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THE PROSERPINE STALAGMITE (HAN-SUR-LESSE CAVE, BELGIUM): PRELIMINARY ENVIRONMENTAL INTERPRETATION OF THE LAST 1000 YEARS AS RECORDED IN A LAYERED SPELEOTHEM

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(11 figures & 2 tables)

ABSTRACT. A two meters long core from the big "*Proserpine*" stalagmite of the Han-sur-Lesse cave, deposited from 1800 (±75) U/Th years BP to recent, displays seasonal layering with two layers deposited during one year. Growth rates of up to 2.1 mm per year are observed. Similar changes are observed in the speleothem fabric, the $\delta^{18}O$ and $\delta^{13}C$ isotopic compositions and the growth rate along the longitudinal axis, suggesting common climatic and/or environmental controls. A first multi proxy approach is attempted, in order to interpret the constructed time-series in terms of changes in climate and environment. We interpreted the changes in proxy data from this speleothems mainly as changes in water recharge or precipitation minus evapo-transpiration. The stalagmite recorded a generally wet Little Ice Age period with two drier and/or colder decennia around 1600 and 1710. The stalagmite also registered the passage of early visitors in the cave from 1700 a AD with traces of fires made on top of the stalagmite and the incorporation of straw from torches in the stalagmite. During the upper 170 years an increasing number of visitors are responsible for the incorporation of soot in the calcite.

Keywords: Speleothems, paleoclimate, multi-proxy, archaeology, Little Ice Age.

1. Introduction

Calcite speleothems or secondary chemical cave deposits, are now generally considered as valuable high resolution palaeoclimatic proxy tools. Recent studies demonstrated their important contribution, especially in establishing precise chronologies for climate changes on the continent (Barr-Matthews et al., 1999; Genty *et al.*, 2003; Spötl & Mangini, 2002; Wang *et al.*, 2001, 2005). TIMS or ICP-MS uranium series dating of only a few 100 milligrams allow dating speleothem calcite with a precision of up to 1% (Li *et al.*, 1989; Shen *et al.* 2002). Counting of visible seasonal layering in laminated speleothems, permit dating with a precision of up to 0.5 year. Even if transfer functions are not always clearly determined, still valuable environmental and climatic information are successfully extracted from speleothems. In most speleothems from our regions, growth rate may give indications on the occurrence of favourable (warm and humid conditions) or unfavourable conditions for calcite precipitation (Genty et al., 2001). A good correlation between layer thickness and effective rainfall, i.e. rainfall minus evapo-transpiration, was observed in speleothems from Belgium (Genty & Quinif, 1996). By contrast, a correlation with precipitation and temperature was observed in Chinese stalagmites (Qin *et al.*, 1999; Tan *et al.*, 2003). Stable oxygen isotopic composition of speleothem calcite is related to a series of climatic factors, such as temperature, rainfall amount, source and storm tracks (McDermott, 2004). Fleitman *et al.,* (2004) related lower values of $\delta^{18}O$ and higher layer thickness to higher effective moisture and monsoonal rainfall in Oman stalagmites. Carbon isotopic composition is mainly related to soil activity (Genty *et al.*, 2003).

The Proserpine stalagmite comes from the speleothem ensemble "Le boudoir de Proserpine" located in the

Salle du Dôme within the touristic part of the Han-sur-Lesse cave system (southern Belgium). It is a tabular white stalagmite, which is fed by a rain of seepage water throughout the year. According to Perette (2000), this kind of "tam-tam" stalagmite is characterized by a rapid transfer of climate and environmental information from the surface to the cave. We expected therefore that the Proserpine stalagmite recorded seasonal and/or annual changes. A 10 cm diameter core, which was drilled along the longitudinal axis of the stalagmite, displays a very clear layering of white porous and dark compact layers over the last 700 years. We present here i) a description of the internal structure of the stalagmite and the influence of humans on the deposition of the stalagmite, ii) $\delta^{18}O$, $\delta^{13}C$ and growth rate time-series, and iii) the first results of the measurement of layer thickness. We discuss first indications for a climatic and environmental interpretation of the data, in the light of isotopic measurements of local rainwater and cave seepage water.

2. Methodology

A 200 centimetre long core was drilled in the Proserpine stalagmite in February 2001. Nine U-series age determinations were performed at Minnesota University (USA), using the procedures for chemical separation and purification of uranium and thorium of Edwards *et al.* (1987) and Cheng *et al.* (2000) on powdered samples of 100 to 400mg, taken with a dental drill. Measurements were performed on a Finnigan ELEMENT ionization coupled plasma mass spectrometer (ICP-MS) equipped with a double-focusing sector magnet in reversed Nier-Johnson geometry and a single MasCom multiplier, following procedures modified from Shen *et al.* (2002). Corrected ^{230}Th ages assume the initial ^{230}Th/^{232}Th atomic ratio of 4.4 ±2.2 x10^{-6}. Those are the values for a material at secular equilibrium, with a bulk earth ^{232}Th/^{238}U value of 3.8. The errors are arbitrarily assumed to be 50%. Uncertainties on the ages are given at 2σ level.

One ^{14}C dating was performed on straw embedded in the calcite at the Koninklijk Instituut voor kunstpatrimonium following the methodology of Van Strijdonck & van der Borg (1991). The ratio ^{12}C/^{14}C ratio was measured with AMS at Leibniz Labor (Christian-Albrechts Universität, Kiel, Germany). Fractionations were corrected with $\delta^{13}C$ measurements. Calendar ages were calculated with the OxCal software program (Stuiver *et al.*, 1998; Bronk Ramsey, 2001).

The $\delta^{18}O$ and $\delta^{13}C$ stable isotope analyses on speleothem calcite were performed following the method of McCrea (1950) at the Vrije Universiteit Brussel and at the Vrije Universiteit Amsterdam with a Kiel device coupled a Delta plus XL and a Finigan Mat 253 respectively. Results are given in ‰ VPDB. Analytical precisions were (2σ) ≤0.1‰ ($\delta^{18}O$) and ≤0.05‰ ($\delta^{13}C$), variations in measurements between the two laboratories lie within these uncertainties. Sub-samples from the Proserpine stalagmite were taken along the longitudinal axis of the core, every 0.5 cm for the upper layered part and every 2 cm for the lower un-layered part. Higher resolution sampling (approximately every mm) was carried out at chosen heights in the stalagmite (the upper seven cm, 24 to 28 cm, 50 to 56 cm. Very high resolution (every 100 micrometer, 200 micrometers deep) were performed with a *Micromill* at the Vrije Universiteit Amsterdam for the last 20 years.

