Microstratigraphic logging of calcite fabrics in speleothems as tool for palaeoclimate studies

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Abstract: The systematic documentation of calcite fabrics in stalagmites and flowstones provides robustness to palaeoclimate interpretation based on geochemical proxies, but it has been neglected because it is difficult to transform crystal morphologies into numerical values, and construct fabric time series. Here, general criteria that allow for coding fabrics of calcite composing stalagmites and flowstones is provided. Being based on known models of fabric development, the coding ascribes sequential numbers to each fabric, which reflect climate-related parameters, such as changes in drip rate variability, bio-mediation or diagenetic modifications. Acronyms are proposed for Columnar types, Dendritic, Micrite, Microsparite and Mosaic fabrics, whose use could then render possible comparison of calcite fabrics in stalagmites and flowstones from diverse latitudinal and altitudinal settings. The climatic and environmental significance of similarities in the geochemical signals and trends analysed in coeval stalagmites and flowstones (or differences in the signals and trends) will be more robust when compared with fabric time series. This is particularly true where, such as in the Holocene, changes in geochemical values may be subtle, yet fabrics may show changes related to variations in supersaturation, drip rate or input of detrital particles or organic compounds. The proposed microstratigraphic logging allows recognition of changes in stable isotope ratio or trace element values that can be ascribed to hydrology and diagenesis, with considerable improvement of reconstructions based on the chemical proxies of stalagmites and flowstones composed of calcite.

Keywords: speleothems; fabrics; calcite; aragonite; diagenesis; microstratigraphy

Received 22 June 2014; Revised 29 October 2014; Accepted 3 November 2014

Citation: Frisia S., 2015. Microstratigraphic logging of calcite fabrics in speleothems as tool for palaeoclimate studies. *International Journal of Speleology*, 44 (1), 1-16, Tampa FL (USA), ISSN 0392-6672 <http://dx.doi.org/10.5038/1827-806X.44.1.1>

INTRODUCTION

Documentation of speleothem fabrics is fundamental for a robust interpretation of speleothem time series where stable isotopes and trace elements are used as proxy (Fairchild & Baker, 2012). Fabrics provide valuable information on post-depositional phenomena (diagenesis), such as phase transformations (aragonite to calcite), aggrading neomorphism (Frisia, 1996), dissolution and re-precipitation processes, which may alter the original geochemical signal. Petrographic observations should not be confined to the sole identification of diagenesis, particularly when U-series dating is problematic (Ortega et al., 2005; Hoffmann et al., 2009; Lachniet et al., 2012). For example, by observing changes in columnar calcite types, high-frequency variability of $\delta^{13}\text{C}$ values in stalagmites could be explained as related to changes in drip rate and degassing (Boch et al., 2011).

A comprehensive classification of fabrics which accounts for growth mechanisms and relates

morphologies to hydrology and drip-water properties (chemistry, presence of organic compounds, particulate etc.) was compiled by Frisia & Borsato (2010). Yet, there has been little advance in the use of fabrics as a complement to palaeo-proxy datasets since Frisia et al. (2000). Although it has been demonstrated that speleothem fabrics record hydrology, bio-influenced and post-depositional processes that influence the way geochemical proxy data are captured by cave minerals (Fairchild & Baker, 2012) their use is limited by the absence of a system that allows to construct a fabric time series comparable with geochemical time series.

Here, a speleothem microstratigraphic log is proposed as a logic system of creating fabric time series. The log is based on the hierarchical coding of fabrics typical of stalagmites and flowstones and builds upon accepted models for their development. The microstratigraphic log relies on the attribution of a progressive number to fabrics as observed by Plane Polarized (PPL) and Cross Polarized Light (XPL) at the optical microscope, which reflects a hierarchic system

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that gives the lowest numbers to fabrics developed under conditions of constant drip of waters at low supersaturation with respect to calcite and low Mg/Ca ratio and/or negligible presence of impurities, and the highest to post-depositional (diagenetic) fabrics. The coded fabrics are thus plotted against age model, thus allowing comparison with geochemical time series. The microstratigraphic log is an inexpensive tool that requires a microscope and thin sections and some practice. It is recommended to use more sophisticated techniques, such as electron backscattered diffraction and TEM techniques need to be used (Fairchild & Baker, 2012) only when there is the need to know processes of crystal nucleation and growth processes and/or complex phase transformation.

The documentation of fabrics in a petrographic log was pioneered in a speleothem-based European Holocene palaeoclimate reconstruction (McDermott et al., 1999). The log highlighted for the first time that certain fabrics, such as a scaffold-like dendritic fabric, coincided with systematic shifts to ^{13}C -enriched calcite. More recently, Luetscher et al. (2011) used microstratigraphic logs to reconstruct glacier mass balance fluctuations in the Swiss Alps. Belli et al. (2013) compared microstratigraphic logs, growth rates and stable isotope time series of two speleothems from the same cave in NE Italy to recognize how “site-specific” processes drove stable isotope ratios variations which allowed for the distinction of the hydrologic vs. hydroclimatic component to the isotope signal.

The coding of fabrics in the construction of the microstratigraphic log must be conceptually connected to parameters related to climate, which influence speleothem crystal growth. Only in such conceptual framework, the log becomes useful to provide robustness to the geochemical data or test the overall accuracy of the speleothem record. It becomes, thus, apparent that the most critical issue in its development is the appropriate coding of fabrics, which are significant for the type of speleothem-based study that is being conducted. Given that fabrics are recognized and defined by microscopy observation, coding is a somehow a subjective evaluation. It is, therefore, proposed here that coding shall be rooted on a general knowledge of how fabric relate to their environment of formation. Critically, there are uncertainties in recognizing the origin of fabrics whose development can be primary or the result of post depositional (diagenetic) processes. The study of speleothem diagenesis is still in its infancy, therefore, examples are here provided for fabrics that are accepted as being the result of diagenesis. It is then proposed that some fabrics, such as mosaic calcite and microsparite, are unequivocally of diagenetic origin. The basis of the hierarchy for coding fabrics as proposed here is provided in the following section.

MODELS OF FABRIC DEVELOPMENT: BASELINE FOR CODING

The logic underneath ranking stalagmite and flowstones calcite fabrics as proposed in Table 1 is based on our current understanding of the parameters

that result in their development. Fabrics develop under certain environmental parameters, most important fluid flow and presence of impurities, which influence a certain spatial arrangement of crystals with a dominant form (or a few dominant forms). Calcite shows a large variety of morphologies, but known speleothem fabrics can be grouped in a few broad categories: columnar types, dendritic, micrite, microsparite and mosaic calcite (cf. Frisia & Borsato, 2010). The proposed order, from the columnar types to mosaic calcite, is a plausible hierarchic system that takes into consideration changes in drip rate (flow), progressive increase in calcite saturation index (where known) and Mg/Ca ratio and, finally, diagenetic transformation.

Columnar Fabrics

Columnar compact and open

Columnar calcite proper (C in Table 1) consists of crystals with the c axis commonly at 90 to 60° to the substrate, unit extinction, length fast, with a length to width ratio < 6:1 (Frisia et al., 2000). The acute and equant rhombohedra are the predominant individual crystal forms (Dickson, 1993). Columnar fabrics are characterized by competitive growth, or geometric selection, whereby crystals compete growing away from an interface. Crystals whose greatest growth vector is perpendicular to the substrate will be favoured, those with greatest growth vector orientated in different directions will be outcompeted. Above the interface, thus, small crystals with “random” growth directions are observed, as testimony of a nucleation episode (Fig. 1C). A crystal fabric with preferred orientation than develop in the “maturation stage” (Dickson, 1993) and the elongation can be with the slow (length slow) or the fast (length fast) optic ray parallel to crystal length. Commonly, columnar fabric consists of length-fast crystals, and Transmission Electron Microscope diffraction patterns have shown that the cleavage rhombohedron (10.4) is a common form (Frisia et al., 2000). Thus, both the cleavage and the acute rhombohedron (40.1) potentially create aggregates of crystals with straight to serrated boundaries with the elongation parallel to the fast optic ray and perpendicular to the substrate (cf. Dickson, 1993). Geometric selection should occur when growth of a stalagmite or flowstone is interrupted by an interface that requires re-nucleation.

