

¹⁴C AND U - SERIES DATING OF SPELEOTHEMS IN THE BOHEMIAN PARADISE (CZECH REPUBLIC): RETREAT RATES OF SANDSTONE CAVE WALLS AND IMPLICATIONS FOR CAVE ORIGIN

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ABSTRACT

Speleothems in 6 sandstone caves in the Bohemian Paradise (Český ráj) were dated by means of ¹⁴C and U-series methods. Stable isotopes of C and O, FAAS, IR, XRD, XRF and SEM were used to characterize the carbonate material and its source. Stable isotopes (C and O) composition of speleothems in two caves corresponds to values characteristic for cave speleothems in Central Europe. In other caves they indicate evaporation and fast carbon dioxide escape during carbonate precipitation. The speleothems from the Krtola Cave were deposited between 8 and 13 kyr BP. Speleothems were deposited 5–8 kyr BP in the Sintrová, Mrtvé Údolí and U Studánky caves. Calcite coatings on smooth sandstone surfaces in studied caves demonstrate that cave walls did not retreat even a few mm in the last 5–8 kyr since speleothem deposition and are thus not evolving under recent climatic conditions. Most of the cave ceilings and walls are at present time indurated by hardened surfaces, which protect the sandstone from erosion.

Sandstone caves probably intensively evolved either during or at the end of the Last Glacial period. There are two different erosion mechanisms which might have formed/reshaped the caves at that time:

- A) In the case of permafrost conditions: Repeated freeze/melt cycles affecting sandstone pore space followed by the transport of fallen sand grains by minor temporary trickles. We expect that heat was transmitted by air circulating between the cave and the surface;
- B) Seepage erosion of sandstone during the melting of permafrost, prior forming of case hardening.

KEYWORDS: speleothems, sandstone, age, cave, pseudokarst, erosion, Holocene

1. INTRODUCTION

There are roughly about 2000 rock overhangs and caves in the Bohemian Paradise (Český ráj) Protected Landscape Area (BPPLA) (Fig. 1). The length of caves varies between a few meters and 75 m (Vítek, 1987). The caves are mostly composed of several chambers connected by smaller passages, predominantly subhorizontal (e.g. Figs. 2, 3). In vertical sections the upward chimneys and pockets are mostly deeper than the hollows in the floor filled with sediment. The cave walls are protected by case hardening. In several caves however, the soft and easily erodible sandstone is exposed in places. The sedimentary fill in the caves is mostly several decimetres thick. The cave entrances are mainly

situated several tens of meters above the valley bottom and the thickness of the cave overburden is mostly 5–15 m. Various hypotheses on the evolution of the caves have been published by Vítek (1987), Šídá (2005), Mertlík (2006), Cílek (2006, 2007), Cílek and Žák (2007), Bruthans et al. (2009b) and Adamovič and Mikuláš (2010) but neither hypotheses nor any combination of hypotheses can fully explain the origin of the caves. It also remains unclear when caves and rock overhangs start to develop and how intensely they were enlarged during the Holocene.

Carbonate speleothems from sandstone caves in the BPPLA have not yet been studied in detail, except for the isotopic signature of C and O of calcareous sediments in rock overhangs sediments (Cílek and

