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ORIGINAL ARTICLE

Speleothems in gypsum caves and their paleoclimatological significance

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Abstract This article highlights the relationship between speleothems growing inside gypsum caves and the particular climate that existed during their development. Speleothems in gypsum caves normally consist of calcium carbonate (calcite) or calcium sulphate (gypsum) and the abundance of such deposits greatly differs from zone to zone. Observations carried out over the last 20 years in gypsum caves subjected to very different climates (Italy, Spain, New Mexico, northern Russia, Cuba, Argentina) highlight wide variation in their cave deposits. In arid or semi-arid climates, the speleothems are mainly composed of gypsum, whilst in temperate, humid or tropical regions, carbonate formations are largely predominant. In polar zones no speleothems develop. These mineralogical details could be useful paleoclimatic indicators of climate change. The interpretation proposed is based on the fact that in gypsum karst the kind of speleothems deposited is determined by competition between the two principal mechanisms that cause precipitation of calcite and gypsum. These mechanisms are completely different: calcite speleothem evolution is mainly controlled by CO_2 diffusion, while gypsum deposits develop mostly due to evaporation. Therefore, the prevalence of one kind of speleothem over the other, and the relationship between the solution–precipitation processes of calcite and gypsum, may provide evidence of a specific paleoclimate. Additionally, other non-common deposits in gypsum caves like moonmilk, cave rafts and dolomite speleothems can be used as markers for the prevalence of long, dry periods in humid areas, seasonal changes in climate, or rainfall trends in some gypsum areas. Moreover, the dating of gypsum speleothems could contribute paleoclimatic data relating to dry periods when calcite speleothems are not deposited. In contrast, the dating of calcite speleothems in gypsum caves could identify former wet periods in arid zones.

Keywords Gypsum · Caves · Speleothems · Paleoclimatology

Introduction and methods

Over the last 20 years, the importance of speleothems as a tool for establishing the recent paleoclimatic record, from the Pleistocene to the present day, has been confirmed by many authors. Accordingly, some speleothems represent a useful paleoclimatic proxy—particularly stalagmites and flowstones, which frequently allow detailed chronology of the morphodynamic, environmental and climatic events of a given karstic area to be investigated (Ford 1997; Baker et al. 1999; Holmgren et al. 1999; Lauritzen and Lundberg 1999; Lauritzen and Onac 1999; Linge et al. 2001; Repisnki et al. 1999; Proctor et al. 2000). Climatic conditions during the formation of speleothems (i.e. temperature, humidity, precipitation, soil conditions, CO_2 production and organic matter content) are intrinsically linked to the growing conditions and mineralogy of the speleothems themselves (Denniston

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et al. 2000; Charman et al. 2001; Zhang et al. 2006). In particular, many studies that analyse growth layers have attempted to identify wet and dry climatic periods (Baker et al. 1997; McGarry and Baker 2000; Genty et al. 2001) as well as the mineralogy of the speleothems, especially Mg and other trace elements (Roberts et al. 1998; Fairchild et al. 2000; Huang et al. 2001). The processes of condensation corrosion can also be detected in speleothem layers, thus providing information about CO_2-carbonate equilibria related to external factors like vegetation and/or soil cover. This has direct implications for identifying the paleoclimatology of a region (BarMatthews et al. 1996).

There is an extended literature dealing with this subject, although it is limited to karstic areas with carbonate lithologies—essentially kartified limestone and dolostone. Until now, virtually no scientific studies have been published concerning possible climatic implications of speleothems growing inside gypsum karst caves rather than carbonate ones (Calaforra et al. 1992; Calaforra and Forti 1999). The aim of the present study is to identify the interrelationships between the various types of speleothems within gypsum caves, and their relationship to the climate that prevailed during their development. Methods in this study were based on both empirical and analytical reasoning. Morphological observations made over the last 20 years by the authors in various gypsiferous areas of the world with very dissimilar climates (Italy, Spain, New Mexico, Russia, Argentina and Cuba) have highlighted the differences between the various chemical deposits which occur today in each of these locations. Additionally, mineralogical analysis (X-ray, Microsond and SEM) was performed to determine the composition of some speleothems whose composition is difficult to evaluate in detail by in situ methods. The analysis of cave raft, moonmilk and hollow stalagmites represent the only three cases where the speleothems were sampled and removed from the cave.

