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PALAEOCLIMATE AND PALAEOENVIRONMENTAL RECONSTRUCTIONS FROM SPELEOTHEMS IN ARTIFICIAL CAVES (LAZIO, ITALY)

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Abstract
Speleothems (stalactites, stalagmites and flowstones) in natural and artificial caves can be used as historical archives to reconstruct palaeoclimatic and palaeoenvironmental conditions in the past. This can be obtained through chemical investigation of the oxygen and carbon contained in the carbonate deposits, whose growth is related to the CO_2 dissolved in the dripping water. This gas may originate from the organic activity in the soil or, in areas affected by volcanic or hydrothermal activity, it may be of deep provenance. Specific investigation on the chemical and physical properties of speleothems, coupled to U-Th dating, allow to distinguish between the two origins. In case of CO_2 derived from the soil, the chemical record can give paleoclimatic and paleoenvironmental information (on temperature and precipitation), whereas deep origin CO_2 may be employed to reconstruct the seismic and volcanic evolution of a given area. The described activities developed from a pilot study focused on an old stone mine (Centroni cave) located at the south-eastern outskirts of Roma, where abundant speleothems have been depositing since 2,000 years ago, with evident periodic hiatuses in the precipitation process. Cyclic interruptions in the speleothems growth, along with an increase in deep CO_2 proportions, are correlated with the opening of fractures preceding earthquakes, historically documented in the city of Roma and surroundings. Further studies have been carried out on the Ancient Roman age artificial outlet of *Gabii* Lake that occupied the crater of Castiglione, not far from the town of Tivoli. A first exploration of the outlet revealed the presence of speleothems. They appeared to have only few growth unconformities. The chemical records do not indicate significant changes or trends that can be interpreted in terms of variation in the proportion of deep CO_2. The chemical signature can be explained more simply as due to small fluctuations related to the climatic and environmental conditions.
Keywords: paloeclimate, speleothems, Italy, isotope geochemistry.

Riassunto
In cavità naturali ed artificiali gli speleotemi (stalattiti, stalagmiti e colate) possono essere usati come archivi storici per ricostruire le condizioni paleoclimatiche e paleoambientali del passato. Questo può essere ottenuto mediante l'indagine chimica di ossigeno e carbonio contenuti nei depositi carbonatici, la cui crescita è legata alla CO_2 disciolta nella goccia d'acqua. Questi gas provengono dalla attività biologica nel suolo o, nelle aree interessate da attività vulcanica o idrotermale, possono avere provenienza profonda. Indagini specifiche sulle proprietà chimiche e fisiche degli speleotemi, accoppiati alla datazione U-Th, permettono di distinguere tra le due origini. In caso di CO_2 derivante dal suolo, il record chimico può dare informazioni paleoclimatiche e paleoambientali (temperatura e precipitazioni), mentre l'origine profonda di CO_2 può essere utilizzata per ricostruire l'evoluzione sismica e vulcanica di una data area. Le attività descritte, partite da uno studio pilota, si sono focalizzate su una vecchia miniera di pietra (Grotta di Centroni) situata a sud-est di Roma, dove gli abbondanti speleotemi sono stati depositati 2000 anni fa, con evidenti iati periodici nel processo di precipitazione. Le interruzioni cicliche nell'accrescimento degli speleotemi, con un aumento in proporzioni profonde di CO_2, sono correlate alle aperture di fratture causate da terremoti, storicamente documentati nella città di Roma e nei dintorni. Ulteriori studi sono stati effettuati nell'antico emissario artificiale del lago di Gabii (di età romana) che occupava il cratere di Castiglione, non lontano dalla città di Tivoli. Una prima esplorazione della presa ha rivelato la presenza di concrezioni che sembrano avere solo poche discordanze nell'accrescimento. I record chimici non indicano cambiamenti significativi o tendenze che possono essere interpretati in termini di variazione nella proporzione di CO_2 profonda. La firma chimica può essere spiegata più semplicemente dalle piccole fluttuazioni legate alle condizioni climatiche ed ambientali.
Parole chiave: *paleoclima, speleotemi, Italia, geochimica isotopica.*

Introduction

Speleothems in natural caves can be routinely used as geological archives to reconstruct palaeoclimatic and palaeoenvironmental conditions in the past (Dorale et al., 2010; Tuccimei et al., 2012). This goal is achieved using the geochemical properties of particular elements in carbonate deposits, such as oxygen, carbon, uranium and thorium. The analysis of these elements allows either the dating of carbonate precipitates or the definition of the geological setting where they formed. In particular, it is very important to investigate the origin of CO_2 dissolved in forming-speleothems dripping water. Carbon dioxide can originate from the organic activity in the soil or, in areas affected by volcanic or hydrothermal activity, can be of deep provenance. The release of deep gas may occur in correspondence with seismic events which cause changes in the subsoil permeability and fluid ascent.