Hydrogen isotopic measurements of local rainwater and cave seepage water, dripping on the Proserpine stalagmite were performed at the LSCE (CEA, France) on a homemade Isotope Ratio Mass Spectrometer (IRMS) with online uranium reduction (Hageman & Lohez, 1978; Vaughn *et al.*, 1998) which is usualy used for ice core water analysis. All the isotopic values are given in ‰ VSMOW. The analytical error on the deuterium measurement is typically 1.0 ‰ (2σ).

High-resolution image of a polished section of the stalagmite was obtained with a flatbed scanner. Each layer was manually marked using Adobe® Photoshop® and layer thicknesses were determined by semi-automatic image processing with ImageJ software (available at http://rsb.info.nih.gov/ij/).

3. Results

This 200 cm long core can be divided into two parts according to the physical properties (Fig. 1): the lower 125 cm (from 65 to 190cm) shows a chaotic pool-like morphology (small crystallization basins) (Fig. 2a), while the upper 64cm are regularly laminated with alternations of smaller compact brown to grey layers and thicker porous white layers up to 2 mm thick (Fig. 2b). From 36 to 33cm the stalagmite is composed of generally darker calcite with ambiguous layering. Between 13.5 and 9 cm, a clear sedimentological perturbation occurs with first deposition of brown layered calcite ending in a white granular calcite crust. Above, disconnected from the crust, un-layered calcite embedding pieces of straw occurs over approximately 2 cm. Above this perturbation, in the upper 9 cm, layered calcite is greyish and layers are much smaller than before. In total, for the layered upper 64 cm of the core, we counted 540 alternations (or 1080 layers) with 140 for the upper 9 cm.

Uranium-series dating indicates that the stalagmite started its growth at 200 (±75) a AD and ends at 2001 a AD, when the stalagmite was cored (Fig. 1, table 1). The U/Th datings are in logical stratigraphic order so they are believed reliable. Straw embedded in the calcite at approximately 10cm (Fig. 1) is dated by AMS, which further supports U/Th dating results (table 2).

Oxygen and carbon isotopic ratios (Fig. 3) display higher values up to -5‰ ($\delta^{18}O$) and -6‰ ($\delta^{13}C$) individually (Fig. 3) and important variations similar for $\delta^{18}O$ and $\delta^{13}C$

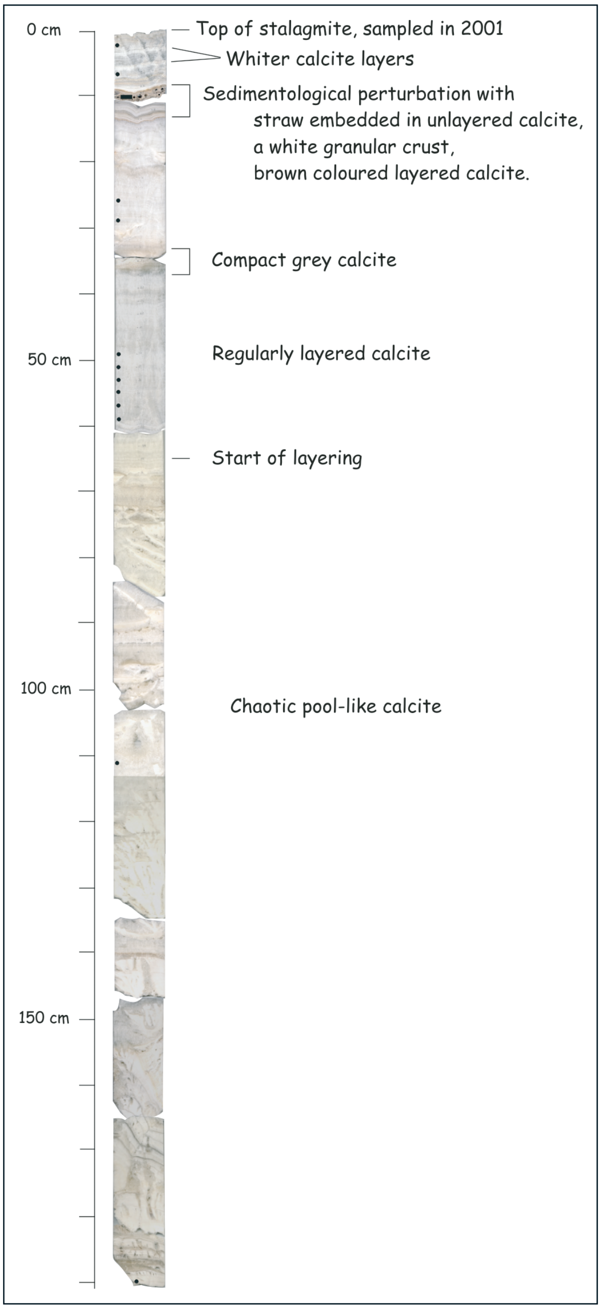

Figure 1. The Proserpine stalagmite core with its lower chaotic pool-like structure and its upper regularly layered calcite. Black dots represent U/Th datings locations. The black rectangle represent the location of the straw that was ^{14}C dated.

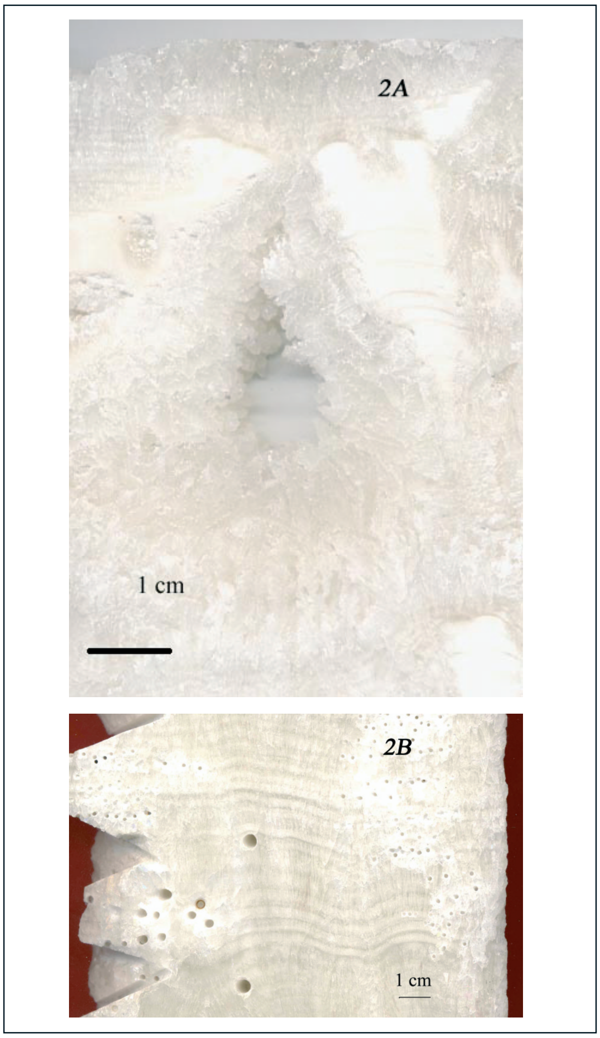

Figure 2. a: Chaotic pool-like structure of the lower part of the core. b: Regular layered calcite of the upper part of the stalagmite.