Results and discussion

In caves that have developed in carbonate rocks, 99% of the speleothems are formed of calcite or aragonite (Hill and Forti 1997). In contrast, in gypsum caves the speleothems are just as frequently formed of calcite as of gypsum (Forti 1997). The relative abundance of each of these minerals varies enormously from one region to another. The influence of climate on speleothem typology as well as the various chemical mechanisms of deposition of gypsiferous and carbonate concretions within gypsum caves need to be considered to explain these differences.

Chemical processes influencing the precipitation of calcite and gypsum in gypsum caves

The fact that, on occasions, gypsum concretions can be found in gypsum caves is logical. These speleothems are formed through the evaporation of water, saturated in calcium sulphate, which has infiltrated into the cave environment:

$$CaSO_4 \cdot 2H_2O + H_2O \leftrightarrow Ca^{2+} + SO_4^{2-} + 3H_2O$$

In contrast, the existence of large deposits of calcium carbonate in the interior of gypsum caves could strike one as rather puzzling. The common explanation for the precipitation of calcite speleothems in gypsum caves refers to the presence of intercalated carbonate of marly strata, which leads to saturation in calcite in the infiltration water. However, since the 1980s the possibility has been recognised that these speleothems could develop in a gypsiferous environment as the sole result of the presence of dissolved carbon dioxide in the infiltration water (Forti and Rabbi 1981). Since this time, other publications have detailed the process of carbonate precipitation with saturation in gypsum, basically linking it to the phenomenon of incongruent dissolution of gypsum (Calaforra 1998; Forti 1997):

$$CaSO_4 \cdot 2H_2O + H_2O + CO_2 \leftrightarrow CaCO_3 + SO_4^{2-} + 2H^+ + 2H_2O$$

In this sense, it is essential to note that the processes of formation leading to either calcite or gypsum precipitation in gypsum caves are completely different from each other. Evaporation is a physical phenomenon controlled principally by temperature and relative air humidity, whereas incongruent dissolution of gypsum and precipitation of calcite is a chemical phenomenon controlled by the partial pressure of dissolved carbon dioxide, which in turn depends on biological activity and soil water residence time.

Figure 1 graphically shows such an idea in outline. Nowadays, in gypsum caves evolving in a dry climate, where precipitation is very scarce, it is usual to find a bare ground surface karst, with a thin soil and a low vegetation cover (Fig. 1a). In this situation, it is not possible for the percolation water to become rich in edaphic CO_2. This is why it is unlikely to precipitate carbonate minerals when the infiltration water reaches the cave atmosphere. If, in addition, we consider the extreme scarcity of water in this situation and the strong evaporation under these climates, it can deduced that the main speleothem deposits in gypsum cave under arid conditions will be mainly gypsiferous. In contrast, in a humid climate with abundant precipitation, it is logical that the soil and vegetation cover will be well developed (Fig. 1b). Under these conditions, contribution