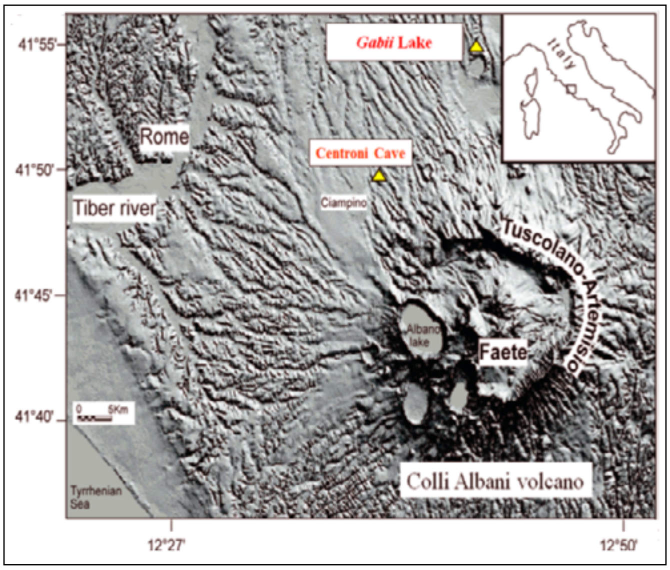

Fig. 1: location of investigated sites in Colli Albani area.
Fig. 1: ubicazione dei siti indagati nell'area dei Colli Albani.

In this study this approach is applied to artificial caves in the Colli Albani volcanic area (Latium, Italy): 1) Centroni cave, a Roman-age stone mine, and 2) the artificial outlet of former Gabii lake, corresponding to Castiglione volcano crater. In particular, the focus of this research is the reconstruction of the paleo-seismicity, the paleo-degassing history and the potential hazard of Colli Albani Volcano, in addition to the paleoclimatic conditions of the area in the last 2,000 years.

Study area

The Colli Albani (Fig. 1) is a quiescent volcano, which started its activity approximately 600,000 years ago. The volcano is made of the superposition of different edifices (Funiciello & Giordano, 2010). The oldest is a 1600 km^2 plateau with a central caldera, named Vulcano Laziale, and is mainly constituted by large volume ignimbrites (> 350,000 years; 10–100 km^3 in volume), to which the caldera is related. After the eruption of the last large volume ignimbrite (Villa Senni), approximately 350,000 years ago (Freda et al., 1997; Watkins et al., 2002), two complex edifices were built within the caldera area: the Tuscolano–Artemisio edifice and the Faete centre (Fig. 1) which infilled the caldera and presently reaches altitude of 949 m a.s.l.

The Tuscolano–Artemisio and the Faete edifices are partly interfingered and were emplaced between approximately 350,000 and 250,000 years ago (Radicati Di Brozolo et al., 1988; Voltaggio & Barbieri, 1995). Their products indicate a remarkable reduction of erupted volumes with respect to the Vulcano Laziale and a change in eruption style to effusive and mildly explosive.

The most recent and still active, although quiescent, period of activity of the volcano has been characterized by eccentric phreatomagmatic activity, which has formed several overlapping monogenetic and polygenetic crater lakes and tuff cones located along the western and northern slopes of the volcano. The products of this period of activity form the Via dei Laghi succession (Funiciello & Giordano, 2010). The older deposits have been dated at approximately 200,000 years (Marra et al., 2003). The most recent products, distributed around the Albano volcano, have been only recently discovered, described and dated to the Holocene (< 10,000 years ago). Localization of the volcanic centres is related to the presence at relatively shallow depth (below 1–2 km) of the aquifer hosted in the buried Ciampino carbonatic structure (Funiciello & Parotto, 1978). The intense diffuse CO_2 degassing of the area is most likely related to the interaction of the magmatic bodies with the carbonates at depth (Chiodini & Frondini, 2001).