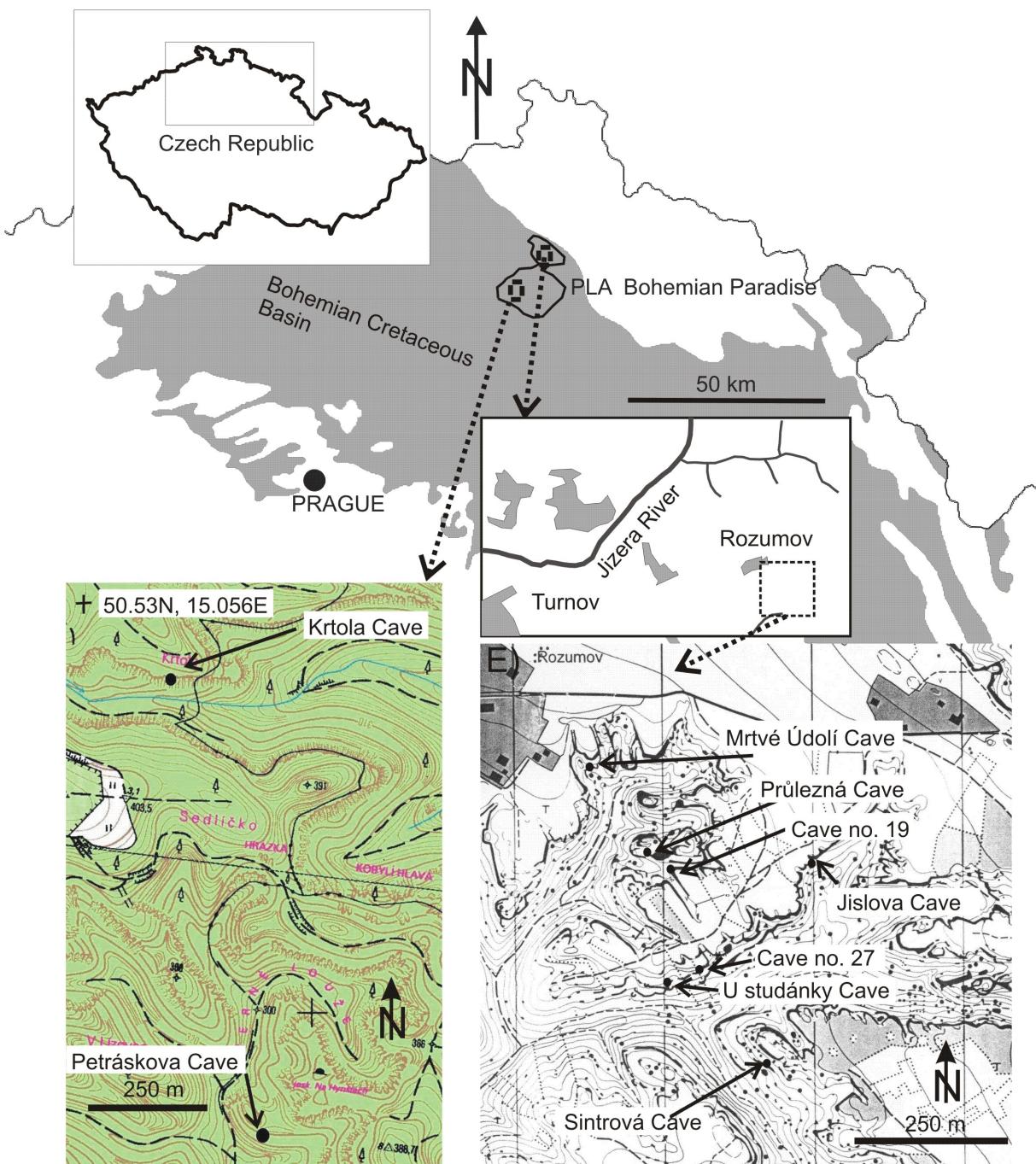


Fig. 1 Location of the study area and caves. PLA – Protected Landscape Area.

Žák, 2007). Speleothems enable radioisotope dating and together with other data may potentially help to reveal the origin of the caves. Since 2006 relatively extensive occurrences of cave speleothems have been found in some sandstone caves in the BPPLA. The preliminary results of dating of speleothems were published by Bruthans et al. (2009a).

The aims of this study were to:

1. Determine the age of speleothems by radioisotope dating (^{14}C and U-series) and find out the source of carbonate.
2. Constrain the minimum age of the caves studied based on radioisotope dating and archaeological

evidence and find out whether the cave walls are retreating under recent conditions.

3. Identify and discuss the processes which may be responsible for the origin of the caves

2. CHARACTERIZATION OF THE STUDY SITES

Caves with speleothems occur in two areas in the BPPLA (Fig. 1): A) the Klokočské Skály area, which is situated 2 km eastward of the town of Turnov, B) the Příhrazské Skály area, which is situated 7 km to the east of Mnichovo Hradiště. Mean annual precipitation is between 600 and 700 mm. Mean

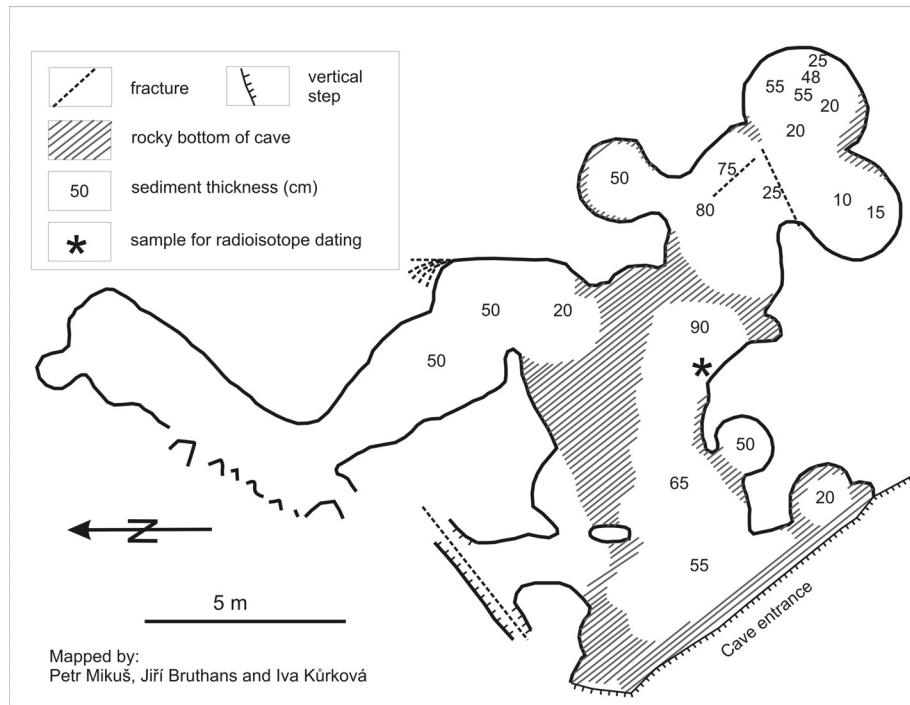


Fig. 2 Horizontal contours (map) of Sintrová Cave.