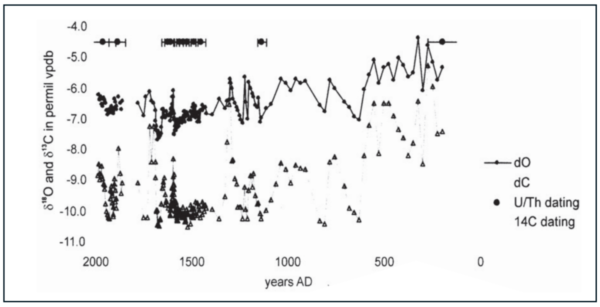

Figure 3. Growth rate, δ^{18}O and δ^{13}C time-series for the Proserpine stalagmite. Chronology is based on layer counting, ICP-MS U/Th dating of the calcite and ^{14}C dating of embedded straw. 2σ errors are displayed for the U/Th data. Horizontal lines represent the mean isotopic values.

in the lower non layered part of the core. A long-term decreasing trend is observed for δ^{18}O as well as for δ^{13}C. In the upper layered 64 centimetres, δ^{18}O displays a more regular large U shaped curve with values between -7.5‰ and -6‰ VPDB with short-term fluctuations. In recent times δ^{18}O increases strongly. In contrast, the δ^{13}C shows a rather flat long-term trend around -10‰ with short-term changes varying between -10.5‰ and -8‰ VPDB.. Six Hendy tests, for isotopic equilibrium (Hendy, 1971) were performed at different heights in the layered part of the stalagmite (Fig. 4). A significant (P≥0.95) correlation between δ^{18}O and δ^{13}C, indicating non-equilibrium deposition, was observed at 8.5 cm (R^2=0.94) and 12.5 cm (R^2=0.62), while no correlation was observed at 26.5, 33,

Table 1. ICP-MS U/Th dates of the Proserpine stalagmite. (2 σ uncertainties are given.)

Sample Number	mm from top	^{238}U (ppb)	$^{230}Th/^{232}Th$ (atomic x10^{-6})	$\delta^{234}U$* (measured)	$^{230}Th/^{238}U$ (activity)	^{230}Th Age (years) (uncorrected)	^{230}Th Age (years) (corrected)
6574-10	12	154.2 ±0.1	5.2 ±0.2	1390.7 ±1.8	0.00359 ±0.00017	164 ±8	**30 ±70**
6574-11	60	118.6 ±0.2	9.8 ±0.4	1396 ±4	0.00426 ±0.00015	194 ±7	**107 ±44**
6574-12	250	42.6 ±0.1	44.3 ±2.1	1329.4 ±2.3	0.00868 ±0.00035	408 ±17	**367 ±30**
6574-13	285	57.2 ±0.1	41.3 ±1.9	1347.7 ±4.1	0.00923 ±0.00039	430 ±18	**384 ±30**
6574-14	505	46.7 ±0.1	185 ±19	1402.9 ±4.2	0.00964 ±0.00051	439 ±23	**428 ±24**
6574-15	525	52.3 ±0.1	184 ±11	1409.8 ±3.0	0.01010 ±0.00040	459 ±18	**448 ±19**
6574-16	545	52.6 ±0.1	188 ±11	1392.8 ±3.3	0.01053 ±0.00039	481 ±18	**470 ±19**
6574-17	565	47.5 ±0.1	219 ±19	1394.9 ±4.2	0.01128 ±0.00048	515 ±22	**505 ±23**
6574-18	585	41.2 ±0.1	90 ±4	1392.9 ±2.9	0.01244 ±0.00039	570 ±20	**540 ±25**
6574-19	1120	54.4 ±0.1	252 ±10	1407.4 ±3.5	0.01916 ±0.00047	870 ±20	**860 ±23**
6574-20	1860	65.1 ±0.3	60.7 ±0.7	1367.3 ±2.0	0.04189 ±0.00049	1947 ±23	**1800 ±75**

$\lambda_{230} = 9.1577 \times 10^{-6}\ y^{-1}$, $\lambda_{234} = 2.8263 \times 10^{-6}\ y^{-1}$, $\lambda_{238} = 1.55125 \times 10^{-10}\ y^{-1}$.
*$\delta^{234}U = ([^{234}U/^{238}U]_{activity} - 1) \times 1000$. ** $\delta^{234}U_{initial}$ was calculated based on ^{230}Th age (T), i.e., $\delta^{234}U_{initial} = \delta^{234}U_{measured} \times e^{\lambda 234 \times T}$. Corrected ^{230}Th ages assume the initial $^{230}Th/^{232}Th$ atomic ratio of 4.4 ±2.2 x10^{-6}. Those are the values for a material at secular equilibrium, with the crustal $^{232}Th/^{238}U$ value of 3.8. The errors are arbitrarily assumed to be 50%.

Table 2. AMS ^{14}C dates of the straw embedded in the calcite.

PROS H (hout) KIA-24821: **255±35 BP**		
68.2% probability	1520AD (11.9%) 1550AD	Improbable compared to the U/Th ages
	1630AD (43,8%) 1670AD	Improbable compared to the U/Th ages
	1780AD (12.5%) 1800AD	probable
95.4% probability	1510AD (25.9%) 1600AD	Improbable compared to the U/Th ages
	1610AD (49.4%) 1680AD	Improbable compared to the U/Th ages
	1760AD (17.1%) 1810AD	**Most probable**
	1930AD (3.0%) 1950AD	Improbable compared to layer counting

34 and 49 cm. Along the longitudinal axis of the stalagmite core (Fig. 5), a strong correlation (at the 0,99 significance level) was observed between $\delta^{18}O$ and $\delta^{13}C$ for the upper 14 cm (R^2=0.71) and for the lower un-layered part (R^2=0.71), while a correlation of only 0.43 (R^2) was observed for the regularly layered part with thick layers in between (Fig. 5).