Centroni Cave

A Roman-age stone mine (Grotta Centroni), consisting of a network of galleries opened about 2,000 years ago for the extraction of building materials for a residential house, is located SE of Roma, along the Anagnina state road. The stone mine, now used for mushroom cultivation, is situated within the "Tufo di Villa Senni" ignimbrite, the last caldera-forming ignimbrite erupted during the period of the Vulcano Laziale centre (Tuccimei et al., 2006). The roof of the stone mine is mostly made by the base of Villa Senni lava flow (Fornaseri et al., 1963) belonging to the Faete volcanic edifice. The Villa Senni lava flow is few metres thick above the mine and hosts a small aquifer containing water rich in calcium bicarbonate. Where the stone mine is covered by the lava flow, the waters circulating in the lava drip down from the roof, loosing CO_2 and depositing abundant speleothems (Fig. 2). Most of them are stalactites because the stone mine floor has been cleared and is now covered by the cases where mushrooms grow. Stalactites mantle almost completely the galleries roof and reach lengths of up to 10–15 cm and diameters of few centimetres. Several stalactites and a stalagmite have been collected in two different galleries and sectioned for studying their stratigraphy and selecting specimens for the analyses.

Artificial outlet of Gabii Lake

Gabii was an ancient city of Latium, located 18 km east of Roma along the Prenestina road, which was in early times known as the *Via Gabina*. It was on the south-eastern perimeter of an extinct volcanic crater lake, approximately circular in shape, named the *Lacus Gabinus*, and then during later times called Castiglione Lake. The water of the lake has been lowered by an artificial gallery by Romans, and the draining of the lake was continued and completed by Borghese family in the XVII century. At present, the former lake is entirely agricultural land. A first exploration of the outlet revealed the presence of speleothems that were successively sampled, sectioned and selected for geochemical investigation.

Methods

Speleothems are powerful paleoclimatic archives, as they contain a multitude of environment-dependent tracers (e.g. oxygen and carbon isotopic composition, trace elements, humic acids). They have been established as one of the best terrestrial archives of past climate because they can be accurately and precisely dated by the U-Th method (Kaufman & Broecker, 1965). Speleothems from Centroni cave and the outlet of Gabii lake have been analysed for carbon and oxygen composition and dated using U-Th method in order to reconstruct the paleoenvironmental conditions and the chronological framework.

Stable isotope geochemistry

Speleothems (stalagmites, stalactites, flowstones etc.) grow in caves as the result of precipitation of calcium carbonate from drip waters that enter the cave's atmosphere. The ratios of the stable isotopes oxygen and carbon of the carbonate ($CaCO_3$) are analyzed as they depend on environmental factors, such as rainfall amount, temperature, vegetation cover and type, etc.

Stable isotopes are chemical elements characterized by the same number of protons, but a different number of neutrons and thus a different mass. In particular, carbon has two main isotopes: ^{12}C (98.9 % abundance) and ^{13}C (1.1 % abundance), and oxygen consists of three isotopes: ^{16}O (99.762 % abundance), ^{17}O (0.038 % abundance) and ^{18}O (0.200 % abundance). The ratio between ^{13}C and ^{12}C of a speleothem is referred to that of a standard material (a fossil of *Belemnitella* in Pee Dee formation, North Carolina, USA) and expressed as $\delta^{13}C$ ‰. The ratio between ^{18}O and ^{16}O of a sample is normalized to the same standard and expressed as $\delta^{18}O$‰.

Stable isotopes ($\delta^{18}O$ and $\delta^{13}C$) are known as powerful atmospheric and hydrologic tracers (Mcdermott, 2004). Oxygen isotope ratios from stalagmites can provide information on the temperature, moisture source, and rainfall amount to name only a few. Stalagmite carbon isotope ratios, although often more difficult to interpret, may provide information on vegetation changes, the origin of CO_2, either from the soil or from depth.

U-Th dating

In addition to stable isotopes, radioactive elements occur in the environment. They are atoms characterized by an excess of energy in the nucleus (parents) that tend to transform (decay) in other elements with lower energy content (daughters). The rate of decay is a function of time and allows dating of a material.