Fig. 3 Typical cave morphology. Inner part of Sintrová Cave. Photo by J. Bruthans.

annual temperature is about 8 °C (Czech Hydrometeorological Institute).

These areas are located in the Bohemian Cretaceous Basin (Fig. 1), which is formed of marine sandstones alternating with marlstones. Sandstone of Teplice Formation was deposited on the submarine delta environment during the Upper Turonian-Lower Coniacian period (Uličný, 2001). The sandstone is fractured and faulted but not folded. Bedding planes are tilted slightly toward the SW with a dip < 5°. The sandstone is composed of mostly quartz grains with minor rock fragments, feldspars, micas. The matrix is a kaolinite and/or illite mixture. The sandstone is

locally cemented by Fe-oxyhydroxides or by secondary silica (Schweigstillová et al., 2005).

2.1. DESCRIPTION OF THE STUDIED CAVES

Krtola Cave (named also Sklep na Chodové, Fig. 4) is 40 m long. It consists of two flat chambers connected by rather small openings (largest chamber 32 × 18 × 3 m; Cílek, 2006). Cílek (2006) noted an occurrence of several types of speleothems (secondary carbonates) in Krtola Cave. The first type, calcite coating, was porous, up to 14 mm thick, partly plastic (soft), fibrous on a microscopic scale (usually called moonmilk, e.g. Pakr, 1979) in different degrees of

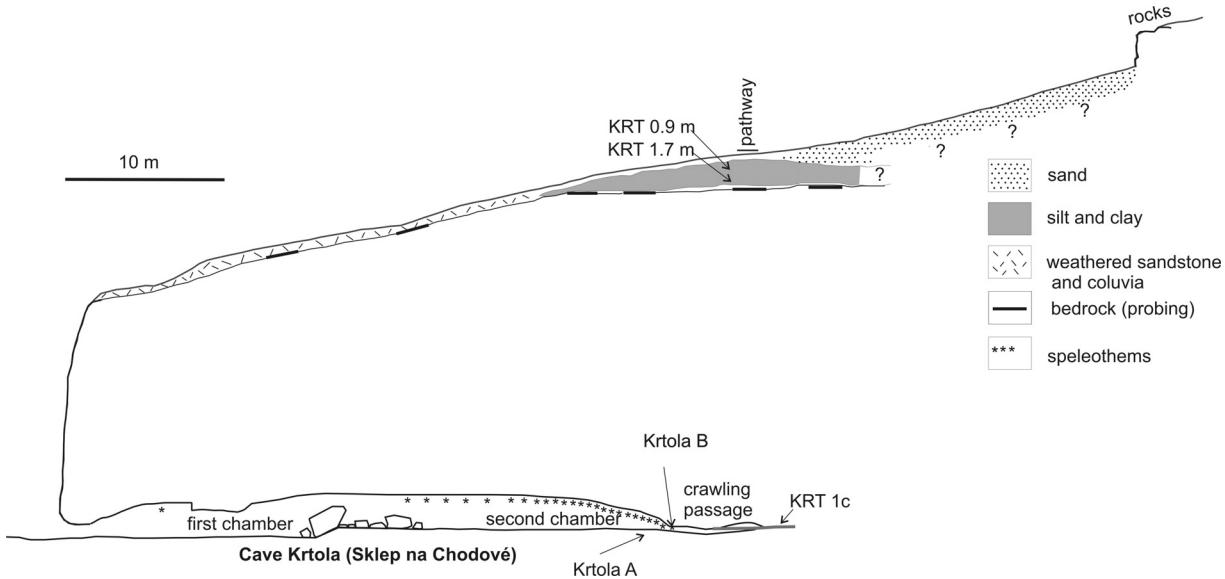


Fig. 4 Cross-section of Krtola Cave and its overburden with type and thickness of superficial deposits and sampling sites. Blocks in cave collapsed along subhorizontal fractures (not visible).

fossilization. The second type is formed by calcite-indurated layer of sand grains and sandstone fragments. It was found in an inner part of the second chamber 40–45 cm below the sediment surface in the dug hole. Krtola Cave was originally completely filled with cave sediments behind the second chamber. The crawl space dug through the sediments by cavers revealed the presence of a collapsed sandstone ceiling blocking the cave passage. Whitish calcite precipitates fill fractures in the collapsed material.