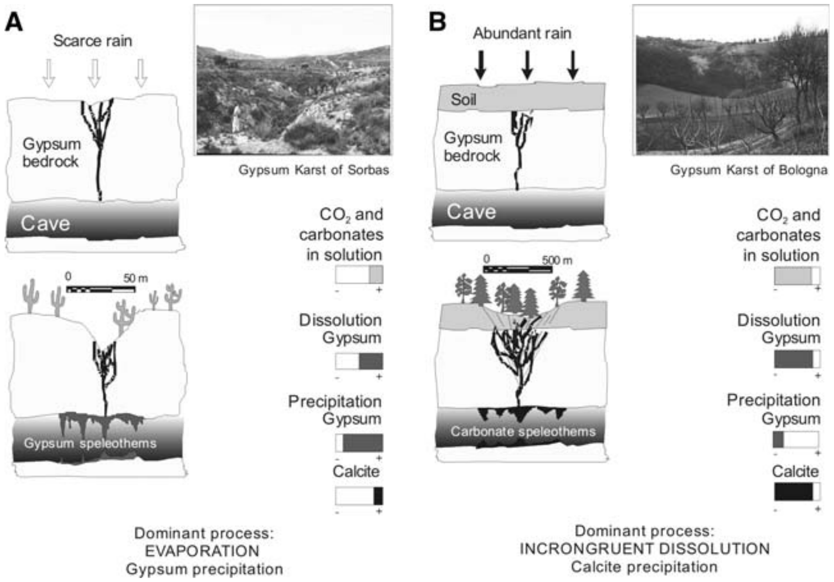


Fig. 1 **a** In arid or sub-arid areas, gypsum outcrops are largely devoid of vegetation and so the content of CO_2 in percolating waters is low. This situation means that the process of incongruent dissolution does not arise. The dominant process is therefore that of evaporation, with the consequent formation of mainly gypsum speleothems. **b** In wet, temperate or tropical areas, gypsum outcrops are covered by vegetation that guarantees an elevated presence of H_2CO_3 in the percolation water. On the other hand, the abundant rainfall means that evaporation is less efficient. In this situation the dominant process is that of incongruent dissolution, with the consequent formation of mainly carbonate speleothems

of CO_2 to the percolation water is much more important and is the reason why the process of incongruent dissolution of gypsum with calcium carbonate precipitation will take place inside the cave.

From a chemical point of view, the exclusive presence of either calcite or gypsum speleothems in the interior of a gypsum cave, or as is more commonly the case, the predominance of one type over the other, depends on the efficiency of each of the two deposition mechanisms (evaporation vs. incongruent dissolution). Accordingly, in areas characterised by arid or semiarid climates, speleothems in gypsum caves are found to consist basically of gypsum, whilst in wet temperate or tropical areas carbonate speleothems clearly dominate.

Recorded variations in speleothem mineralogy in gypsum caves

In addition to the brief discussion of the various mechanisms of concretion in gypsiferous caves, examples are given of speleothems which are in equilibrium with the current climate changes found within some gypsum caves. These speleothems afford direct (paleo) climatic information. In line with the idea that deposition of gypsum in gypsum caves is controlled by evaporation, gypsum speleothems will be more abundant in caves with scarce infiltration water, low CO_2 content, high relative temperature and quite low humidity. In fact, the greatest development of gypsum speleothems has been observed in the desert of New Mexico (USA) (Calaforra and Forti 1994), in the subdesert area of Sorbas (Spain) (Calaforra 1998) and in the gypsum areas of North Africa. In contrast, formation of calcite linked to incongruent dissolution of gypsum will occur when aqueous carbon dioxide content is high. Such a condition requires the presence of well-developed soil and vegetation cover overlying the karstic system. For this reason, the areas in which it would seem logical to expect significant development of carbonate speleothems are the gypsiferous karsts in humid climates, in which the temperature regime is not too extreme. As expected, the most extensive carbonate speleothems in gypsum caves are found in northern Italy and Cuba (Fagundo et al. 1993; Dalmonte and Forti 1995; Piancastelli and Forti 1997; Dalmonte et al. 2004).

Other extreme conditions of speleothem deposition in gypsum caves can be described using the example of the Pinega area in Russia (Forti 1990; Klimchouk et al. 1996), close to the Arctic Circle. These caves are characterised by the total absence of permanent concretions in their interior. Some speleothems can develop during the autumn, before the available water totally freezes up. Such deposits are ephemeral, and dissolve completely during the thaw at the end of spring. In addition, the hydrodynamic regime within the cave is such that during the brief period of late spring and summer, the caves undergo intense flushing and erosion, a feature that does not permit any kind of gypsum or carbonate concretion. The discovery of an upper gallery in one of the most important caves in the area has brought to light the existence of an old carbonate pavement several tens of centimetres thick. Currently, this is subject to erosion and corrosion, but it demonstrates unequivocally a past climate that allowed the precipitation of carbonates inside the cave. Finally, we can cite the karstic area of Sorbas (Spain), one of the most arid in Europe, whose caves are among the most important in the world for their abundance of gypsiferous speleothems (Calaforra and Forti 1990). The present-day carbonate speleothems are limited to very particular hydrological situations (Calaforra et al. 1992), while the fossil galleries encrusted with carbonate flowstone are currently being eroded or corroded away. The Sorbas calcite speleothems probably developed during a time of greater rainfall when vegetation cover was more abundant.