Hydrogen isotopic measurements of local rainwater (taken every two weeks between November 2003 and February 2004) indicates a high variability between -29‰ and 80‰ VSMOW, but a constant value of -51.8‰ ±1.0‰ (2σ) VSMOW for the cave seepage water, dripping above the Proserpine stalagmite and taken every week, between July 2003 and February 2004) (Fig. 6).

Measurements of total layer thickness between 1900 and 2000 a AD show a very similar variation pattern to the $\delta^{18}O$ (Fig. 7), with thicker layers corresponding to lower $\delta^{18}O$ values. Since dark layer thicknesses generally vary in the same way as white layer thicknesses, we further worked with total layer thickness.

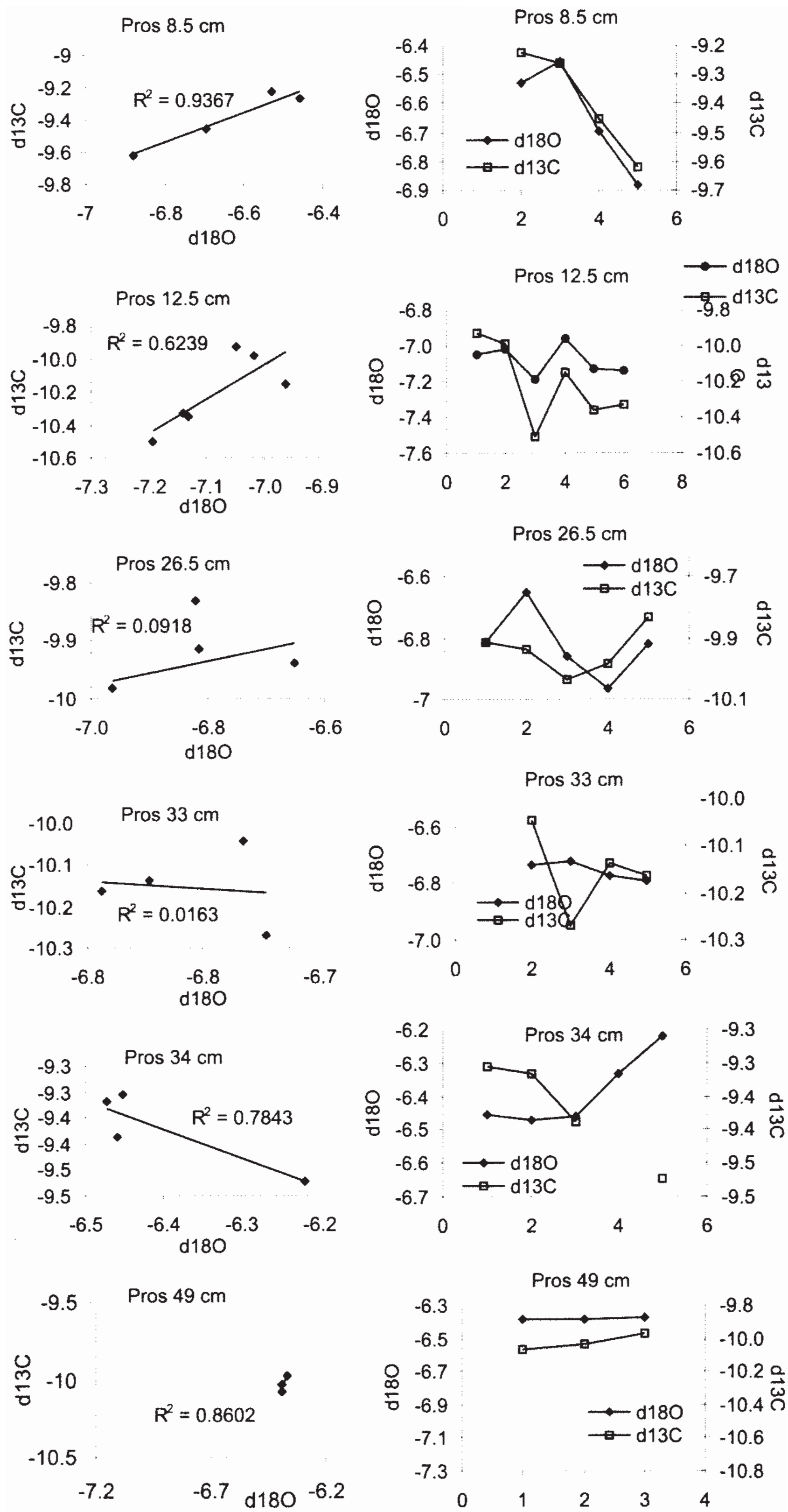

Figure 4. The Hendy tests (indicating non equilibrium deposition if $\delta^{18}O$ varies along a single layer and if $\delta^{13}C$ variations are similar to the $\delta^{18}O$ variations), performed at different heights in the Proserpine stalagmite indicate a deposition close to isotopic equilibrium for the calcite with a regular thick layering (from 64 to 14 cm) and non equilibrium conditions for the upper 14 centimetres.

4. Interpretation and discussion

4.1. Morphology

The differences in crystal fabric between the unlayered lower and layered upper part of the core is more probably due to the location of the coring at the side of the stalagmite than to climatic or environmental changes (Fig. 8), because in the upper part of the stalagmite, the core was well centred in the flat part, where regular laminated calcite is deposited directly from the dripping cave seepage water. In the lower part, the core crosses the side of the stalagmite where pool-like structures were deposited after a first deposition of calcite at the top. At the side of the stalagmite, precipitation from partly degassed water and evaporation processes, common in pool like structures, may change the initial isotopic and chemical signal contained in the water and carbonate. For this reason, we concentrate our study on the layered part of the stalagmite. Changes of calcite fabric inside the layered part may be of climatic, environmental or anthropogenic origin. If the onset of the sedimentological perturbation at 14 cm was not clearly related to a natural cause, the embedded straw is obviously anthropogenic and indicative of early visitors in the cave using torches. The greyish calcite in the upper 9 cm is most probably due to the incorporation of soot from the torches used by the visitors (Fig. 9). The deposition of whiter layer packages at 2.5 and 5.5 cm is possibly related to a decrease in the number of visitors. Since the end of the use of torches in 2000, a spectacular whitening of the speleothems is observed in the Salle du Dôme, especially for soda straw stalactites (Fig. 10).