U Studánky Cave is 25 m long. The cave is composed of an entrance chamber 8 × 6 m connected with several small crawl passages, which were originally filled by sand. Sintrová Cave is 42 m long. It is composed of a branching network of cave passages (Fig. 2). The cave passages are blindly terminated in spherical cavities, which are up to 3 m in diameter. Průlezna Cave (no. 17) is a simple straight gallery 22 m long, ~1 m in diameter. It has two entrances with an elevation difference of 4 m (Vítek, 1987). Mrtvé Údolí Cave is about 7 m long. It consists of three spherical chambers (about 2–4 m in diameter) connected by openings ~1 m in diameter. Petráskova Cave is a large gallery about 30 m long, approx. 3–5 m wide, predominantly filled with sediment. The entrance is very small (0.5 × 0.5 m) and was enlarged by digging from fox-earth in 1996 year. Cave no. 19 is 26 m long and has two entrances. It consists of several chambers up to 5 m in size connected by small openings. The elevation difference between the entrances is ca 5 m (Vítek, 1987). Cave no. 27 is 16 m long with two entrances (Vítek, 1987). It consists of 4 spherical cavities up to 4 m in diameter connected by small openings. Caves are situated at an altitude between 320 and 380 m a.s.l.

2.2. DEPOSITION RATE IN CAVES BASED ON ARCHAEOLOGICAL FINDS

Nearly 130 speleo-archaeological sites were registered in the BPPLA prior to 2005, giving movable finds from the Prehistoric hunting period to the Modern Period (Jenč, 2006; Jenč and Peša, 2007). At present time there are approximately 170 such sites.

By means of standard speleo-archaeological methods, i.e. interdisciplinary archaeological-natural research with detailed spatial documentation of all the artefacts and ecofacts (biofacts) uncovered and the sieving and washing of deposits, several Mesolithic sites (dating back to the 9th–6th millennium BC) have recently been identified (Svoboda ed., 2003; Matoušek et al., 2005). Knowledge concerning the use of rock overhangs and caves in the BPPLA at the end of the Prehistoric Hunting Period has been significantly enhanced by fresh critical evaluation of the stone industry obtained from earlier excavations (1st half of the 20th century) in the Turnov region (Šídá and Prostředník, 2007).

The Prehistoric Hunting period is represented by at least one Palaeolithic site, Jislova Cave (age approx. 90 kyr – 40/35 kyr BC, archaeological find horizon in 0.8 – 1.2 m; Filip, 1947; Fridrich, 1982; Šídá, 2005) and 10–12 Mesolithic sites, two of which, in Borecké rocks, are outside the BPPLA. New research into the Mesolithic settlement in the BPPLA has also yielded several radiocarbon data, indicating – after calibration – the 8th millennium BC to the 1st half of the 6th millennium BC (Abri Pod Pradědem – Hrubá Skála region, Věžák lake rock overhang, Kristova Cave in the Klokočské skály area) (Prostředník and Šídá, 2006). The upper surface of deposits with Mesolithic finds in rock overhangs in

the Česká Lípa region and in České Švýcarsko is situated at depths of between 50 and 100 cm, in rare cases even at 20 cm (Arba near Srbska Kamenice or Okrouhlík near Vysoká Lípa; Svoboda ed., 2003). Similar depths have been identified for deposits dating back to the early to mid Holocene in rock overhangs in the Bohemian Paradise, such as the bed of layers in Kristova Cave (Prostředník and Šídá, 2006). In the case of very shallow rock overhangs (Dolský Mlýn) the upper surface of deposits with Mesolithic finds can be situated much deeper (200 cm).

In Portal Cave in Mužský hill a late Neolithic horizon (1st half of the 5th millennium BC) is situated at a depth of 60–70 cm, which is the typical depth for this sandstone area (Jenč and Prostředník, 1995; Jenč, 2006). An archaeological find horizon dating back to the 5th–4th millennium BC in Oko Cave near Branžež was identified at a depth of only 20–40 cm (Jenč, 2006).

The archaeological find horizon from the beginning of the Late Holocene (Lusatian culture, 1400–100 BC), as regards shallow caves, lies some 40–50 cm deep, such as at Portal Cave in Mužský hill (Jenč and Prostředník, 1995, Jenč 2006), Kristova Cave (Prostředník and Šídá 2006; Hartman et al., in press), Velký Mamut'ák rock overhang and Kopřivák 1 (Jenč, 2006). Deeper caves little affected by outside transfers of sediment may contain finds from the end of the 2nd millennium BC at a depth of around 20 cm (Postojná and Jislova caves) (Filip, 1947; Šídá, 2005). The cases of a cultural layer of this age, lying only a few centimetres below the surface, are not extraordinary, see Zlatá Cave in the Klokočské skály area (finds of the Lusatian culture have been identified 2–10 cm below the surface). The calculated mean long-term sediment deposition rate for sandstone caves and rock overhangs is 3–25 cm/kyr based on the above values. The depth of archaeological find horizons generally decreases from the entrance part of the cave into the inner part of the cave. The highest thickness was found in very shallow rock overhangs.