Table 1, based on observations made over the course of the last 20 years, shows a scheme of the relative relationships that exist, and the general characteristics of gypsiferous or carbonate speleothems within gypsum caves, for various climate zones over the world. The table summarizes what kind of speleothems are essentially found in gypsum caves according to the general climate of the area.

Detailed observation of some of the concretion sequences in gypsum caves in areas that are climatically distinct from each other has demonstrated how such caves constitute a record of climate change in that area. We will go on to describe some particular examples of such climatic variation.

Gypsum crystals over calcite

In Entella Cave, Sicily, large carbonate flowstones exist. These are currently inactive, as is demonstrated by an almost total covering by gypsum efflorescences (Fig. 2a), which in places exceed half a centimetre in thickness. The good state of conservation of these carbonate speleothems, which show no signs of erosion, corrosion, together with their fine, gypsiferous coating suggests that environmental change occurred relatively recently. In all probability, it may have been anthropogenic in nature, and could have taken place during the last century or a little before, when vegetation in the area was cleared, causing erosion of the soil cover overlying the gypsum, and so altering the hydrodynamics and the chemistry of the infiltration water.

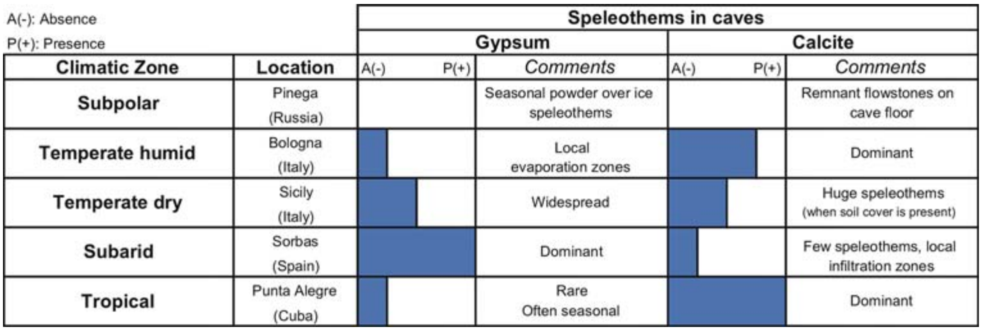
Dolomite in gypsum caves

During extreme dry periods it is possible to find some speleothems composed of dolomite. This is the case of the moonmilk found in the Spipola Cave in Italy (Forti et al. 2004), which was the first time this mineral formation had been described in a gypsum cave and the conditions were quite unusual for the precipitation of this mineral (10°C, cave atmosphere and low water salinity). It is likely that the mechanism of precipitation of the dolomite moonmilk is by direct evaporation of a magnesium-rich flow through the gypsum–marl strata, and the main factor that determined its development could be related to the long dry spell that occurred in this area at the beginning of 2002, prior to the discovery of this speleothem.

Gypsum hollow stalagmites

Other interesting speleothems that indicate the close interaction between the precipitation of calcite and gypsum in gypsum caves are the hollow stalagmites found in the gypsum karst of Sorbas (Spain) (Calaforra and Forti 1990). The most amazing feature of this speleothem is that the dripping stalactites above them are of calcite but the hollow stalagmites themselves are made of gypsum. The stalagmites are formed by the effect of common-ion interaction between gypsum and calcite (calcium):

Table 1 Summary of empirical observations of speleothems in different gypsum karsts of the world



A(-): Absence
P(+): Presence

		Speleothems in caves					
		Gypsum			Calcite		
Climatic Zone	Location	A(-)	P(+)	Comments	A(-)	P(+)	Comments
Subpolar	Pinega (Russia)			Seasonal powder over ice speleothems			Remnant flowstones on cave floor
Temperate humid	Bologna (Italy)	▨		Local evaporation zones	▨		Dominant
Temperate dry	Sicily (Italy)	▨		Widespread	▨		Huge speleothems (when soil cover is present)
Subarid	Sorbas (Spain)	▨	▨	Dominant	▨		Few speleothems, local infiltration zones
Tropical	Punta Alegre (Cuba)	▨		Rare Often seasonal	▨	▨	Dominant