The U-Th method is the most widely used dating technique applied to speleothems and is based on the extreme fractionation of the parent isotopes ^{238}U and ^{234}U from their long-lived daughter ^{230}Th in the hydrosphere. Uranium, markedly more soluble than Th in the surface and near-surface environments, is readily mobilised, whereas Th is adsorbed on detrital particles. Uranium is co-precipitated with $CaCO_3$ on release of CO_2, while Th is generally negligible. In the absence of detrital Th, ^{230}Th only forms in situ by radioactive decay of co-precipitated U. In a closed system the value of $^{230}Th/^{234}U$ activity ratio is a function of time, and allows the sample dating (Kaufman & Broecker, 1965).

Fig. 2: the roof of Centroni cave decorated with abundant actively growing stalactites (after Tuccimei et al., 2006).

Fig. 2: la volta della grotta di Centroni decorata con abbondanti stalattiti in crescita attiva (da Tuccimei et al., 2006).

If a speleothem is not pure calcium carbonate, but contains detrital particles, it is necessary to correct for it in order to obtain a more reliable age. Different models of corrections are available.

Sampling

Several speleothems have been collected in the Centroni cave and also in the outlet of Gabii lake, and sectioned for studying their stratigraphy and selecting specimens for the analyses.

Centroni Cave

The speleothems have been cut transversely relative to the growth axis using a diamond saw and the obtained sections have been accurately examined (Fig. 3). The most relevant features were the unconformities separating the growth layers of all the speleothems, suggesting that there have been periodical interruptions of the deposition process during the last 2,000 years, the age of the stone mine quoted from archeological evidences. The possible explanation of that, besides a strong periodic reduction of the rainfall amount, not recorded in any published pollen or isotopic record from Rome and Central Italy (Alessio et al., 1986; Calderoni et al., 1994; Ricci Lucchi et al., 2000; Tuccimei et al., 2005), could be the periodic groundwater acidification due to the increase of dissolved CO_2 in a region of diffuse and localised degassing processes. Microscope studies have shown that the speleothems are entirely calcite. The hiatuses do not show evident signs of selective dissolution.

Artificial outlet of Gabii Lake

Several stalagmites have been collected in the Gabii lake outlet. They appeared to have only few growth unconformities (Fig. 4) if compared with speleothems from the Centroni site. A second difference with the latter seemed to be represented by a higher proportion of detrital minerals. Microscope studies have shown that the speleothems are mainly calcite with minor clay minerals.