3. METHODS

3.1. SAMPLING AND FIELD EXPERIMENTS

The cave spaces and the profile above Krtola Cave were mapped with tape, a compass and a precise inclinometer. The thickness of sediments inside and above the caves was measured using a steel probing bars hammered from the surface to depths of 1 and 2 m. Samples of sediment core were taken by means of these probes. The carbonate content was checked with 10 % HCl in the field for fast screening; samples were taken for precise laboratory analyses.

The temperature and humidity of the cave air were measured using a GMH 3350 (Greisinger electronic, Germany). The airflow velocity in the caves was measured using an Air Velocity Meter (TA410).

3.2. SAMPLE TREATMENT AND LABORATORY ANALYSES

We sampled 8 sites with speleothem occurrence from six caves. Sets of two samples were taken in Krtola and Petráškova caves. Sand grains were removed from samples of speleothems prior to ¹⁴C and U-series dating. The only exception is Krtola Cave A sample, which was analysed in bulk, including dispersed sandstone fragments in the carbonate material. Carbonate samples were cleaned both mechanically and chemically (washed briefly in 10 % HCl).

A Leica DMRX microscope with Leica DC300 photocamera was used to take photos of the speleothems. A Quanta 450 (FEI) scanning electron microscope was used to study of the speleothem surface morphology. Observations of the fresh uncovered samples were performed in secondary electron (SE) mode under low vacuum (80 Pa) with the energy of the electron beam 15 kV.

The chemical composition of the material from KRT 1C, KRT 0.9 m and KRT 1.7 m was determined by flame atomic absorption spectroscopy (FAAS, Czech Geological Survey). The chemical composition of the material from U Studánky Cave, Sintrová Cave and Krtola Cave B was analysed by X-ray fluorescence (XRF, Institute of Chemical Technology, Prague) (Table 1). The carbonate content from the samples was determined by infrared spectrometry after decomposition with acid as CO₂ amount (IR, Czech Geological Survey) (Tables 1, 3).

The mineral composition of the samples (Tables 1, 3) was determined by X-ray powder diffraction (XRD) with a PANalytical X'Pert PRO diffractometer equipped with a conventional X-ray tube (CuKα 40 kV, 30 mA, line focus) in transmission mode. An elliptic focusing mirror, a divergence slit of 0.5°, an anti-scatter slit of 0.5°, a Soller slit of 0.02 rad and a mask of 20 mm were used in the primary beam. A fast linear position sensitive detector PIXcel with an anti-scatter shield and a Soller slit of 0.02 rad were used in the diffracted beam. All patterns were collected in the range of 2 to 88 deg. 2theta with the step of 0.013 deg and 100 sec/step.

Qualitative analysis was performed using the HighScorePlus software package (PANalytical, the Netherlands, version 3.0d), Diffrac-Plus software package (Bruker AXS, Germany, version 8.0) and JCPDS PDF-2 database. For quantitative analysis of XRD patterns we used Diffrac-Plus Topas (Bruker AXS, Germany, version 4.2) with structural models based on the Inorganic Crystal Structure Database. This program permits to estimate the weight fractions of crystalline phases by means of Rietveld refinement procedure.

The sample material was reacted with 100 % H₃PO₄ in a vacuum at 25 °C, following McCreas' method (1950) for the stable isotope determination. Stable isotope determinations of the prepared CO₂ gas were performed in the laboratories of the Czech

Table 1 Chemical and mineralogical composition of selected speleothems.
(XRF – X-ray fluorescence; IR - infrared spectroscopy; XRD – X-ray diffraction)

method	XRF	XRF	XRF	XRF	XRF	IR	XRD
sample	CaO (%)	MgO (%)	SrO (%)	SiO ₂ (%)	P ₂ O ₅ (%)	CO ₂ (%)	
U Studánky Cave	54.35	0.341	0.0155	1.99	0.065	39.97	calcite
Sintrová Cave	55.76	0.127	0.01	0.087	0.034	43.02	calcite
Krtola Cave B	55.33	0.565	0.03	0.03	0.03	43.24	calcite

Table 3 Chemical and mineral composition of clays from and above Krtola Cave.
(FAAS – flame atomic absorption spectroscopy; IR – infrared spectroscopy ; XRD – X-ray diffraction)

method	FAAS	FAAS	IR	XRD	XRD	
sample	CaO (%)	MgO (%)	CO ₂ (%)	major	minor	
KRT 1C	0.58	1.1	< 0.01	quartz	K-feldspar, illite, kaolinite, smectite	
KRT 0.9 m	0.28	0.92	< 0.01	quartz	K-feldspar, illite, smectite, chlorite	
KRT 1.7 m	0.43	1.15	< 0.01	quartz	smectite, illite, chlorite, K-feldspar	

Table 2 Frequency of grain diameters of KRT 0.9 sample of silt sediment

Diameter (μm)	Frequency (%)
0.3	0.6
0.5	1.3
1	3.2
2	6.9
4	11.4
6	8.3
8	7.3
10	6.6
12	5.9
15	7.8
20	10.4
32	15.1
45	7.7
63	4.7
90	2.2
140	0
180	0
230	0
300	0
400	0

Geological Survey, Prague, using a Finnigan MAT 251 mass spectrometer. The overall analytical uncertainty, established by repeated analyses of the NBS-19 international carbonate standard, was ±0.1 % for both δ¹³C and δ¹⁸O values.