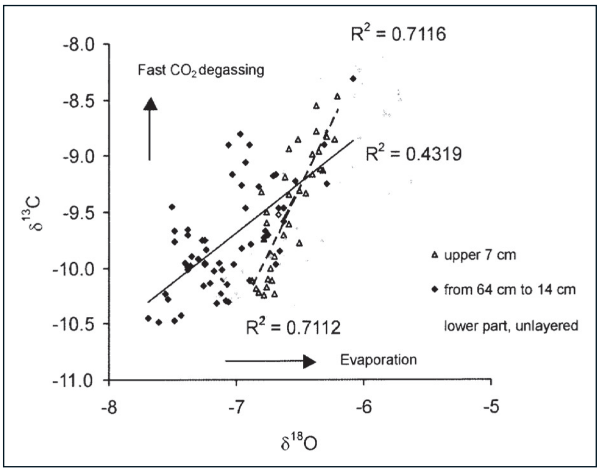

Figure 5. The upper 7 cm, as well as the lower unlayered lower part display a good significant correlation between $\delta^{18}O$ and $\delta^{13}C$ along the longitudinal axis, due to non-equilibrium deposition. The regularly layered part displays a poor but significant correlation, which expresses the climatic link between both variable.

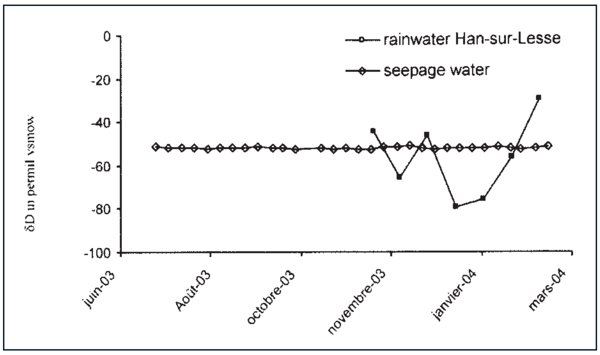

Figure 6. Deuterium isotopic composition of Han-sur-Lesse rainwater displays large variations while and Han cave seepage water is rather constant throughout the year.

4.2. Chronology and seasonal layering

The time-series are constructed based on layer counting, 9 ICP-MS U/Th dates and one ^{14}C date. The top of the core was deposited in 2001, date of coring. Layer counting from the top (2001 AD) down to the sedimentological perturbation (133 layer alternations) places the restart of layering at 1868 AD. The ^{14}C dating of the straw embedded in the calcite, deposited before the restart of layering, gives several possibilities after calibration (table 2). The most recent ages (between 1930 and 1950 AD) are in opposition with the layer counting. The ages, before 1680 AD are very unlikely since they are in opposition with the U/Th dating performed ±15 centimetres below and indicating an age of 1634 ±30(2s) AD. As a consequence and with a mean growth rate of 1 mm/year, the sedimentological perturbation, containing the straw, should start at least at 1750 AD (=1634-30(2s)+150mm/1mmper year). Therefore the most probable cal ^{14}C date seems to us the date between 1760 and 1810 (17% within 95% probability). It means that between the straw embedded at the end of the perturbation (maximum 1810 AD) and the restart of the layering (1868 AD), a hiatus of at least 60 years occured. For the part deposited before the perturbation, the U/Th dates were interpolated.

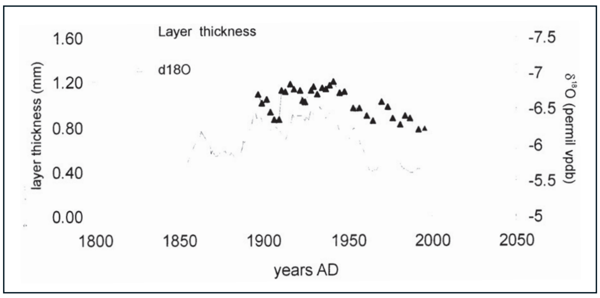

Figure 7. Total layer thickness and $\delta^{18}O$ values for the last century.

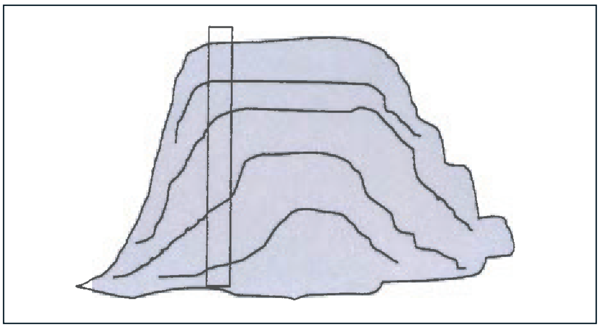

Figure 8. The core taken in Proserpine stalagmite. The upper part of the core crosses the stalagmite in its flat top, while the bottom of the core crosses the flank of the palaeo-stalagmite with a typical "micro pool" like structure.

Based on the datings it can be inferred that the studied 200cm high stalagmite was deposited in less than 2000 years, which indicates a high mean growth rate of over 1mm per year. The 540 layer alternations correspond well with the U/Th age of 540±25 a BP at the base of the layering and strongly support the seasonal character of the layers with an alternation of a white porous layer and a dark compact layer corresponding to one year. The time periods between the different U/Th ages correspond, within dating uncertainties, to the number of layer alternations. It is still unclear during which period of the year the different layers are deposited but former monitoring of the cave water in the Père Noël cave indicates a sudden increase of the drip rate and the conductivity of the cave water during autumn, when the decrease in vegetation activity permits an increase in water recharge (Genty & Deflandre, 1998). This suggests that the larger white porous layers with smaller crystals due to rapid calcite deposition from ionically charged water with chaotic dripping formed during the autumn, winter and maybe even spring period, while the dark compact layers with large elongated crystals formed during summer season when deposition from less charged water was slow with a more regular dripping.

Figure 9. The upper 10 cm of the Proserpine stalagmite. At its base, the straw embedded in brown calcite. Arrows indicate two whiter bands, related to the decrease in number of visitors during last two world wars.

4.3. Anthropogenic perturbation

We know from written historical sources that first touristic visits of the Han-sur-Lesse cave occurred around 1770 a AD (Timperman, 1989 and references herein). However, until 1814 a AD only a small part of the cave was visited and till now, we had no information about the knowledge and /or regular visit of the Salle du Dôme. The presence and dating of the straw, a relict of used torches or fires lit on top of the stalagmite as possibly suggested by the white crust of granular calcite indicates that the Salle du Dôme was already known between 1760 and 1810 AD. If the sedimentological perturbation starting at 1700 AD is of anthropogenic origin, visits could have started much earlier than inferred from the first known publications about the Han-sur-Lesse cave in 1743 AD. Regular visits have been recorded as grey soot-polluted calcite since 1870, when Eduard de Spandl bought the cave and equipped it to attract tourists (Bastin & Dulière, 1995). He placed 300 candles and a dozen fires in the Salle du Dôme. The visits continued up to sampling in 2001 AD with modern equipment but a historical show in the Salle du Dôme still used torches, which explains the soot incorporation up to 2001. During WWI and WWII, the cave was closed, explaining the whiter calcite deposited around 1917 a AD and between 1940 and 1945 a AD (Fig. 9). Since the tourist attraction with the torch was abandoned in 2000, a spectacular whitening of recently deposited calcite is observable in soda straws and in the upper layers (2001-2005 a AD) of the Proserpine stalagmite.