When the crystals form a compact aggregate, with welded crystal boundaries, where inter-crystalline porosity is not discernible at the optical microscope, the fabric is columnar compact (C in Table 1). When intercrystalline boundaries are marked by the presence of linear inclusions, or pores, then the fabric is columnar open (Co in Table 1 and Fig. 1 A-B).

The established general model for the development of compact (complete coalescence of crystallite) and open (incomplete coalescence) columnar calcite was established by Kendall & Broughton (1978), whereby the first type forms from a thin film of fluid, under relatively slow drip rate and enhanced degassing conditions (enhanced cave ventilation) and the latter forms from a thicker film of fluid, under higher drip

rate and less efficient degassing. Frisia & Borsato (2010) observed that, in temperate climate settings, C fabric forms under relatively constant discharge, low (up to 0.35) calcite supersaturation state (SI_{cc}), low Mg concentration in the dripwater (Mg/Ca ratio < 0.3) and negligible

presence of particulate and/or organic colloids. Boch et al. (2011) illustrated that lamina couplets of compact and open columnar calcite in stalagmites from Katerloch cave in Austria developed from drip waters characterized by low Mg/Ca ratio (mean 0.01). The complete

Table 1. Calcite Fabrics, types and acronyms, code numbers and acronym of special features, which may provide additional information.

Fabrics	Type	Acronym	Progression in coding	Acronym for additional features
Columnar	Compact	C	1	C_{rt} ; C_{lf}
Columnar	Open	Co	2	Co_{rt} ; Co_{lf}
Columnar	Elongated	Ce	3	
Columnar	With lateral overgrowths	Ce_{lo}	4	
Columnar “spherulitic type growth”	Fascicular Optic	Cfo	5	
	Radiaxial fibrous	Crf	5.5	
Columnar	Microcrystalline	Cm	6	
Dendritic		D	7	D_o (Dendritic open)
Micrite		M	8	Hm = hiatus with micrite layer
Diagenetic	Microsparite	Ms	9	
Diagenetic	Mimetic replacement of aragonite fabrics by microsparite	Ms_{mra}	10	
Diagenetic (?)	Mosaic calcite	Mc	11	
Diagenetic	Mosaic calcite with visible aragonite needle relicts	Mc_{an}	12

Fig. 1. Columnar Types. A) Elongated columnar, XPL, stalagmite from Liang Luar Cave, Flores Indonesia; B) Protruding crystal terminations in a stalagmitic flowstone, PPL, from Cave 370 in the Nullarbor, Australia; C) Columnar fabric showing lateral overgrowths marked by red arrows, XPL, Conturines Cave, N Italy. Note that the lateral overgrowths are not the same as randomly oriented crystals that, after nucleation, do not survive for the law of geometric selection. The lateral overgrowth “stem” from the stepped faces of a columnar individual, which survived thanks to geometric selection. D) Fascicular Optic Calcite (FOC), XPL, from a stalagmitic flowstone collected in Cave 370 Nullarbor, Australia. E) Map showing average Mg concentrations in FOC calcites in the same sample illustrated in D. Average Mg concentration in FOC is ca. 20,000 ppm. The FOC nucleated on a micrite/microsparite layer with Mg concentration up to 30,000 ppm.

coalescence of “crystallites” in the Katerloch stalagmites was attained under low drip rates, high rate of degassing and water pH of ca 8.4, and a low HCO_3^-/CO_3^{2-} ratio, which favoured horizontal linear growth rate. By contrast, the open columnar type resulted from incomplete coalescence of “crystallites” in laminae produced by high drip rates, low rate of degassing, pH from 7.4 to 8 and, consequently, high HCO_3^-/CO_3^{2-} ratios, which promoted vertical linear growth rates. More positive C isotope ratios values in the compact columnar laminae then should reflect enhanced degassing, typical of low drip rates combined with intense cave ventilation (Boch et al., 2011).

Within the open and compact columnar calcite fabrics, it is common to observe growth surfaces characterized by well-developed rhombohedra terminations, and/or flat terminations. Rhombohedra terminations are more common in the open columnar type. Flat terminations have been observed in association with dark, organic-rich laminae

(Frisia et al., 2000 and Frisia et al., 2005), which suggests the possibility that organic matter oxidation may promote localized dissolution resulting in the overall “flat” appearance of the crystal tips (cf. Frisia, 1996). The different types of crystal terminations, thus, encapsulate environmental significance and their distinction in a microstratigraphic log can be useful information. A subscript to describe morphology of crystal terminations is suggested in Table 1: C_{rt} , Co_{rt} or C_{lf} where *rt* stands for protruding rhombohedra terminations (cf. Fig. 1B) and *lf* for laminae, flat.

Because columnar compact and columnar open columnar fabrics result from growth processes that imply a combination of low/high drip rate and high/low degassing, the C isotope ratios in the two types may be more/less modified by kinetics. Given the common, low Mg/Ca ratio range measured to date in their parent waters (0 to 0.3), trace element variability may likely reflect impurities trapped within intercrystalline spaces in the open type. The distinction between C (compact) and Co (open columnar) thus becomes important, as it provides useful information to distinguish a climatic from a local hydrological signal (Belli et al., 2013).

Elongated Columnar type

Elongated columnar (Ce) is composed of crystals with a length to width ratio of 6:1, most commonly 2 up to 60 mm long, and 0.2 up to 5 mm wide. These figures can be, in certain cases, exceeded, particularly where stalagmites do not show visible interfaces and re-nucleation phenomena. According to Dickson (1993), the elongated columnar fabric is the result of the preferential growth of the acute rhombohedron (40.4). Gonzalez et al. (1992) observed that development of acute rhombohedra in speleothems from Indiana, Kentucky, New Mexico and Puerto Rico resulted from the combination of supersaturation state, Mg/Ca ratio in the parent water and flow. Elongated arrays of crystals required fluid flow over the surface of the speleothem, a Mg/Ca ratio in the parent fluid ranging from 0.85 up to 2.8 and SI_{cc} ranging from 0.1 to 0.4. Frisia et al (2000) reported that elongated columnar fabric in speleothems from temperate settings in Europe developed under constant drips, SI_{cc} ranging from 0.1 to 0.35 and Mg/Ca ratio higher than 0.3.

Gonzalez et al. (1992) further proposed that elongated columnar fabrics develop from relatively fast flow that accelerates crystal growth perpendicular to the substrate and high nucleation rate (Gonzalez et al., 1992). This interpretation was then contrasted by Kendall (1993), who demonstrated that the formation of elongated columnar fabrics neither imply fast flow, nor high nucleation rates. Thus, it seems plausible to hypothesize that elongated columnar type forms from constant drip (water flowing at the speleothem surface as inferred by Gonzalez et al., 1992), similar SI_{cc} but higher Mg/Ca ratios (>0.3) than for columnar compact and open.

Elongated columnar calcite fabric is common in flowstones developed from parent waters whose catchment intersected dolomitic or Mg-rich rocks. Turgeon & Lundberg (2001) observed elongated

columnar type in flowstones from a cave in the Klamath Mountains (Oregon, USA) where the host rocks consist of marble, argillite and basalt. The elongated columnar fabric they described is characterized by large, acute crystal terminations, exceeding 750 μm , which have been interpreted as indicative of high flow in interglacial periods (Turgeon & Lundberg, 2001). The protruding terminations should favour a relatively turbulent flow, which would prompt degassing, increase the precipitation rate and promote incomplete coalescence of crystals and development of linear inclusions. Flat terminations, or rhombohedra terminations <200 μm then, should be interpreted as documenting low flow (Turgeon & Lundberg, 2001).