Carbon dioxide was released by phosphoric acid, and through lithium carbide and acetylene it was transformed to benzene. The radiocarbon activity of benzene was measured using a Tri Carb 3170 liquid scintillation spectrometer. The samples were prepared

and ¹⁴C determined in the radioisotope laboratory of Charles University in Prague.

Selected samples (Krtola Cave B, Krtola Cave B* and U Studánky Cave) were analysed for ²³⁰Th/²³⁴U ratios with alpha spectrometry in the laboratories of the Institute of Geological Sciences, Polish Academy of Science, in Warsaw. For uranium and thorium separation from carbonates, a standard chemical procedure was used (Ivanovich and Harmon, 1992). ²²⁸Th/²³²U spikes were used for the control efficiency of the chemical separation procedure. The samples were dissolved in 6M nitric acid. The U and Th fractions were separated by the chromatography method using DOWEX1×8 as an anion exchanger. The alpha particles spectrum was measured at OCTETE PC spectrometer (EG&GORTEC). Spectra analysis and age calculations were done using the URANOTHOR software, version 2.6, which is the standard software used in the U-Series Laboratory in Warsaw (Gorka and Hercman, 2002; half-life values after Cheng et al., 2000). The quoted errors are 1σ.

4. RESULTS

4.1. SOIL AND SEDIMENTS ABOVE THE CAVES

As sandstone does not contain carbonate, the soil zone above the caves was studied to search for fine sediments, which might contain carbonate and thus serve as a source for speleothems in the past. The soil zone above sandstone caves is very shallow or missing with the exception of Krtola Cave, where sediments and soil above the cave were studied in detail.

A layer of soil and weathered sandstone up to 50 cm thick was found on the surface above the first half of Krtola Cave. A yellow silty material (sample KRT 0.9 m taken 0.9 m below surface; Fig. 4), was detected by probing above the cave, at a distance of 30–47 m from the cave entrance. The maximum

detected thickness of the silty layer is about 1.5 m, and its base is nearly horizontal (Fig. 4). Grey clay about 30 cm thick occurs in places below the yellow silt (sample KRT 1.7 m, taken 1.7 m below the ground surface). The silt and clay are partly covered by sand from weathered sandstone (Fig. 4). None of the materials excavated within 2 m of the surface contain carbonate at the present time (tested in the field with 10 % HCl and confirmed by laboratory analysis; Table 3). The grain size distribution of the KRT 0.9 m sample is bimodal (Table 2) and it is similar to some samples of loess deposits in the Střeleč Quarry (approx. 12 km west of Krtola Caves, the same bedrock lithology; Černý et al. 1976). The <0.063 mm fraction is overwhelmingly dominated by quartz. The heavy mineral assemblage of KRT 0.9 m is composed of 90 % of opaque minerals. The remaining part consists of 70 % staurolite, 13 % pyroxene, 10 % tourmaline, 5 % rutile and 2 % other minerals. These are interpreted as an ultra-stable assemblage (L. Lisá, pers. comm.).

The mineral composition of fine fractions of samples KRT 0.9 and 1.7 m was compared with marine bioturbated clay (KRT 1C), which is exposed in the wall of Krtola Cave (Fig. 4, Table 3). There is a distinct difference between these two materials in terms of the clay composition: The sediments above the cave contain chlorite and do not contain kaolinite, while the opposite is true for the KRT 1C marine clays. The sediment above the cave may be interpreted as loess, but additional analyses are needed for confirmation.

4.2. WATER FLOW AND AIR CIRCULATION IN CAVES

At the present time the caves are mostly dry. Only after an intense thaw or rain do trickles flow on the bottom of some caves for a few hours or days. In Cave no. 19 the trickle 0.05 l/s was observed inflowing via upper cave entrance and intensively transporting sand on the cave bottom with inclination of a few %. Even a flow rate of 0.001 l/s can transport sand in a channel several cm wide with gradient 6 % only (Bruthans et al., in rev.). The longitudinal profiles of the caves thus allow the loose material to be transported by very small amounts of flowing water. In most of the caves visitors disturbed the sediment bottom.

