations are likely to be dependent on soil CO<sub>2</sub> production, which in turn has the potential to correlate with surface temperature. Such a result has important implications in the interpretation of stalagmite growth rates over the Quaternary period, as it suggests that with careful calibration and in association with other proxy evidence, growth rate may be used as a relative palaeoclimate index. However, the significant intra-site variations suggest that absolute palaeoclimate proxies cannot be inferred from growth rate. except for recent samples where it is possible to undertake a correlation with historical climate parameters.

Intra-annual calcium ion concentrations and drip rates at five sites demonstrate that there are seasonal intra-annual growth rate variations. For the majority of our study sites, we have chosen relatively fast drips, and for these sites drip rate is not an important determinant of intra-annual growth rate variations. Instead, growth rate is primarily dependent on the dissolved  $Ca^{2+}$  concentration. However, for very slow drip rate stalagmites ( < 0.01 drip s<sup>-1</sup>). or slow drips that show seasonal variations (< 0.01 drip  $s^{-1}$ ), drip rate becomes an important control on growth rate (Fig. 4). The understanding of such intra-annual growth rate variations is important for the interpretation of variations in annual laminae structures. Genty and Ouinif (1996) demonstrate that visible annual laminae can vary in structure in terms of the ratio of the width of the white porous calcite to dark compact calcite over the period  $10^{1}$ – $10^{2}$ years. In addition, previous research had suggested that when comparing visible and luminescent growth laminae, the dark compact laminae formed at the same time as the luminescent laminae. (Genty et al., 1997). It appears, in this study, that depending on the site, the storage time of the seepage water may be several months and can still maintain discrete geochemical signatures, and that calcium drip water concentration reaches maximum values at different seasons. This suggests that laminae maximum growth rate and possibly, laminae type (dark compact or white porous), may occur at a different season from one site to another. But we must keep in mind that prior calcite precipitation might occur in the unsaturated zone during seepage. This phenomenon might be also an important cause of the dripping water [Ca<sup>2+</sup>] variation and more work (Sr/Ca and Mg/Ca measurements) have to be done in order to demonstrate the significance of this process. Understanding the mechanisms of intra-annual growth rate variations goes some way towards the understanding of these structural variations. Baker et al. (1999a,b,c) demonstrate than annual luminescent laminae may vary in shape both in terms of the number of peaks and their shape and skewness. Such luminescent laminae are dependent both on the timing and intensity of organic matter fluxes as well as the relative growth rate within the year; the understanding of intra-annual growth rate variations should also permit the interpretation of band structures in terms of organic matter fluxes.

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