Elongated crystals may show lateral overgrowths (Ce_{lo} in Table 1), in particular when micrite-rich layers are present in flowstones (cf. Frisia, 1996) (Fig. 1C). The formation of this type of elongated columnar calcite has not yet been fully explained. A plausible model of formation would be growth under relatively fast flow, similarly to the open columnar calcite, where intercrystalline spaces are occluded by organic colloidal particles that subsequently become the nucleation sites for micrite (cf. Freytet & Verrecchia, 1999). Freytet & Verrecchia (1999) also documented diagenetic replacement of micrite by elongated calcite crystals with overgrowths in freshwater stromatolites. When “clots” of micrite are observed in sinters (flowstones) consisting of elongated columnar fabric with lateral overgrowths, thus, diagenesis may have occurred. Thus, the interpretation of elongated columnar calcite with lateral overgrowths and relicts of micrite clots may imply diagenesis in flowstone layers, which were once porous and rich in organic particulate. This interpretative model does not disagree with the mechanism of formation of elongated columnar calcite proposed by Kendall (1993), although, it may imply post-depositional changes (Frisia, 1996). In such case, it is expected that some chemical properties were re-set and caution should be taken when interpreting stable isotope ratio values variability (or lack of it) recorded in flowstones consisting of Ce_{lo} associated with relict micrite.

Spherulitic type growth in columnar fabrics: Fascicular Optic, Radial.

These fabrics consist of columnar polycrystals (length to width ratio > 6:1, commonly > 10:1) characterized by undulatory extinction due to the systematic variations of the orientation of the c-axes with respect to the substrate of “fibre-like” individuals that compose each polycrystal. If the c-axes of the crystal units that compose the polycrystal diverge from the centre outward, these are similar to the Fascicular Optic Fibrous Calcite (FOFC) described in Kendall (1985), (Fig. 1D). This fabric can then be labelled Columnar Fascicular Optic (Cfo) and its overall appearance is that of a sector of a spherulite, or a fan, consisting of bundles of elongated crystals bending outward. If the polycrystals possess a pattern of converging fast vibration directions, that is undulatory extinction converge away from the

substrate, then they are similar to radiaxial calcite (Kendall & Tucker, 1973; Kendall and Broughton, 1978; Kendall, 1985). Fabrics consisting of polycrystals showing brush extinction converging away from substrate when the rotating table is turned clockwise should then be labelled Columnar radiaxial fibrous (Cr_f). Both C_{fo} and Cr_f columnar types are characterized by curved cleavage, with upper concave curvature in the case of Cr_f and downward concave curvature in the case of C_{fo}. The extinction characteristics of Radiaxial calcite and Fascicular optic calcite are thoroughly described and illustrated for polycrystals in Neuser & Richter (2007).

These two fabrics are here grouped together because they form through a mechanism typical of spherulitic growth (cf. Sunagawa, 2005). This implies splitting of a crystal into a number of units with slightly diverging lattice orientation. When split growth is systematic, it is manifested as an “extinction brush”, or sweeping (undulatory) extinction on a polarizing microscope. Kendall (1993) proposed that the generation of fascicular optic and radiaxial calcites was related to crystal splitting caused either by high supersaturation or by the presence of ions (or colloids) that poisoned crystal surfaces. The formation of polycrystalline aggregates with spherulitic-type growth has been documented through experimental settings that used organic compounds and Magnesium (Mg) as additives. Magnesium typically affected crystal morphologies by altering the calcite nucleation sites and eventually caused crystal splitting (Meldrum & Hide, 2001).

In stalagmites from St. Michaels Cave in Gibraltar, low-Mg calcite polycrystals with sweeping, fascicular optic type extinction, formed under low drip rates (0.04 to 0.06 litres/day), from parent water with Mg/Ca ratio > 1.5 and SI_{cc} of 0.5 (Mattey et al., 2010). Radiaxial Fibrous Calcites were documented in speleothems sampled in caves cut in dolomitic rocks (Neuser & Richter, 2007), which confirms an important role for Mg in driving the formation of spherulitic-type fabrics. The occurrence of C_{fo} and Cr_f seem to be typical in stalagmites and flowstones whose calcite Mg content is high (cf. Folk & Assereto, 1976), but still within the range of Low-Mg calcite, typically from >10,000 to <30,000 ppm (this study, Fig. 1E). Yet, the boundary conditions that shift the system from one that favours the development of elongated low-Mg calcites, to one that favours that of spherulitic-type fabrics (and finally the precipitation of aragonite) depend on a series of variables factors, including: temperature, the SI_{cc} and Mg/Ca ratio, the carbonate ion concentration and the presence of organic compounds in the parent waters (Meldrum & Hide, 2001). Because all these factors interfere with each other, it becomes difficult to set precise Mg/Ca ratio and SI_{cc} values for cave waters at which the transition from elongated to fascicular optic or radiaxial fabric occur. It is reasonable, however, to infer that Mg/Ca ratio of the parent water needs to exceed 0.35.

In terms of environmental and/or climate significance, an increase in Mg concentration in speleothems is commonly related to dry periods, when drip rate is low, transport of colloid from the soil

should be reduced, Mg, Sr, Ba concentration should increase and $\delta^{18}\text{O}$ values of the drips should become more positive (Orland et al., 2014). It is reasonable to assume, then, that C_{fo} or Cr_f are indicative of Prior Calcite Precipitation (PCP) and/or prolonged water-rock interaction (WRI) in dolomitized aquifers during dry episodes. The observation that C_{fo} is common in flowstones from caves cut in dolomite, however, also suggests that flow may influence crystal splitting, as already inferred by Gonzalez et al. (1992). For spherulitic growth, in fact, it is necessary that the growth front receives a continuous supply of “building material” from the solution, in order for the “platelets” originating from a nucleus to keep growing in the fastest direction (Sand et al., 2011). Thus, it is here proposed that the presence of C_{fo} in a speleothem implies an increase in Mg concentration in the parent water and laminar flow. In the case of the stalagmitic flowstone from a Nullarbor cave illustrated in Fig. 1D and E, the highest concentration of magnesium, up to 30,000ppm, is recorded in micrite layers capping the C_{fo} fabric and marking surfaces above which re-nucleation and geometric selection occurred. Hence, it is likely that the C_{fo} fabrics formed only when the speleothem surface was continuously wetted by the regular flow of a thin film of fluid. Under conditions of strong cave breathing in the Nullarbor caves PCP increased the Mg/Ca ratio in the parent waters (Wong et al., 2011). When fluid was not regularly flowing at the surface, than micrite rich in magnesium, rather than C_{fo}, developed.

Overall, the presence of C_{fo} and/or Cr_f indicates that speleothems formed under conditions of at least constant flow, and suggest that cave breathing or ventilation may result in kinetic modifications of the geochemical signals encoded in the two fabrics. From the available literature data, and this study, it is reasonable to assume that the columnar fabrics with spherulitic type growth most likely develop from dripwaters with Mg/Ca $\geq \sim 0.4$ and $\leq \sim 3$ and SI_{cc} $\geq \sim 0.3$ and $\leq \sim 0.5$, that is, higher than those required for the development of elongated, compact or open columnar fabrics.

Columnar Microcrystalline type

The columnar microcrystalline type (C_m in Table 1) has been observed in stalagmites, not in flowstones and distinguished as a distinct type of columnar fabric in specimens from Italian Alpine settings. The C_m fabric is characterized by polycrystals with length to width ratio < 6:1, highly irregular intercrystalline boundaries, uniform extinction, punctuated by inter- and intra-crystalline microporosity (Frisia et al., 2000). The C_m is typical in speleothems showing laminae rich in organic colloidal particles (Fig. 2 A, B) (cf. Frisia et al., 2000). The notable difference with the typical columnar fabric is in the highly irregular crystal boundaries, which developed because of the presence of foreign particles that favoured the formation of crystal defects (see the examples illustrated in Fig. 7.18c in Fairchild & Baker, 2012) visible at the Transmission Electron Microscope (TEM). At the TEM, C_m polycrystals are clearly defect-ridden, with dislocations, lamellae and subgrain boundaries

Fig. 2. Microcrystalline, dendritic and associated fabrics. Note how subtle changes in the arrangement of crystals distinguish open columnar, where the single composite crystals are mostly elongated along the c axis, from dendritic, where crystals branch in a scaffold-like texture. A) and B) Microcrystalline fabric at XPL, in a stalagmite from Grotta di Ernesto (N Italy) showing irregular boundaries, porosity, and domains of uniform extinction with irregular boundaries. These domains consist of polycrystalline aggregate. C) and D) Co fabrics at XPL in a stalagmite from Grotta di Carburangeli (S Italy) characterized by strong seasonal contrast in drip rate. In C) the composing crystals are $> 50 \mu\text{m}$ long and single crystals, thus the fabric is not microcrystalline. Crystals do not branch, thus the fabric is not dendritic. D) shows incipient branching crystals with interfingering boundaries. The branching fabric inside the circle can be considered dendritic, because the composing crystals form an angle and show diverse extinction patterns. Nevertheless, the distinction is very subtle and, for the sake of simplicity, the overall fabric can still be classified Co. E) Real dendritic fabric, with scaffold-like morphology clearly visible in PPL in a stalagmite from Tham Doun May, Laos. F) The SEM image of the same specimen highlights the presence of semi-spheres attached to the calcite crystal surfaces, which consist of amorphous SiO_2 , a likely a by-product of microbial metabolism. This observation suggests both hydrologic instability and presence of microbial populations at the stalagmite surfaces as possible necessary component in a genetic model for the development of dendritic fabric.