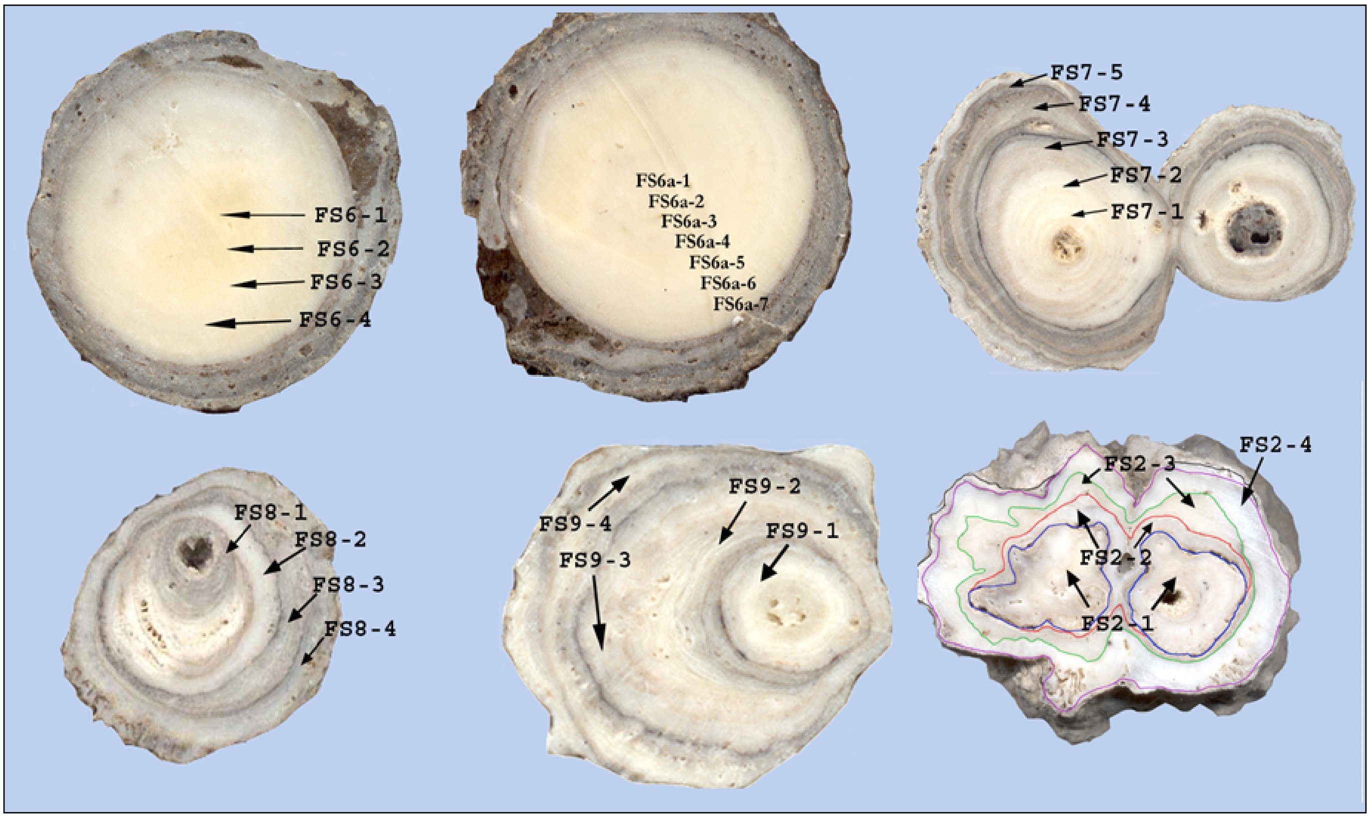

Fig. 3: transverse sections of stalagmite FS6 (two mirror slices) and stalactites FS2, FS7, FS8 and FS9 with location of studied samples. All of them have been analysed for the O and C isotopic composition and some speleothems have been also dated (after Tuccimei et al., 2006).

Fig. 3: sezioni trasversali della stalagmite FS6 (due sezioni speculari) e stalattiti FS2, FS7, FS8 e FS9 con posizione dei campioni studiati. Per tutti è stata analizzata la composizione isotopica O e C e alcuni speleotemi sono stati anche datati (after Tuccimei et al., 2006).

Results and discussion

Centroni Cave

Based on U-Th dating (Tuccimei et al., 2006), ten cycles of deposition have been identified from 90-110 A.D. to 1798-1820 A.D., some of which are recognised across different speleothems (Tab. 1). The age gap dividing different growth layers is in the order of one to few hundred years giving a temporal span for periodic interruption of speleothems deposition. These are probably due to episodic release of deep fluids from the geothermal-hydrothermal system that uprises and acidifies drip water. No speleothem growth has been documented in the period 1350-1800 A.D. at the Centroni site.

$\delta^{13}C$ ‰ and $\delta^{18}O$ ‰ of speleothems generally becomes more positive within a single growth layer, showing an increase of deep CO_2 (Tuccimei et al., 2006) This consideration depends on the strongly different composition distinguishing CO_2 in soil (more negative) and CO_2 from deep aquifer (more positive). Using these data, it is possible to assess that the input of deep fluids, nearly absent at the beginning of the deposition, reached values of about 20–30% approaching the depositional break, that is when that layer stopped forming. The likely interpretation for this evidence is that the increasing contribution of deep CO_2 made the dripping waters more acid, and therefore interrupted the deposition. The remarkable changes in isotopic composition occurred in a very short time period and demonstrates that is not likely due to climatic-induced changes of mother solutions.

One possible agent of the recurrent release of deep CO_2 in the last 2,000 years at the Colli Albani volcano, as recorded by the speleothems, could be the periodic increase of crack development preceding the largest historical earthquakes affecting the area of Rome which have produced increase of springs and groundwater temperature, changes of groundwater level and occasionally also intense gas emissions (Funiciello et al., 2003). When examining a list of the earthquakes with effects in Rome (Molin et al., 1995; Donati et al., 1999) some chronological correspondences have emerged between seismic events and the speleothem depositional cycles dated by U/Th dating (Tab. 1).