The table shows, proportionally, the characteristics of speleothem deposits (gypsum or carbonate) inside gypsum caves for various climate zones

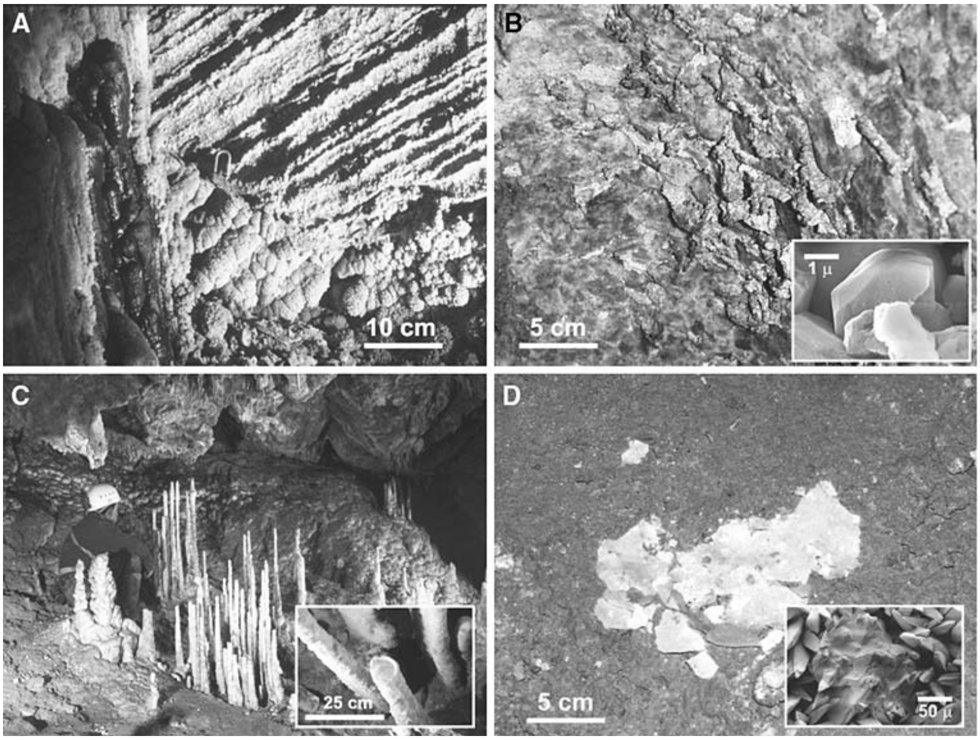


Fig. 2 Some examples of speleothems in gypsum caves that are strongly related to changes in climatic conditions. **a** Gypsum over calcite speleothems. In climates that are becoming drier, many of the carbonate formations are no longer developing and they are starting to be covered by gypsum deposits (Entella Cave, Sicily, Italy), **b** dolomite moonmilk in Spipola cave (Bologna, Italy) has been detected after a long period (90 days) without rainfall in the recharge area of the cave, **c** gypsum hollow stalagmites are a very complex speleothem that show the finely equilibrium between the precipitation of calcite (stalactites) and gypsum (the hollow stalagmites), **d** gypsum-calcite rafts in Spipola cave indicate dry periods when some lakes in the cave dried up and evaporation occurred with the simultaneous precipitation of gypsum over the calcite

$$2H_2O + 2CO_2 \leftrightarrow 2HCO_3^- + 2H^+$$

$$CaSO_4 \cdot 2H_2O + H_2O \leftrightarrow Ca^{2+} + SO_4^= + 3H_2O$$

$$2HCO_3^- + Ca^{2+} \leftrightarrow CaCO_3 + H_2O + CO_2$$

which allows subsaturation of the dripping water with respect to gypsum but its later precipitation by evaporation. Because this equilibrium is very complex, any change in climatic conditions can shift the balance of primacy in the precipitation of gypsum or calcite.