(Frisia et al., 2000). This results in high intracrystalline microporosity that distinguishes Cm from the Co type, which is characterized by intercrystalline porosity. When comparing Cm and Co in thin section (Fig. 2A-C), the open columnar type shows relatively large, linear pores between each crystals, whereas the microcrystalline type has serrated boundaries, and because of the composite nature of each individual with uniform extinction, some of the “crystallites” of adjacent polycrystals cross-cut each-other, creating areas where the extinction is different from that of cross-cut individual. The presence of intracrystalline

porosity and impurity-rich layers results in the opaque and milky appearance of Cm type in the polished hand specimen with visible “flame-like” polycrystals. Co type is also opaque and milky at the naked eye, but shows regular arrangement of parallel columnar crystals, similar to an open palisade.

The distinction between Co and Cm is useful when logging the microstratigraphy of a speleothem. Although they both form from parent fluid at low supersaturation with respect to calcite (SI_{cc} 0.1 to 0.35, Frisia et al., 2000) and variable drip rate, Cm formation implies input through the feeding system of organic colloids and particulate “in excess” when compared to the conditions of formation of Co. The “foreign” particles form a regular layer at the growing surface when the combination of low degassing and high drip rate results in an undersaturated (or barely at saturation) state for the film of fluid wetting the speleothem. Colloidal particles are most probably adsorbed at defect sites on the rugged surface of calcite crystals, and subsequently incorporated in the speleothem, possibly between single “crystallites” composing any of the polycrystal (see Fig. 5F in Frisia et al., 2000). The adsorption of foreign particles or organic molecules triggers development of crystal defects (Cölfen, 2003) typical of the Cm fabric.

In stalagmites from Alpine settings, the combination of low supersaturation, low degassing and increase in flushing of colloidal particles occurs in autumn, when soil efficiency diminishes because plants become dormant (Frisia et al.,

2005). It is, thus, reasonable to hypothesize that the development of Cm is typical in stalagmites from temperate regions characterized by marked seasonal contrast in temperature, vegetation activity and autumnal rainfall. Its development requires also seasonal changes in cave ventilation, with a less efficient exchange between cave and atmospheric air occurring when the inflow of colloidal particle from the soil is greater. To date, available information about the vegetation cover suggests that Cm forms in Alpine caves developed below mixed conifers and deciduous forest (Frisia et al., 2000).

Dendritic Fabric

The Dendritic (D in Table 1) fabric is composed of branching polycrystals, with each branch consisting of stacked (10.4) rhombohedra. Under the optical microscope, two branches form an angle of ca. 90°, and are inclined of ca. 45° relative to the substrate. This V-shaped aspect of the fabric, which is particularly visible in cross polarized light, distinguishes D from Co, and its overall appearance resembles a scaffold, or warp and waft in a woven cloth, defining rounded to elongated pervasive porosity, rather than series of linear pores as in Co (Fig. 2D). The well-defined, rod-like crystal boundaries, branching into the adjacent crystal and the high intercrystalline, rather than intracrystalline, porosity distinguishes D from Cm. In the hand specimen, D fabric is characterized by milky, opaque appearance due to high intercrystalline porosity, related to the voids between the “warp and waft” which may then be sites where particles, impurities or fluid inclusions are trapped. Figures 2E and F illustrate dendritic fabric characterizing a stalagmite from cave Tam Doun May, in Laos. The dendritic fabric did not develop in all the stalagmites from the cave, but only in those with relatively fast drip, where a portion of the dripwater infiltrated within the speleothems, rather than flowing at their surfaces. Scanning Electron Microscopy observations show that the fabric is indeed very porous, where voids are defined by the arrangements of rods in a scaffold-like structure. The flanks of each rod are characterized by macrosteps that accommodate hemispheric particles. The hemispheres entirely consist of Si, most probably an amorphous phase, and it is reasonable to assume that they are indicative of bio-influenced precipitation (Frisia et al., 2012). Preliminary culture data on swabs taken at the tip of stalagmites in Tam Doun May cave reveal the presence of diverse microbial species, some derived from the soil zone, but others endemic (Evans, pers. comm., 2014). This supports growing evidence for bio-influenced calcium carbonate precipitation in caves (Banks et al., 2010) and suggests that some speleothem fabrics, such as D, may be the result of bio-influenced mineralization. McDermott et al. (1999) documented for the first time the dendritic fabric in stalagmites from Crag Cave, and reported that the layers consisting of D fabric were characterized by more positive $\delta^{13}\text{C}$ values relative to C fabric layers in the same specimen. This phenomenon was attributed to more enhanced CO_2 degassing during D fabric formation. Subsequently, Frisia et al. (2000) documented that the development of D fabrics required variable discharge, and a slightly higher SI_{cc} (up to 0.4) than the C fabrics formed in the same Alpine regional settings. The Tam Doun May D fabric reported in the present study suggests that bio-influenced precipitation should be included in its model of development in addition to degassing and hydrological instability. If microbial influence in carbonate precipitation plays the most important role in the formation of the scaffold-like structure typical of D fabric, then it becomes possible that a stalagmite consisting (or having layer) of D-fabric should not be discarded on the assumption that degassing influences the C isotope ratio. More specifically, if the D fabric is

still porous in a sample, that is, if there has not been syndepositional occlusion of the pores by cementation, then the fabric may preserve geochemical signals that record rainfall and soil efficiency at the time of formation. To test the accuracy of D fabric in recording climate and environmental signal, it is recommended to run a few stable isotope ratio analyses in sectors of the stalagmites to be used for palaeoclimate reconstructions that are characterized by more and less porous D fabrics, and compare the results with C fabrics from the same sample (if present). If the C isotope ratios of D have more negative values than in the compact C, one could infer that kinetic processes did not modify the signal in D. In palaeoclimate studies, the best strategy of sampling when speleothems show D fabric is to compare geochemical time series in two coeval specimens from the same cave with diverse stalagmite morphologies.

Micrite fabric

By micrite fabric (M in Table 1) it is here intended a fabric consisting of crystals whose max dimension is 2 μm , which commonly appears dark in PPL (Fig. 3A) and dark brown in XPL (Fig. 3B). Micrite is one of the most intriguing speleothem fabrics, because the precipitation of micrite in experimental studies commonly requires an exceptionally high number of pre-existing nuclei, high supersaturation and/or the presence of organic compounds (Morse et al., 2003). In seawater, which has a calcite saturation index that greatly exceeds that of cave waters, abiotic formation of micrite appears to require nucleation on “old” sediment and is clearly favoured by organic molecules (Kaźmierczak et al., 1996; Morse et al., 2003). In continental environments, mechanisms of micrite formation in laminar calcretes and tufas implies a biotic intervention, such as precipitation mediated by cyanobacteria (Kaźmierczak et al., 1996; Alonso-Zarza & Wright, 2010). Micrite fabric has been observed associated with bio-influenced calcite moonmilk fibres and in calcareous tufa (Borsato et al., 2000; Frisia & Borsato, 2010). Similarly, micrite fabrics characterizes stromatolite-like structures in speleothems from the Swiss Alps grown under a glacier, where these structures mark periods of glacier retreat, highlighting that micrite is a palaeoglaciological proxy (Luetscher et al., 2011). Stromatolite-like micrite layers were observed in Holocene stalagmites from a mid altitude cave in the Dolomites and in Pliocene stalagmitic flowstones from the Nullarbor, where they coincide with reduction or cessation of the common abiotic speleothem growth processes (Frisia & Borsato, 2010; Frisia et al., 2012). Epifluorescence (UV) observations support the hypothesis that micrite fabric in stromatolitic-like structures is related to organic compounds and, most likely, to microbial laminae (Fig. 3C-D). Studies on Ca-homeostasis of cave bacteria suggest that cave microbes influence the formation of micrite (Banks et al., 2010). Thus, it appears plausible that the presence of micrite fabric is indicative of bio-influenced processes, which has important implications for palaeoclimate studies as micrite layers may be associated with shifts to more