Artificial outlet of Gabii Lake

Based on a preliminary macroscopic observation of sampled speleothems that showed very few unconformities, the isotopic investigation was carried out at first to detect any repeated cycles due to degassing. The isotopic records do not indicate significant changes or trends that can be interpreted in terms of variation in the deep CO_2 component, due to the opening of cracks preceding seismic events (Fig. 4). The isotopic signatures could be explained more simply as due to small fluctuations or plain trends related to the climatic variations in the course of the last 2,000 years. A detailed dating of speleothems is necessary to

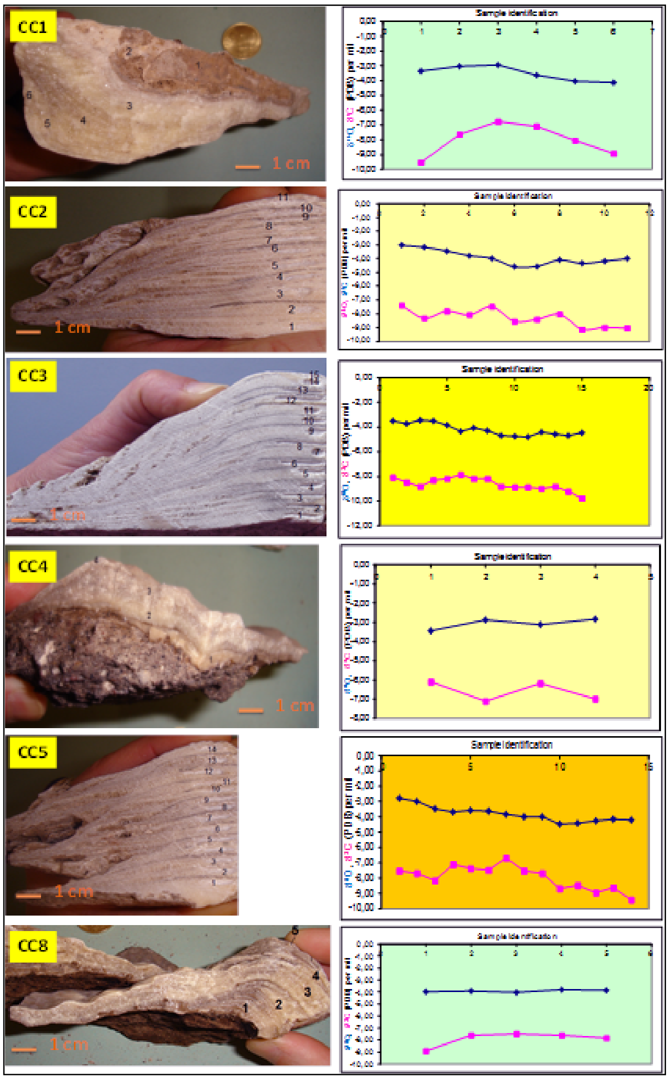

Fig. 4: carbon (lower curve) and oxygen (upper curve) isotopic composition of six speleothems from Gabii lake outlet (Roma, Italy). Numbers indicate subsamples location within the speleothems. Isotopic values are expressed as δ 13C and δ 18O PDB (per mil). See text for explanation.

Fig. 4: composizione isotopica del carbonio (in basso) e dell'ossigeno (in alto) di sei concrezioni dell'emissario del lago di Gabii (Roma, Italia). I numeri indicano la posizione dei sottocampioni all'interno delle concrezioni. I valori isotopici sono espressi come δ13C e δ18O PDB (per mille). Vedere il testo per la spiegazione.

gather more data and to better constrain paleoclimatic and paleoenviromental conditions of the study area.

Conclusions

The study of speleothems from the Centroni cave showed that the growth and the interruption of deposition is connected to periodic input of deep CO_2 in the past. Present and past degassing phenomena seem to be generated by fluid release from aquifer hosted in buried structures of the Mesozoic carbonate basement (Fig. 5). These aquifers act as traps for the gases of deeper origin and become sources of high CO_2 flux toward the surface. In this setting a correlation between the periodic interruption of speleothems growth and the opening of fractures preceding largest earthquakes has been found. It is evident that degassing phenomena are more extended than previously known and future research should include peripheral areas to better understand the location and extension of the geothermal/hydrothermal system at depth and its connection with the shallower aquifers.