Figure 10. A soda straw stalagmite in the Salle du Dôme close to Proserpine. Note the recent whitening of the calcite at the tip of the soda straw.

4.4. Stable oxygen and carbon isotopic composition of the calcite

An important distinction can be made between un-layered and layered parts of the speleothem core. The heavier isotopic composition and larger variations in the lower part of the stalagmite may be related to non-equilibrium evaporation and degassing processes (common in pool environments) (Fig. 3). This is also suggested by the high correlation between $\delta^{18}O$ and $\delta^{13}C$ (R^2= 0.71, n=43, P≥0.999) along the longitudinal axis of the core (Fig. 5).

The layered part of the stalagmite consists of calcite with a $\delta^{13}C$ value in agreement with the C_3 type vegetation at the surface and with the carbon composition of modern Belgian stalagmites, deposited in conditions close to equilibrium (between -12‰ and -10‰) (Verheyden 2001). The oxygen isotopic composition is in agreement with close to equilibrium deposited calcite at ±10°C from cave water with $\delta^{18}O$ of -7.5‰ (Verheyden, 2001), corresponding to present-day conditions in the cave. The Hendy tests (Fig. 4) suggest that only the very regular layered part with thick layers is deposited close to isotopic equilibrium. The upper 14 cm with thin grey and brown layers, deposited during the last 300 years are deposited out of isotopic equilibrium. In these upper 14 cm, we also observe a high co-variation between $\delta^{18}O$ and $\delta^{13}C$ along the longitudinal axis as a consequence of non-equilibrium deposition. However, the non equilibrium processes may not be important enough to fully overprint the climatic signal, since the $\delta^{18}O$ and $\delta^{13}C$ values are still relatively close to the expected values.

In temperate climate settings and in isotopic equilibrium deposited calcite, lower $\delta^{13}C$ values are related to an increased soil activity, mainly occurring during warm and humid conditions (Genty *et al.*, 2003). The oxygen isotopic composition depends on a combination of factors such as air temperature, changes in the source and storm-track trajectories of the rain water and amount of precipitation. In general (with exception of Norwegian speleothems (Lauritzen & Lundberg, 1999)), a lower $\delta^{18}O$ was interpreted as representative of a colder period in Europe (Genty *et al.*, 2003; Baldini *et al.*, 2002; Spötl & Mangini, 2002). Verheyden (2001) calculated a change of approximately 0.2 to 0.3‰ per °C for our regions.

For calcite deposited in isotopic disequilibrium, $\delta^{18}O$ and $\delta^{13}C$ are dependent on kinetic processes such as evaporation and fast CO_2 degassing. In the studied regions, heavier $\delta^{18}O$ and $\delta^{13}C$ values have been observed to correspond to drier conditions (Verheyden, 2001, Niggeman *et al.*, 2003). Mickler *et al.* (2004) also observed that ^{18}O and ^{13}C enrichment measured in present-day Barbados speleothems are related to higher growth rates, due to increased CO_2 degassing or higher bicarbonate concentrations. However, experimental studies in the laboratory failed to demonstrate a dependency between precipitation rate and oxygen isotope partitioning in the calcite-H_2O system (Jimenez-Lopez *et al.*, 2001; Kim & O'Neil, 1997). It seems therefore obvious at this stage that both in-situ and experimental studies still not provide an understanding of the different controls on the isotopic incorporation in speleothems. Nevertheless, the different controls (growth rate, metal and bicarbonate concentrations, drip rate, temperature,…) are more or less influenced by climate factors as demonstrated by recent studies relating speleothem proxies to climate (McDermott, 2004 and references herein). More and more evidence indicate that isotopic equilibrium is rarely reached in speleothems and a combination of temperature and water availability or recharge, governing the drip rate, the residence time of water in host rock (itself controlling the ionic charge of the water), the local humidity conditions seem to be major controlling facors on speleothem growth, chemical and maybe even isotopic composition of the speleothems.

In recent times, anthropological perturbations should also be considered. Deforestation would increase the $\delta^{13}C$ as well as the $\delta^{18}O$ value, since it decreases the input of light carbon due to respiration and decomposition of soil organic matter and increases the effect of pure soil water evaporation, resulting in an increase of vadose water $\delta^{18}O$.

Since the layered part was deposited in quasi-isotopic equilibrium, we interpreted the long-term $\delta^{18}O$ change as a change in mean annual surface temperature. The stalagmite recorded a difference of approximately 1°C from the coldest period around 1600 - 1700 AD to the warmest period (last 30 years). Short-term changes are more susceptible to reflect changes in the extent of kinetic processes. Changes in water recharge, as main control, may be interpreted in terms of climate changes in different ways. A lower water recharge may indicate higher temperatures and thus higher evaporation at the surface before it infiltrates the soil or a lower rainfall amount or a combination of both.

The long-term $\delta^{13}C$ is rather constant around -10‰ during the last 700 years, indicating that the vegetation did not change drastically. Short-term changes, however, are important and especially anomalies similar to $\delta^{18}O$ anomalies should be regarded as characteristic of kinetic processes (changing both $\delta^{18}O$ and $\delta^{13}C$ in the same way) and therefore be brought in relation to dry-wet changes. An increase in $\delta^{13}C$ may therefore be interpreted as a dry and hot or a dry and cold but also as a wet and cold year, if $\delta^{13}C$ changes are due to a lower vegetation activity (in that case the change in $\delta^{13}C$ is independent of the change in $\delta^{18}O$). A decrease in $\delta^{13}C$ means a hot and wet or a wet and cold year (but where the cold is not strong enough to harm vegetation activity). Superimposed on that, anthropogenic deforestation also needs to be taken into account. This complexity clearly demonstrates that the sole isotopic composition of speleothem calcite will not provide enough information to reconstruct climatic or hydrological changes and that accurate climate reconstruction should rely upon the comparison of different proxy data in the same speleothem, as well as in the comparison between different speleothems from the same cave and from different caves.