positive values in the C isotope ratios (cf. Kaźmierczak et al., 1996). In the Nullarbor sample illustrated in Fig. 1E, for example, the $\delta^{13}\text{C}$ values shift from -10.5‰ in Cfo to -4.0‰ in stromatolitic-like micrite (M) layers. This phenomenon was interpreted as a possible result of microbial colonization of the speleothem surface during a relatively dry period (Frisia et al., 2012).

Micrite may be also a “destructive” fabric (Cañaveras et al., 2001). Etching, biomechanical “micritization”, and condensation-corrosion may all create micrite

(Cañaveras et al., 2001). Micrite associated with aragonite relicts has been considered a “destructive” fabric, formed through condensation-corrosion (Martín-García et al., 2009; Martín-Pérez et al., 2012). However, aragonite needles in many speleothems are replaced by microsparite ($> 2\ \mu\text{m} < 30\ \mu\text{m}$), rather than by micrite (Folk & Assereto, 1976; Frisia et al., 2002). Furthermore, in Antro del Corchia cave speleothems, micrite associated with aragonite forms layered “clots” that are not converted to microsparite (Fig. 3E).

Because of the high surface to volume ratio of micrite, one would expect that the small crystals were converted to a microsparite mosaic when aragonite rays were transformed into low-Mg calcite, as observed by Folk & Assereto (1976). It is, therefore, unclear whether micrite fabric is a diagenetic product of condensation-corrosion or a primary fabric. Most probably, micrite associated with stromatolite-like structures is a primary fabric, whereas micrite associated with aragonite can be diagenetic or not. To date, a general model for the genesis of micrite in stalagmites and flowstones does not exist because all occurrences of micrite reported in the literature pertain to fossil specimens. From the analogy with stromatolitic-structures and experimental data, it is plausible to infer that micrite development in speleothems consisting of calcite is favoured by bio-mediation in a regime of relatively low discharge (Frisia et al., 2012).

The diagenetic fabrics: Microsparite and Mosaic calcite

Microsparite (Ms in Table 1) consists of crystals $>2\ \mu\text{m}$ and $< 30\ \mu\text{m}$ in diameter arranged in a mosaic of anhedral to sub-euhedral crystals. Its origin is still uncertain, but a model for the genesis of microsparite needs to be proposed to assist in the logics behind the construction of a petrographic and microstratigraphic log that bears palaeoclimate significance. Microsparite is inferred to be the product of aggrading neomorphism of micrite (Folk, 1965; Folk & Assereto, 1976). The process of neomorphism implies the *in-situ* transformations of a mineral in itself (replacement of calcite by calcite) or by a polymorph (replacement of aragonite by calcite) (Armenteros,

Fig. 3. Micrite and microsparite. A) and B): flowstone from Cave Grotta di Conturines, N Italy. A) Micrite (M) in PPL marking protruding both rhombohedra and flat crystal terminations of columnar fabric. The black arrow shows a thin micrite layer coating crystals with flat terminations, indicative of a very thin film of fluid wetting speleothem top. Ms, microsparite (Ms) aggrades micrite and, most probably, transforms in coarser columnar calcite via diagenesis (ripening). B) Thin micrite layer (white, thin arrow) aggrading into microsparite. Above the microsparite, and partially incorporating it, is an example of columnar calcite with extreme splitting, which induces a “feathery” appearance (the “coconut meat calcite” of Folk & Assereto, 1976) to the crystals when viewed, as in this photo, in XPL. The “feathery” calcite has a relatively high content of Mg, similar to that of the FOC illustrated in Fig. 1D. C and D: Micrite, “stromatolite-like” layer capping columnar crystals in a stalagmite from Cogola di Giazzeria, N Italy. C) is viewed in PPL and D) in UV epifluorescence. The micrite layers exhibit fluorescence, which indicates that the fabric incorporates organic compounds. By contrast, microsparite (Ms) and columnar crystals between the micrite layers, do not show fluorescence. The micrite forming stromatolite-like coatings (see Fig. 13 in Frisia and Borsato, 2010) has been interpreted as associated to bio-influenced precipitation (see text for details. E) Micrite associated with aragonite transformation. The aragonite needles are being replaced by a mosaic of microsparite, but micrite has not been aggraded into microsparite. Photo taken in PPL, in a stalagmite from Antro del Corchia Cave, Italy. F) Epifluorescence image of microsparite rhombs grown on aragonite rays in a subaqueous speleothem from Tommy Graham Cave, Nullarbor, Australia. The microsparite rhombs have a fluorescent core and growth bands. This suggests that organic compounds may influence microsparite growth. The microsparite may then act as the trigger for the replacement of aragonite.

2010). Neomorphism is believed to take place through dissolution of the “precursor” on one side of a film of fluid and precipitation of the neomorphic phase on the other side of the film. The replacement of micrite by low Mg-calcite microsparite is most probably a process of aggrading neomorphism (Frisia, 1996). This is driven by the instability of very small crystals with high surface to volume ratio. But in order to occur at the low temperatures under which speleothems commonly form, the crystal size must be very small, which is the case of micrite (cf. Fairchild et al., 1994). Aggradation would occur when a flow of saturated solutions enters in contact with micrite (Fairchild et al., 1994), thus, the replacement of micrite by microsparite should imply a partial (or complete) opening of the system.

Microsparite is commonly observed in association with micrite in stromatolite-like layers within stalagmites (Fig. 3C, D) and in dark, organic-rich laminae (Frisia, 1996). In these cases, organic matter oxidation may have promoted sin-depositional dissolution-re-precipitation, with little consequence on the preservation of original geochemical signals, including the mobilization of U. The Microsparite fabric is also typical as a replacement phase in aragonite needles (Fig. 3E), in the form of mosaics of calcite crystals that mimic single aragonite crystals morphologies (Frisia et al., 2002). In speleothems, the process of neomorphism with replacement of a precursor by calcite microsparite may be favoured when discharge resumes after a dry period, where either aragonite or micrite had previously formed.

The process of microsparitization may also be influenced by the presence of inorganic or organic additives, which may control both the morphology of the precipitates and polymorphism (Sand et al., 2011; Rodríguez-Navarro & Benning, 2013). In speleothem-forming environments, the presence of organic compound and microbes is bound to be ubiquitous, and speleothem formation may not be dictated by sole inorganic processes (cf. De Choudens-Sánchez & González, 2009). For example, in some phreatic speleothems from the Nullarbor, calcite microsparite crystals growing on aragonite needles show at their nuclei UV fluorescence, which also marks growth stages of the rhombohedra (Fig. 3F). It is, thus, reasonable to infer that the development of microsparite is facilitated by the presence of organic compounds. It is plausible to infer that a bio-influenced mechanism favour the mimetic replacement of

speleothem aragonite fabrics into microsparite (cf. Frisia et al., 2002). In summary, it is here proposed that occurrence of microsparite fabric in speleothems is indicative of diagenetic processes and it is suggested to consider Ms as a diagenetic fabric.