Temperature, air humidity, airflow direction and velocity were measured in several caves to test if the air circulation could possibly transport the heat between the cave and the surface (see chapter 5.3). Cave air temperatures vary greatly during the year. In wintertime the air temperature even in the deep parts of caves often drops below freezing point. In summer the temperature of cave air is often just a few °C lower than the temperature of the outside air (Caves no. 19 and 27). The temperature, airflow direction and velocity were studied in detail at 3.8.2009 in caves no. 19 and 27 in the Klokočské Skály area. The summer

period was selected to simulate the air flow circulation during the Last Glacial summer periods (the absolute air temperature was much lower at that time but the sandstone walls were cooler than the air, like in recent summer periods). The measurements clearly demonstrated that air circulates even in the deepest parts of caves. Relatively warm air enters the cavities along the ceiling; it cools, falls, and returns back along the cave bottom. Circular cavities with an entrance only 50 cm wide are subject to active air circulation (Fig. 5).

4.3. CAVE SPELEOTHEMS

Two samples of speleothems were taken from Krtola Cave at two different sites (Fig. 4). The Krtola Cave A sample was taken 31 m away from the cave entrance, from an indurated layer of sand grains and sandstone fragments about 15 cm thick, strongly cemented by carbonate. The sample was taken from excavated material from the crawl passage that leads to the far end of the cave (Fig. 4). This is probably the same material described by Cílek (2006) from a hole dug in cave sediments. The second sample (Krtola Cave B) was taken from the side of the crawl passage (Fig. 6) about 35 m from the cave entrance. It consists of white calcite 1 cm thick with no admixture of the sand grains. Calcite filled the fracture in the material collapsed from the ceiling. The Krtola Cave B* subsample is additional material from the same place as Krtola Cave B, but with a higher content of non-carbonate fraction.

The Krtola Cave A sample dates the minimum age of the last erosion phase in the cave, when the cave had no sediment cover. The Krtola Cave B sample dates the minimum age of the collapse of the passage behind the second chamber.

Speleothems in the U Studánky, Sintrová, Mrtvé Údolí, Průlezná and Petráskova caves are developed in the form of calcite coatings (thickness up to ca 20 mm) covering the smooth surface of the sandstone (cave wall and/or ceiling) on an area about 0.1-2 m². The speleothems strongly resemble the "blankets of calcite moonmilk" (Sintrová and Průlezná caves) and "crusts of cauliflower-like calcite speleothems" (U Studánky Cave) described by Urban et al. (2007a). Later in the text we use the term calcite coating to encompass all these types. In neither cave do the cave walls/ceilings show any traces of surface retreat in the surroundings of the calcite coatings (Fig. 7). The calcite coating samples in all these caves thus date the minimum age when the retreat of the cave walls ceased.

In Sintrová Cave the calcite coating ~1 cm thick located 7 m from the entrance was sampled (Fig. 7). Similar material situated 3 m from the lower entrance was sampled in Průlezná Cave. In the U Studánky and Mrtvé Údolí caves, the calcite coatings ~1.5 cm thick covering the small circular cavities in the ceiling of the entrance chamber were sampled about 4 m and 3 m from the entrance, respectively. In Petráskova