4.5. Stable isotopic composition of the cave water

A solution to better constrain transfer functions between climate and isotopic composition of speleothems is to directly study this transfer for recent times, in order to use a similar transfer in the past. First measurements on δD isotopes (Fig. 6) display a rather constant δD composition for the cave seepage water, while, as expected, a high variability is observed in the δD of local rainwater. Since δD is generally more sensitive than $\delta^{18}O$, we expect a similar behaviour for the $\delta^{18}O$. Previous results on $\delta^{18}O$ in other parts of the caves of Han (Verheyden, 2001) indicate a constant $\delta^{18}O$ for the cave water, representing the annual mean of local rainwater $\delta^{18}O$. Measurements are in progress to confirm this for the seepage water feeding the Proserpine stalagmite. Based on isotopic measurements, the cave seepage water may, therefore, be considered as mean local rainwater, thus with an annual residence time in the vadose zone. The fact that the drip rate rapidly reacts to variations in the amount of rainfall as observed by the cave guides, should be explained by the occurrence of a "piston effect", with new recharge water pushing down old already mixed vadose water (Tooth & Fairchild, 2003) or by the occurrence of complex flow patterns (pore and fissure flows) with different residence times and where the isotopic characteristics of the water are influenced mainly by a long (approximately annual) residence time (Bottrell & Atkinson, 1992). As a consequence, it will probably not be possible to observe important seasonal variations in isotopic composition of the calcite layers, but other proxies may display a clear seasonal signal or register flood events due to differences in drip rate. A clear example of such a seasonal signal is the layered structure of the stalagmite.

4.6. Growth rate, layer thickness and $\delta^{18}O$.

Once chronological markers are present, the growth rate of the different parts of the stalagmite can be calculated. We reconstructed a rough growth rate time-series by comparing packages of 10 to 20 layer alternations (or years) with their corresponding length of calcite deposition along the longitudinal axis. The growth rate is commonly 1 mm/year, and extremely high growth rates (up to 2.1 mm/yr) are observed between 1450 and 1550 AD. A drastic decrease in growth rate (under 1 mm/yr) together with a change in calcite fabric occurs at ca 1610 AD, 1530 AD, around 1650 AD, and at 1700 AD (Fig. 12). During the embedding of the straw at ca 1785 a growth hiatus of approximately 100years probably exists.

In association with other proxies, the growth rate of stalagmites may be used as a relative palaeoclimate indicator (Genty *et al.*, 2001). Growth rate is primarily dependent on the cave seepage water calcium ion concentration, linked with the residence time of the water in the limestone and thus with water recharge and with the amount of soil CO_2 controlled by vegetation activity. In addition, there is a correlation between growth rate and mean annual temperature, due to the correlation between calcium ion concentration, soil pCO_2 and surface temperature (Genty *et al.,* 2001) or due to a pH effect, dominated by soil activity (Frisia *et al.*, 2003). Fleitmann *et al.* (2004) demonstrated a strong correlation between layer thickness (and thus growth rate) and precipitation in the Oman monsoon climatic system. A correlation between growth rate and water supply is demonstrated in our regions by Genty & Quinif (1996) and by Genty *et al.* (2001). At this point of the study, we could not relate the growth rate to temperature or precipitation in the Proserpine stalagmite.

A precise time-series (with annual resolution) of layer thickness established by computer-aided techniques, is available for the last 100 years. Since layer thickness was demonstrated in our regions to be linked with water supply (Genty & Quinif, 1996) and since $\delta^{18}O$ values are related to precipitation (Verheyden, 2001, Niggeman *et al.*, 2003), a first comparison between these two parameters was performed (Fig. 7). The variations in layer thickness for the period between 1850 and 2000 AD display similar variations as $\delta^{18}O$ (despite a poor overall correlation of $-0.4 = R^2$), which may suggest a relationship between $\delta^{18}O$ and water amount. Correlation between temperature and rainfall data for the last 100 year are not conclusive but further investigation in the exact relation between these both potential proxies are continuing. Water supply (or recharge) can be very different from rainfall amount, especially during hot years when evapo-transpiration is high. Therefore, further investigations will focus on the relation between effective precipitation or water recharge (precipitation minus evapo-transpiration) and $\delta^{18}O$ and layer thickness.

The close similarity between $\delta^{18}O$ and layer thickness (or growth rate) changes may also suggest a dependence between growth rate or precipitation rate and isotopic composition, which therefore joins the idea of Mickler *et al.,* (2004). Till now, however, no significant seasonal $\delta^{18}O$ changes were observed in the Proserpine calcite layers, while seasonal variations in growth rate between the layers are important. Ongoing higher resolution measurements may bring new insights in this problematic but also further detailed in-situ studies are necessary to empathize with the isotopic behaviour.

4.7. A preliminary multi proxy approach

As each individual proxy has its own limitations and problems, it seems clear that a multi-proxy approach is necessary. The different proxies may be partially interdependent but they are all controlled in a more or less direct way to climate and environment. The Proserpine stalagmite displays simultaneous changes in isotopic composition, growth rate or layer thickness and crystal fabric (Fig. 11). We made a chronological reconstruction with the most important features and a first reflection on possible environmental or climatic controls.

The start of layering at 1365 a AD, is probably more a sedimentological change due to the position of the core in the stalagmite than linked to an environmental or climatic change. The non layered lower part with microgours was deposited at the flank of the paleo-stalagmite with occurrence of strong non equilibrium processes affecting the oxygen and carbon isotopic composition as suggested by the high correlation between $\delta^{18}O$ and $\delta^{13}C$ along the longitudinal axis. The upper layered part is therefore more suitable for paleo environmental reconstruction since this part, with exception of the upper 14 cm was deposited close to equilibrium. U/Th datings indicate a higher growth rate of 2.44 mm/yr between 1140 and 1365 a AD (onset of layering) than for the base of the core (0.79 mm/yr) deposited between 200 a AD and 1140 a AD. This high growth rate may be related to favourable conditions for speleothem deposition (hot and relatively wet) of the Medieval Warm Period (800 – 1300 a AD). After a short decrease around 1350-1400 a AD (onset of Little Ice Age), highest growth rates are observed from 1450 to the end of the 16th century (between 1.2 and 2.1 mm/yr). According to the Proserpine speleothem, the climate during this period did not affect vegetational activity drastically and low $\delta^{18}O$ values are consistent with a high water recharge and thus a sufficient humidity level at the surface. Around 1600 a AD, a clear simultaneous signal occurs in the calcite fabric (deposition of grey compact calcite), the growth rate (sudden decrease) and the Oxygen and carbon isotopic compositions (sudden decrease of $\delta^{18}O$ and $\delta^{13}C$). All these changes suggest a decrease in water availability, combined with a lower vegetational activity (since the $\delta^{13}C$ increase precedes the $\delta^{18}O$ increase, the $\delta^{13}C$ change can not be only due to kinetic processes). The deposition of compact translucent calcite (with low growth rate) seems typical of cold periods as also seen during the 8.2 ka BP event in a stalagmite from the Père Noël cave (Verheyden, 2001). Therefore the time-series suggests a colder and drier period around 1600 in our region, probably characterised by cold and dry winters and humid fresh summers, unfavourable for agriculture. This interpretation can account for the drop in growth rate together with an increase in $\delta^{18}O$ and $\delta^{13}C$ and deposition of darker compact calcite at 1650 a AD. On the contrary, the drop in growth rate of 1630 a AD is confirmed neither by a change in isotopic composition, nor by a signal in crystal habit.