The Mosaic fabric (M in Table 1) consists of euhedral to sub-euhedral low-Mg calcite crystals > 30 µm and < 1 cm in diameter, commonly showing triple junctions (angle between faces of three adjacent crystals = 120°). Each crystal in the Ms fabric typically includes ghosts of dark, needles, which are a distinguishing characteristic of calcite replacing aragonite (Frisia et al., 2002; Fairchild & Baker, 2012). The occurrence of Ms fabric has been, to date, mostly related to speleothem aragonite transformed into calcite (Fig. 4 A-C). Mosaic calcite, however, has been reported also as the product of dissolution and reprecipitation of former low-Mg calcite (cf. Fairchild & Baker, 2012). It is unclear, as yet, if mosaic calcite is the final product of neomorphism in a micrite → microsparite → sparite series in continental carbonates (Armenteros, 2010) or a product of dissolution of columnar calcite and re-precipitation of mosaic calcite driven by influx of undersaturated waters causing dissolution of a pre-existing fabric, and reprecipitation of sub-euhedral crystals. The latter case would be expected, as an example, when glacial meltwaters invaded cave systems during deglaciations.

Fig. 4. Diagenetic fabrics and Aragonite. A) Mosaic fabric. General appearance of mosaic calcite (Mc) replacing aragonite. Photo taken in PPL, stalagmite from Liang Luar cave, Flores, Indonesia; B) The same as A but seen at XPL; C) Relic aragonite clearly visible within the subeuhedral to euhedral mosaic of calcite spar, same speleothem as in A and B; D) Micrite (M) associated with aragonite needles in a stalagmite from Antro del Corchia, Italy. Note the dark appearance in PPL of the micrite, due to its fine grained size. Microsparite (Ms) appears lighter, has crystals larger than micrite and replaces aragonite needles, but there is no clear replacement of micrite. The mosaic calcite (Mc) almost completely blurs the original aragonite fabric.; E) Elongated columnar calcite capped by acicular fabric radiating from a micrite layer. The needle-like crystals entirely consist of calcite, although their terminations may appear commonly square. There is no indication that the calcite needles replaced aragonite needles in the form of relic aragonite. Rather, it seems as if the crystals grew constrained by impurity-rich micrite. Flowstone from Grotta di Collalto, Italy, PPL.

Given that most speleothems formed at relatively low temperature, and that microsparite is more stable than micrite, it seems plausible that sparite mosaic is not the product of ripening of microspar (cf. Fairchild et al., 1994). Then, mosaic fabric consisting of sparite, without evidences of former aragonite needles within any euhedral or sub-euhedral individual, should be considered a diagenetic product of dissolution and re-precipitation of any of the primary calcite fabrics, most probably of the most porous, such as dendritic or microcrystalline or columnar open.

Aragonite transformation into Calcite as documented by the diagenetic fabrics

Microsparite and Mosaic calcite are considered, in the present study, diagenetic fabrics, which, in most cases, document aragonite to calcite transformation. Diagenetic mosaic calcite destroys the original aragonite fabrics, be it rays or needles (see Figs. 4C, D), and the preservation of metastable polymorphs “ghosts” (Fig. 4C) depends on the flow (cf. Armenteros, 2010), whereby “diagenetic” fluids may (or may not) be in contact with a “metastable” phase depending on the original porosity. The unequivocal presence of relict aragonite must be identified by X-Ray diffraction (XRD) with a diffractometer or by Electron Backscattered diffraction on a polished section. Quantification of the percentage of relict aragonite within the calcite is recommended to correctly interpret the chemical data.

In some cases, the transformation process preserves, at least partially, the fabric of the precursor aragonite. It is common to observe “transformed” aragonite speleothems characterized by an arrangement of mosaics of calcite crystals outlining columnar aragonite crystals with quadratic terminations (see Kendall & Broughton, 1978). Columnar aragonite crystals are pseudo-hexagonal individuals formed by twinning, and commonly radiate from the substrate forming Rays (Folk & Assereto, 1976; Frisia et al., 2002; Frisia & Borsato, 2010). Replacement has been observed to occur preferentially at crystal defects (Frisia & Wenk, 1985), such as twin planes, and, in this case, transformation results in the partial preservation of the aragonite speleothem fabric (Fig. 4A). Aragonite in speleothems commonly shows acicular or elongated crystals (Railsback et al., 1997; Railsback et al., 2011; Wassenburg et al., 2012), and these morphologies may still be visible after transformation into calcite as illustrated in Fig. 4C and D.

Mimetic replacement is a possible candidate to explain occurrence of highly elongated columnar calcite crystals, very similar to acicular aragonite crystals, as the product of transformation. The case illustrated in Fig. 4E is that of an ancient flowstone, whose age is ca. 120,000 years (unpublished data, analyses performed by John Hellstrom), which formed in Collalto Cave in NE Italy cut in dolomite rock. The age implies the possibility that the flowstone underwent dissolution and re-precipitation when the cave was invaded by glacial meltwater during the deglaciation. Because the host rock is dolomite, it is reasonable to assume that the flowstone may have originally consisted of aragonite. However, petrographic observations

of crystal tips show most common rhombohedra termination, rather than square terminations. In addition, the highly elongated columnar crystals and “needles” consist of single individuals, not of a mosaic of calcite microsparite as would be expected as a product of mimetic transformation. Finally, the crystals do not show any clear evidence, such as micro-inclusions, of the presence of aragonite. Therefore, the fabric illustrated in Fig. 4E is most probably primary, not diagenetic, or should be considered primary in the microstratigraphic log.

It is here suggested to consider the presence of aragonite relicts in speleothem calcite “mimicking” aragonite as the unequivocal evidence for diagenetic transformation. Relicts of former primary or secondary aragonite are less frequent when aragonite is replaced by columnar calcite rather than by mosaic calcite (Perrin et al., 2014), yet relicts of aragonite still attest to the diagenetic origin of columnar calcite encasing them. Samples similar to that illustrated in Fig. 4E consist exclusively of calcite as documented by XRD analyses, whereby an aragonite precursor should not be hypothesized on the basis of morphologic similarity.

To summarize, the presence of acicular fabric or of very elongated columnar individuals is not, by itself, indicative of aragonite transformation. Not even the presence of square terminations is unequivocal. Thus, if in doubt, XRD analyses and Transmission Electron Microscopy observations remain the best options to investigate diagenetic transformations in speleothem fabrics (Frisia et al., 2002; Wassenburg et al., 2012).

CODES AND MICROSTRATIGRAPHIC LOGGING

The microstratigraphic log is constructed by ascribing a progressive code, which is the same as a progressive number, to fabrics observed in thin section, in PPL and XPL, at the optical microscope. For each fabric, an acronym, or symbol, is provided in Table 1 as well as the hierarchic code. The construction of the microstratigraphic log in the basis for the fabric time series, and relies on a logic progression in the coding, from fabrics which are related to the most stable hydrological conditions to those which formed through diagenetic processes. Fabric recognition and coding is a subjective operation, because it relies on a human operator, but, if based on accepted models for fabric development as proposed in Section 2, it has the potential to become a common tool shared by speleothem-based palaeoclimate researchers.

The coding of fabrics proposed here to produce follows that already used by McDermott et al. (1999) and ascribes the lowest numbers to primary fabrics whose accepted model of formation implies relatively constant drips (hydrologic stability), and parent waters characterized by relatively low supersaturation state with respect to calcite and with low concentration of inorganic and/or organic “additives” to the solution. These are the columnar proper (C) and Columnar open (Co) fabrics (Table 1). The highest numbers should then be ascribed to the diagenetic fabrics, such as microsparite (Ms) and mosaic calcite (Mc), because

these indicate opening of the system and re-setting of the original climate signals. The assignment of a code between “end members” then takes into account the whole spectrum of hydrological instability, changes in supersaturation state, input of impurities and bio-influenced precipitation that may be recorded in speleothems through the development of different fabrics. Obviously, if only one fabric constitutes the whole speleothem, then the microstratigraphic logging may have little significance. But when at least two primary fabrics are present, logging is useful, as it may highlight changes in degassing or drip-rate worth mentioning for the correct interpretation of the paleodata (cf. Boch et al., 2011).