Gypsum/calcite cave rafts

This speleothem, recently found in Spipola Cave (Italy) (Forti 2003), is composed of a very thin lamina (<0.2 mm) of scalenohedral calcite (75%), with anhedral gypsum precipitated over the calcite without corrosion forms. This kind of deposit has been related to the action of two mechanisms: double exchange in the case of calcite, and oversaturation due to evaporation in the case of gypsum. The presence of this speleothem indicates dry periods when some lakes inside the cave dried up and evaporation occurred with the simultaneous precipitation of gypsum over the calcite.

These four examples -shown in Fig. 2 confirm how effectively climatic change can be registered in the speleothems of a gypsum cave. The sketch in Fig. 3 summarizes the expected climatic trends in the evolution of chemical deposits in gypsum caves: in general, an increase in temperature should enhance gypsum deposition, while an increase in rainfall would enhance calcite deposition.

Finally, it is necessary to point out that while some climatic variations give rise to modifications in the pattern of concretions that may be preserved and documented after a relatively long period of time, others may be preserved for a much shorter period and so are much less likely to be detected in the speleothem palaeoclimatic record. This is due to the fact that gypsum and calcite have completely contrasting physico-chemical properties. In particular, the solubility and erodibility of gypsum is much greater than calcite. Thus, it is much more probable that modification of concretions involving the fossilization of calcite and development of gypsum will be observed than will

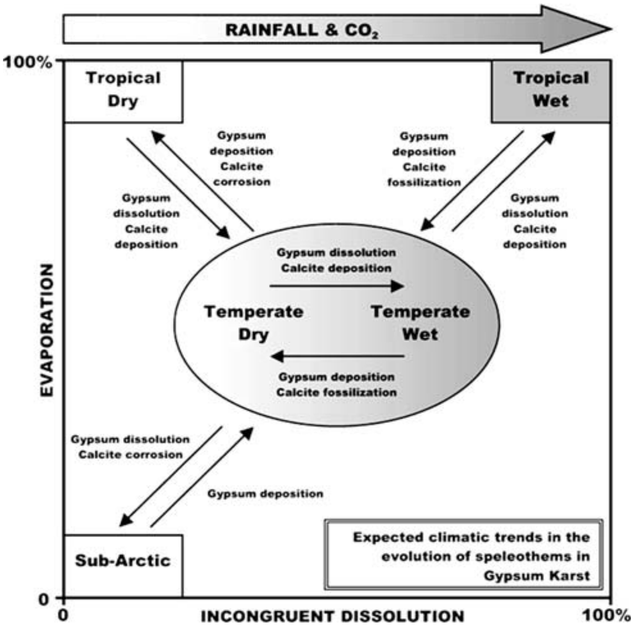


Fig. 3 General Scheme showing the variations induced in calcite/gypsum speleothems growing in gypsum caves as a function of rain, CO_2 content in the seeping water, evaporation and/or incongruent dissolution efficiency (after Forti 2004, modified)

modifications in the opposite direction. In other words, at least theoretically, the gypsum caves are more likely to retain a ‘‘memory’’ of climate changes moving from wet-temperate to arid/semi-arid than a change in the opposite direction.

Conclusions

The observed differences in active cave formations within gypsum caves of the world strongly support the idea that these chemical deposits may be considered as proxy records of climate and paleoclimate of a given gypsum area (Klimchouk et al. 1996; Forti and Hill 2004). In gypsum caves, climatic factors have been proved to have a stronger influence upon calcite and/or gypsum deposits than in carbonate karst: this is the consequence of the different depositional mechanisms active in gypsum karst (incongruent dissolution and/or CO_2 diffusion for calcite, and evaporation for gypsum), which are influenced in very different ways by climatic variables. This close relationship to climate gives rise to deposits in the gypsum environment that are potentially of very great importance for paleoclimatology (Forti 2004), as well as being indicators of contemporary climatic change. Therefore, sudden changes in chemical deposits of a particular cave (e.g. from predominantly calcite to predominantly gypsum or vice versa, or from either type of deposit to the absence of speleothems) should be considered proof that the climate has changed. Finally, it must be stressed that climatic effects on gypsum speleothems are expected to be much faster than those on calcite because gypsum is much more soluble and more prone to erosion than calcite.

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