The sedimentological perturbation at 1700 a AD occurs simultaneously with an important increase of the $\delta^{18}O$ and $\delta^{13}C$ compositions suggesting a climatic cause for the onset of the perturbation (decreased recharge with a less important soil activity) rather than an anthropogenic cause. In addition, from between 1760 and 1810 a AD, humans visited the cave and left behind their burned torches or lit fires on top of the Proserpine stalagmite. Calcite deposition stopped for approximately 100 years possibly not only due to the visits but also due to unfavourable calcite depositional conditions, as indicated by the high $\delta^{18}O$ (decreased water recharge) and high $\delta^{13}C$ values (lower soil activity) at both sides of the hiatus. The lower recharge also explains why Proserpine was an ideal

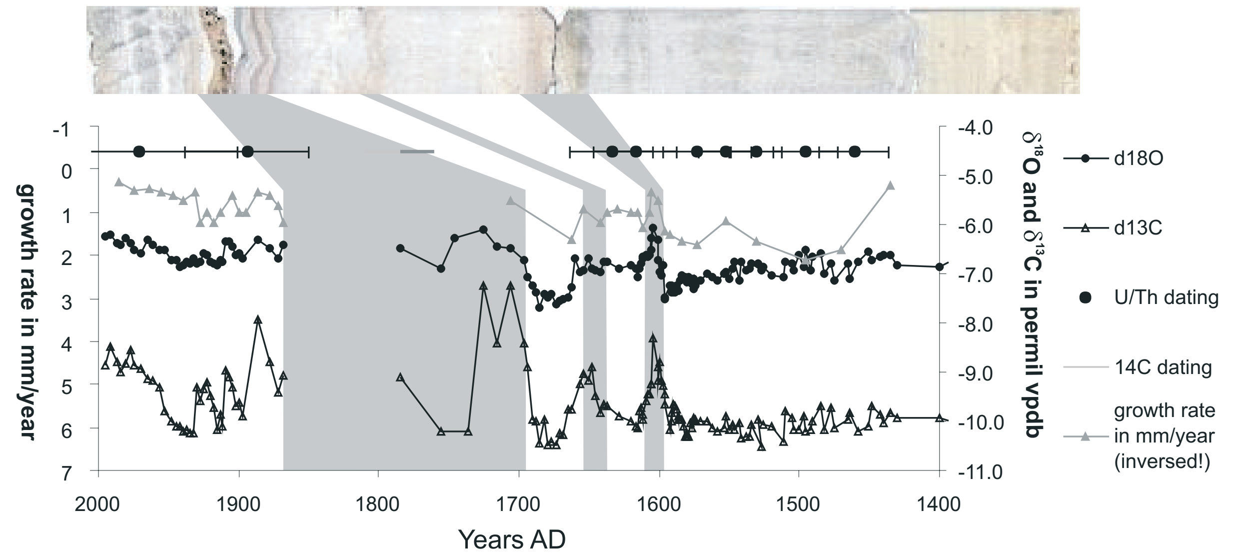

Figure 11. $\delta^{18}O$, $\delta^{13}C$ time-series and growth rate time-series for the layered part of the Proserpine stalagmite. Chronology is based on layer counting, ICP-MS U/Th dating of the calcite and ^{14}C dating of embedded straw. 2sigma errors are displayed for the U/Th data. Horizontal lines represent mean isotopic values.

place to make a fire, since besides of the strategic location in the Salle du Dôme and the flat top, less water was probably dripping on the stalagmite. Today, this stalagmite would probably not be chosen due to the continuous "rain" falling on the entire surface of the stalagmite. For the last 100 years, $\delta^{13}C$ values were low while $\delta^{18}O$ seems rather stable (high soil activity and stable recharge conditions) around 1930-1950 a AD. A recent strong increase in $\delta^{13}C$ (decreased soil activity) and $\delta^{18}O$ values (lower water recharge due to dryer/warmer conditions?) is observed in the speleothem since 1950 a AD. Since 1880 a AD regular visits with torches in the cave deposited a grey soot layer on the stalagmites. Deposition of white calcite around 1917 and between 1940 and 1945 a AD is consistent with a decrease of visits during the world wars. The soot is disappearing only recently (visible in recent soda straws) with the end of the use of torches as tourist attraction in the Salle du Dôme.

5. Conclusions

The Proserpine stalagmite grew between 200 and 2001 a AD and displays a seasonal layering since 1350 a AD. Simultaneous changes in different parameters such as crystal fabric, growth rate or layer thickness and isotopic composition, indicate that they are controlled by a common mechanism: climatic, environmental and/or anthropogenic processes. However, caution is required since high similarity of changes may suggest a common control, probably linked with depositional dynamics. Nevertheless, even if partially interdependent, these parameters depend more or less directly on climatic fators, mainly the amount of water recharge, which is closely related to precipitation, air temperature, and vegetation coverage.

The Proserpine stalagmite provides information on the climatic and/or environmental evolution of the Han-sur-Lesse region as well as archaeological information, such as the early visiting of the Salle du Dôme from 1760-1810 and a lowering in frequency of visits during both world wars. Oxygen and carbon isotopic composition, combined with growth rate and crystal habit suggests warm and humid conditions for the 11th to 14th centuries. Coldest periods occurred at 1600-1610, 1650 and 1700-1750 a AD. During the last 100 years a gradually dryer and/or hotter climate is registered in the stalagmite.

The exact transfer functions between climate/environment and proxies in the stalagmite still need to be studied more in detail by further in-situ studies of the nowadays transfer of the cave water and its characteristics. δD measurements reveal the lack of seasonal signal in the isotopic composition of cave seepage waters. Comparisons between recent and historical meteorological data and proxies in the stalagmite indicate a more complex interaction than a simple correlation between a single proxy and temperature or amount of rainfall.

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