The very first step to construct a microstratigraphic log is to obtain a continuous sequence of fabrics. This can be done by cutting the top and bottom of each section at an angle as shown in Fig. 5. Thus, the top of the lower section overlaps with the bottom of the upper section even if there is a minimum loss of material due to sawing. Thin sections should be 30 µm thick, polished and uncoated to allow for successive analyses in epifluorescence, or backscattered electron microscopy, or by LA-ICP-MS. Transmitted light microscopy observations should be carried out at a research microscope with high quality optics, equipped with: a rotating table, an x-y moving stage with micrometer steps, at least 3 objectives (2x, 10x, 40x), analyser and polarizer lenses, a condenser and a camera attached to a PC. If the x-y stage is not available, it is recommended to mark on a clear image of the whole sample (for example a scan at high resolution) specific bounding surfaces (“markers”). Alternatively, one could mark directly on the thin sections increments of 1 or 5 mm from top to bottom (Fig. 5). The operator should bear in mind that it is essential to keep precise track of distances from the top where changes in fabric occur.

To build the microstratigraphic log in a electronic spreadsheet one column is labelled the “distance from top” and a second column “fabric code”, following the progression in coding given in Table 1. It is recommended to perform a first rapid check of the fabrics present in the thin sections before starting the actual microstratigraphic logging. In this first screening, the fabrics are recognized and organized hierarchically in a legend, where each code is assigned to its fabric acronym (Table 1). Then, sequential cells in the “distance from top” column report the spatial increments in mm where a specific fabric is observed,

and the cells in the “fabric code” column report the code ascribed to the fabric.

In Fig. 6 a large thin section is illustrated with its microstratigraphic log overlain to show the correspondence between codes and actual fabrics. From Fig. 6 it becomes evident that coding is a subjective operation, because each observer may privilege one fabric over another where subtle differences are present. Yet, in the thin section shown in Fig. 6, the most important information is the occurrence of compact, open and elongated columnar calcite, and of micrite layers. According to the models of development for each of these fabrics, the stalagmite portion logged in Fig. 6 suggests changes in hydrology through time from constant (compact) to variable (open), with periods when particulate and foreign ions are transported by dripwater to the surface of the speleothem (elongated). The presence of micrite indicates that the system experienced periods of reduced flow, when microbial films could have colonized the speleothem surface. Critically, the fabrics in the speleothem illustrated in Fig. 6 have been found to record rainfall amount variability, and support the $\delta^{18}\text{O}$ interpretation in terms of monsoon intensity changes in the Holocene (Griffiths et al., 2009).

The selection of the spatial resolution of the microstratigraphic log should be optimized for the research question. The highest resolution used to date in a microstratigraphic log was 20 to 50 µm, which was the resolution required to reconstruct hydrological variability in the Younger Dryas from a speleothem portion characterized by very slow growth rate (cf. Belli et al., 2013).

The progression in the coding system proposed in Fig. 6, follows the models of fabric development discussed in section 2 and summarized in Table 1, where 12 calcite fabrics are listed with their respective acronyms. Table 1 also supplies in the “Notes” column, acronyms that supplement the general information encoded in the number.

In any single speleothem, commonly only a few fabrics (three or four) are present. Thus, once the codes are replaced by the acronym for each fabric, the microstratigraphic log will appear as a relatively simple curve that shifts from the fabrics to the left hand side (lower code numbers), which identify relatively “regular” flow of water at low supersaturation and transporting few impurities, to fabrics on the right hand side of the graph, which mark dry events and/or diagenetic processes (higher values).

In the example shown in Fig. 7, there are four columnar calcite types present, and the order is: C, Co, Ce and Cfo. This progressive order follows the criteria provided in section 1, where the lowest code of C indicates development of the stalagmite from waters at low supersaturation state for calcite and little Mg or organic molecules

Fig. 5. Suggested sampling strategy to obtain a continuous thin section microstratigraphic sequence. Each section is labelled progressively from the top. A scanned picture of the whole specimen with the outline of the thin section sequence is handy at the microscope to keep constant track of the stratigraphy. It is recommended to mark major features, such as hiatuses, and mark also the thin section outlines each cm. Thus, even if the stage of the microscope is not equipped with an x-y microstage system, it is possible to keep track of the distance from top for each passage of fabric observed in thin sections.

Fig. 6. Microstratigraphic log of a thin section from a stalagmite predominantly consisting of columnar, columnar open and columnar elongated fabrics. A thick micrite layer occurs at 2mm from top. Thinner micrite layers are outlined in the log. Columnar fabric showing rhombohedra terminations is also highlighted by the acronym Crt, but not reported as different fabric in the log (see text for details).

as “additives”, and the highest code for Cfo indicates higher supersaturation and most likely the relatively highest Mg/Ca ratio reached by the feeding drip waters throughout the formation of the specimen. There are other two fabrics, micrite and microsparite, which suggest, respectively, a prolonged period of dryness and aggrading neomorphism. The association of micrite and microsparite in the same layers testifies to the ripening of micrite when its conditions of formation changed, possibly through the recovery of flow. The rationale for constructing the microstratigraphic log of the sample is hierarchizing the fabrics according to the progression provided in Table 1. Then, in the “Fabric code” column of the spreadsheet, depths from top when fabric changes occur are reported. For example: from 0 to 1 mm from top the fabric code is 1, or C, from 1 to 2 mm from top the code is 8 (or M), from 2 to 3 mm from top the code is 9 (or Ms), from 3 to 4 mm from top the code is 8 (M), from 4 to 5 mm from top the code is 1 (or C), ... from 9 to 13 mm the code is 1 (or C) and so on and so forth. Once all the sequence of fabrics from top to bottom has been rendered in numerical form through the codes, and the log constructed, the

numbers are replaced by the fabric acronyms. It is, therefore, important to use the acronyms provided in Table 1 for the speleothem calcite fabrics, so that researchers can compare fabric datasets that use a common reference.

The issue of a common “reference” for transforming fabric changes in a curve becomes critical when fabrics are plotted against geochemical curves. In Fig. 7 it is clear that the shift of C isotope ratio values at ca. 25 cm from top occurs at a layer characterized by M, and the most positive C isotope ratio values in the series pertain to M and Ms. For example, if different speleothem records from the same region show coeval M layers associated with similar shifts to more positive $\delta^{13}\text{C}$ values, then it would be reasonable to infer that the occurrence of M reflects a regional history of hydrological stress when stalagmite tops became almost dry. The sole coeval shift to more positive $\delta^{13}\text{C}$ values without support of the microstratigraphic log would be more equivocal, as the shift could be ascribed to different processes, such as more intense degassing, a change in vegetation or prior calcite precipitation (cf. Fairchild & Baker, 2012).

If hiatuses are present, because they identify depositional gaps, it is here suggested not to code them. Rather, features characterizing the hiatus can be provided as notes or a red line mark a fabric boundary believed to be a hiatus.

BENEFITS OF THE FABRIC LOG IN SPELEOTHEM RESEARCH

The construction of microstratigraphic log of fabrics in speleothems allows for the recognition of changes through time (vertical dimension) of depositional models (or environments of deposition). Changes in the succession of fabrics are here given a simple, graphic form, which allows direct comparison with the series of geochemical data (Fig. 7). In the conceptual framework proposed here, the hierarchy of fabrics is operated through criteria of: a) progressive “hydrological stress”; b) progressive increase in supersaturation state of the dripwater; c) progressive increase in Mg and impurities; d) diagenesis. (Fig. 7)

In the microstratigraphic log a shift to highest fabric codes from bottom to top, then, suggests that a regime of relatively constant drip rate, although with some variability (Co) was punctuated by periods of lower discharge, when dripwaters were characterized by higher Mg concentration (Cfo), up to a point when the supersaturation of the water was very high, or a microbial coating formed (M). The development of Ce following C or Co, according to the proposed model of development, signifies that the supersaturation state and the Mg/Ca ratio of the parent water changes, but not necessarily the drip rate, which should have been relatively constant.