Finally, the site of the *Gabii* lake outlet does not seem to be affected by these processes, probably because the area is not interested by systems of deep fractures and faults, as in the Centroni area. The isotopic data are interpreted as due to temperature and precipitation changes occurred in the last 2,000 years. Further dating is expected to provide a more detailed chronological framework to constrain and better interpret oxygen and carbon records.

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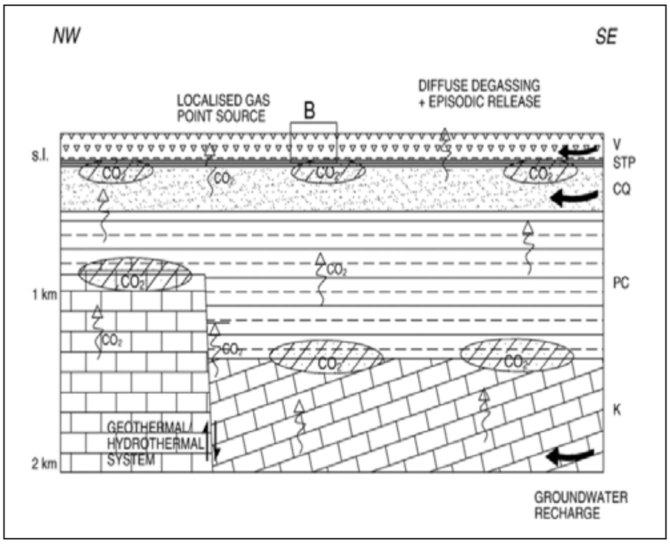

Fig. 5: schematic section of Centroni cave area illustrating a hydrothermal/geothermal reservoir hosted in Mesozoic carbonate formations where the CO_2 produced at depth accumulates, forming gas pockets at the top of the structure. From the gas pockets the gas escapes towards the surface generating CO_2 manifestations, either localised or diffuse. V, STP, CQ, PC and K stands for volcanic products, Tufo pisolitico pyroclastic deposit, Quaternary cover (made up of marine and continental deposits), Plio-Pleistocene clays and Mesozoic carbonates, respectively (modified after Tuccimei et al., 2006).

Fig. 5: sezione schematica dell'area della grotta di Centroni che illustra un serbatoio idrotermale/geotermico ospitato nelle formazioni carbonatiche mesozoiche dove la CO_2 prodotta in profondità si accumula formando sacche di gas nella parte superiore della struttura. Dalle sacche di gas fuoriesce del gas verso la superficie generando CO_2, sia localizzata che diffusa. V, STP, CQ, PC e K stanno rispettivamente per prodotti vulcanici, deposito piroclastico denominato Tufo pisolitico, copertura quaternaria (costituiti da depositi marini e continentali), argille plio-pleistoceniche e carbonati mesozoici (da Tuccimei et al., 2006 modificato).

Sample	Date	Seismic event
FS1-2	1798 - 1820 A.D.	1806 A.D. o 1812 A.D. [b]
FS2-4	1350 -1370 A.D.	09/09/1349 [a]
FS8-4 FS3-2	1250 -1280 A.D.	04/30/1279 [a]
FS2-3 FS9-2b FS9-2c	910 - 960 A.D.	
FS8-3	800 - 820 A.D.	04/29/801 [b]
FS8-2	710 - 750 A.D.	
FS2-1 FS7-2	500 - 550 A.D.	508 A.D. [b]
FS7-1a FS7-1d	430 - 460 A.D.	443 A.D. [b]
FS1-1	300 - 322 A.D.	304 A.D. [b]
FS6-1 [c]	90 - 110 A.D.	94 or 110 A.D. [b]

a, data from DONATI et al. (1999); b, data from (MOLIN et al., 1995).

Tab. 1: interruption of speleothems growth and correlated seismic events from Centroni cave.

Tab. 1: interruzione nella crescita delle concrezioni ed eventi sismici correlati nella grotta di Centroni.

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