In Fig. 8, the coded fabrics are plotted against their relative stable isotope values to produce the *IsoFab* plot (Isotopes + Fabrics). All columnar fabric types seem to have similar $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values, which are more negative with respect to the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of micrite and microsparite. The good correlation

Fig. 7. Microstratigraphic log (left) of the stalagmitic flowstone from Cave 370 in the Nullarbor illustrated in Fig. 1D and E. The log was constructed measuring fabric changes in the sequence of three thin sections shown in the middle (PPL and XPL). To the right, C and O stable isotope ratios profiles are also shown. The microstratigraphic log indicates that layers consisting of micrite (M) and microsparite (Ms) are characterized more positive $\delta^{13}\text{C}$ values. These layers have a stromatolite-like appearance (see text for details) and have been ascribed to relatively dry periods, which would explain the more positive C isotope ratio values. The methods of coding allows for an easy correlation between speleothem fabrics and chemical properties as shown in Fig. 8.

($r^2 = 0.97$) in the inset of Fig. 8, shows that there is progressive kinetic modification from columnar elongated, columnar open, columnar compact, fascicular optic, micrite and, finally, microsparite. The observed correlation is quite intriguing, as it highlights that Ce has the most negative $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values.

Fig. 8. The *IsoFab* plot compares fabrics (here the fabrics coded in Fig. 7 are reported) and their corresponding mean stable isotope ratios values (plotted with their standard deviation). The correlation plot in the inset shows that columnar fabrics (C, Ce, Cfo, Co) have similar $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ signatures, with Ce characterized by the most depleted values. Micrite (M) and microsparite (Ms) are enriched in both ^{13}C and ^{18}O . The regression line shows that there is progressive kinetic modification of the isotope signals from columnar calcite to microsparite, possibly due to combined degassing and evaporation.

The content of Mg of Ce calcite in the specimen is up to 20,000 ppm (see Fig. 1E), thus, in the analysed specimen, the high concentration of Mg has no (or little) kinetic effect on stable isotope fractionation. The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values more negative than in C and Co suggest that the drip rate may have been higher during the deposition of the elongated crystals, which opens questions about the provenance of Mg (associated with particulate in addition to incorporation in the lattice) and the significance of the $\delta^{13}\text{C}$ values. The Cainozoic rock hosting the aquifer that fed the speleothem illustrated in Fig. 7 has $\delta^{13}\text{C}$ values as negative as ca. -12‰ (Miller et al., 2012), which hints at the possibility that Ce recorded wetter periods characterized by the efficient (rapid?) dissolution of Mg-rich clasts in the host rock (Frisia et al., 2012).

In synthesis, hierarchizing fabrics, microstratigraphic logging and the construction of a *IsoFab* plot provide information about potential disequilibrium precipitation, with all the implications for stable isotope fractionation and/or trace element partitioning. For example, the *IsoFab* plot for the sample illustrated in Fig. 7 highlighted that C and Co fabrics may be affected by some degassing and evaporation relative to Ce, which modified the C and O isotope ratios.

There is, thus, the potential to use the *IsoFab* plot in conjunction with a correlation between coded fabrics and stable isotope ratio values to act as test for “equilibrium” incorporation similar

to the Hendy test (Hendy, 1971), with the difference that the test proposed here operates on “bulk” fabrics, rather than on the co-variance (or lack of it) of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values along single growth layers.

In addition, the *IsoFab* plot undoubtedly provides an immediate visualization of those changes in stable isotope ratio values that may be related to diagenetic changes, and in particular aggradation, with considerable improvement in palaeoclimate reconstruction from geochemical proxies.

In conclusion, it is here demonstrated that the construction of speleothem microstratigraphic log and of the combined Isotope-Fabric cross correlation diagram (*IsoFab* plot) are indeed necessary tools to provide robustness to the interpretation of the geochemical data. In Appendix 1 a schematic representation of each fabric, name, acronym, code, characteristics and significance are provided to facilitate both the construction of the microstratigraphic log, and the interpretation of fabrics in terms of environment.

ACKNOWLEDGEMENTS

The Author wishes to thank B. Railsback, for his constructive criticism and review, and the suggestion to produce a summary figure to be “hung on petrographers’ wall”. Russell Drysdale and Petra Bajo provided access to thin sections from Antro del Corchia. Russell provided the Tommy Graham and the Liang Luar thin sections, as well as help with mass spectrometry results.

Nullarbor samples were studied within the framework of ARC Discovery Project DP0985214. Bence Paul performed the Mg map of the Nullarbor speleothem from cave 370 with much appreciated precision and skill. Rebeca Martín-García put together the micrographs for Fig. 8. Thanks to Giovanni Gattolin who introduced me to the fascinating issue of the genesis of clotted peloidal micrite and to microbialite fabrics. Kathleen Johnson agreed to show the samples from Tham Doun May, and allowed me to use the SEM micrograph in Fig. 2D.

Christoph Spötl and the project “Speläotheme der Conturines Höle: Ein potentiell einmaliges Paläoumwelt-Archiv in den Dolomiten” are acknowledged for access to speleothems from Conturines cave.

Last, but not least, my profound thank goes to Andrea Borsato, who always supports me, through light and darkness. He also helped with editing the figures.

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Appendix 1

Type	Symbol (code)	Characteristics	Environment of formation
C (1)	I/w ratio <6:1; competitive growth at interfaces; straight to serrated boundaries; uniform extinction; common "flat" terminations or protruding rhombohedra terminations (~ 2µm high)	Relatively slow and constant drip; Slcc <0.35; Mg/Ca <0.3; pH up to 8.4; low impurity content
Co (2)	I/w ratio <6:1; competitive growth at interfaces; incomplete coalescence of crystals; high intercrystalline porosity, commonly linear; uniform extinction.	Drip rate > than in C; Slcc up to 0.35; Mg/Ca <0.3; pH 7.5 up to 8.
Ce (3)	I/w ratio > 6:1; competitive growth at interfaces; preferential growth of acute rhombohedron; incomplete coalescence of crystals; protruding terminations common; uniform extinction. May show lateral overgrowths, in particular in the presence of impurity-rich layers.	Drip rate constant; Slcc 0.1 to 0.4; Mg/Ca > 0.3. Ce ₀ : relatively fast flow; presence of particulate. Diagenesis?
Ce ₀ (4)
Cfo (5)	Polycrystals I/w ratio > 6:1; undulatory extinction diverge away from substrate when rotating table is turned CCW; split crystal growth (spherulitic type); downward concave curvature.	Low drip rate or laminar flow; Slcc 0.5; Mg/Ca > 1.5; typical in stalagmites & flowstones formed in caves cut in dolomite
Cr _f (5.5)	Polycrystals I/w ratio > 6:1; undulatory extinction converge away from substrate when rotating table turned CW; split crystal growth; upward concave curvature.
Cm (6)	Polycrystals I/w < 6:1; irregular intercrystalline boundaries; uniform extinction with "patches" due to cross-cutting by adjacent crystals; high intracrystalline microporosity; typical of laminated speleothems.	Variable drip rate (seasonal); Slcc up to 0.35; Mg/Ca < 0.3; drip/flow carrying colloidal particles.
D (7)	Branching polycrystals; scaffold-like appearance "warp and waft"; irregular intercrystalline boundaries; high intercrystalline porosity.	Variable drip rate; Slcc up to 0.4. Mg/Ca possibly similar to Cm; strong degassing; presence of particulate/ foreign ions. Bio-influenced precipitation
M (8)	Crystals < 2µm; stromatolitic-like structure; clotted structure. Common geometric selection above micrite layers.	Bio-influenced. Low flow/dry. Condensation/corrosion?
Ms (9)	I/w ratio ~ 1:1; crystal size > 2µm < 30µm; commonly associated with micrite. Fabric-destructive replacement.	Diagenesis. Aggrading neomorphism (micrite to microsparite)
Ms _{ma} (10)	I/w ratio ~ 1:1; Crystal size > 2µm < 30µm; retention of aragonite fabric.	Diagenesis. Mimetic replacement
Mc (11)	I/w ratio ~ 1:1; crystal size > 30µm. Fabric destructive.	Diagenesis. If replacing calcite, no relicts of a former unstable phase are visible.
Mc _{an} (12)	I/w ratio ~ 1:1; Crystal size > 30µm; Fabric destructive; preserves relicts of aragonite, commonly needles.	Diagenesis. Commonly related to the transformation of speleothem aragonite into calcite

Appendix 1. Summary of calcite speleothem fabrics, their symbols, codes, characteristics and known environmental parameters underpinning their development.