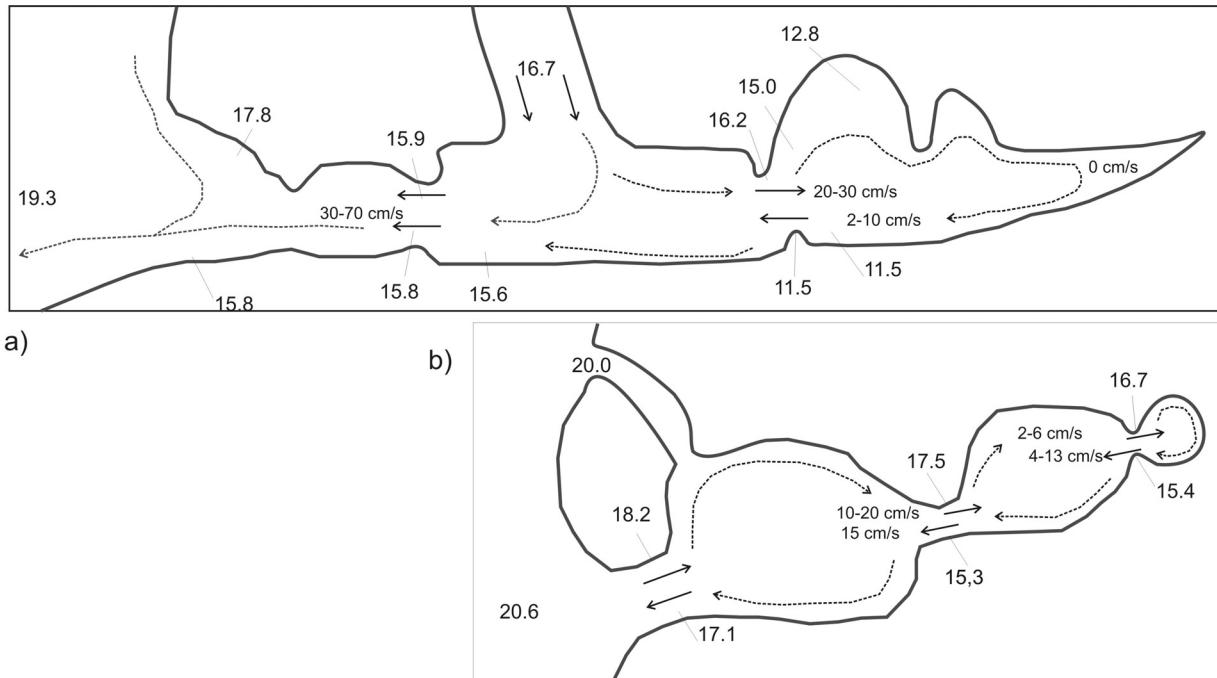


Fig. 5 Sketch of the vertical cross-sections of caves with temperature ($^{\circ}\text{C}$), air flow direction and velocity at 3.8.2009: a) Cave no. 19, b) Cave no. 27.



Fig. 6 White calcite filling fissures in sandstone and fallen blocks equivalent to sample "Krtola Cave B". Photo by J. Bruthans.

Cave the calcite coating ~1 cm thick was sampled at two places on an overhanging wall 3 m from the entrance (Fig. 8): Sample A from Petráska Cave represents the relatively well-preserved calcite coating sampled from the upper part of the cave wall, while Petráska Cave B represents a strongly weathered calcite coating (material that crumbles and falls off easily) situated closer to the cave bottom. Both samples in Petráska Cave were taken from a single calcite coating, which probably started to

precipitate at the same time. The reason for dating both samples was to test the effect of weathering and calcite re-precipitation on an apparent radiocarbon age. However, the Petráska Cave B sample is partly covered by a thin whitish newly - re(?) - precipitated carbonate.

Based on macroscopic inspection, the Krtola Cave A sample is composed of sandstone fragments (size up to 15 mm) cemented and coated by calcite (calcite coating up to 3 mm thick). The calcite is firm.



Fig. 7 Calcite coating on the wall in Sintrová Cave. The smooth sandstone surface stretches under the calcite coating without any changes, which proves that the cave wall did not retreat more than a few millimetres after calcite deposition. Photo by J. Bruthans.

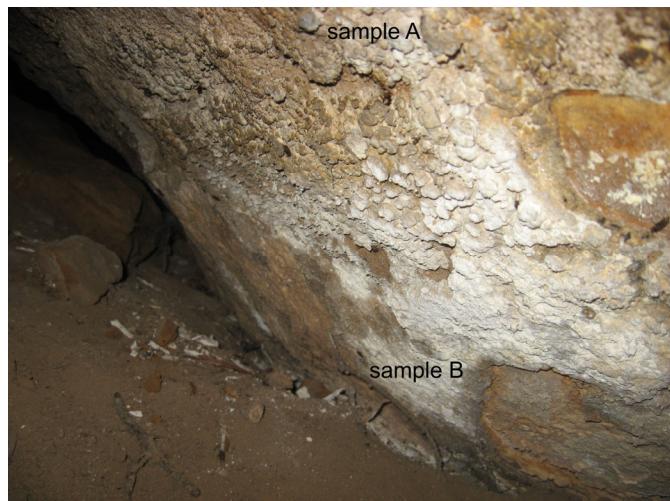


Fig. 8 Calcite coating with coraloids (cauliflower-like forms) on the entrance crawl space in Petráskova Cave. Positions of samples A and B is indicated. The entrance crawl space is on right side of the picture. Photo by J. Bruthans.

There are many air filled pores, which are several mm in diameter. The Sintrová and Průlezná Cave samples are formed by very light and soft calcite material up to 1 cm thick (easily brushed with the fingers). The slightly greenish colour of the Sintrová Cave sample is probably caused by the presence of algae. The U Studánky Cave sample consists of a basal layer about 5 mm thick, which shows internal lamination. This zone is covered by cauliflower-like forms (size up to 10 mm, coralloid – see Hill and Forti 1997). The material is firm. The Petráskova Cave A and B samples are composed of light and rather soft calcite material covered by cauliflower-like forms (size up to 15 mm).

Based on SEM, the carbonate material of Krtola A sample is composed of irregular columnar microcrystals ranging in size approximately between 10 and 20 μm . The individual microcrystals have an irregular probably corroded surface (Fig. 9a). The material is porous in microscale, but the porosity seems to be lower compared to the samples from the other caves. The Sintrová and Průlezná Cave samples are highly porous. The material of these two samples consists of a mixture of more or less elongated microcrystals (1-50 μm in the longer axis) with a micrograined carbonate matter (Fig. 9b). The U Studánky Cave sample is highly porous. It is also composed of microcrystals, which are rather regular in

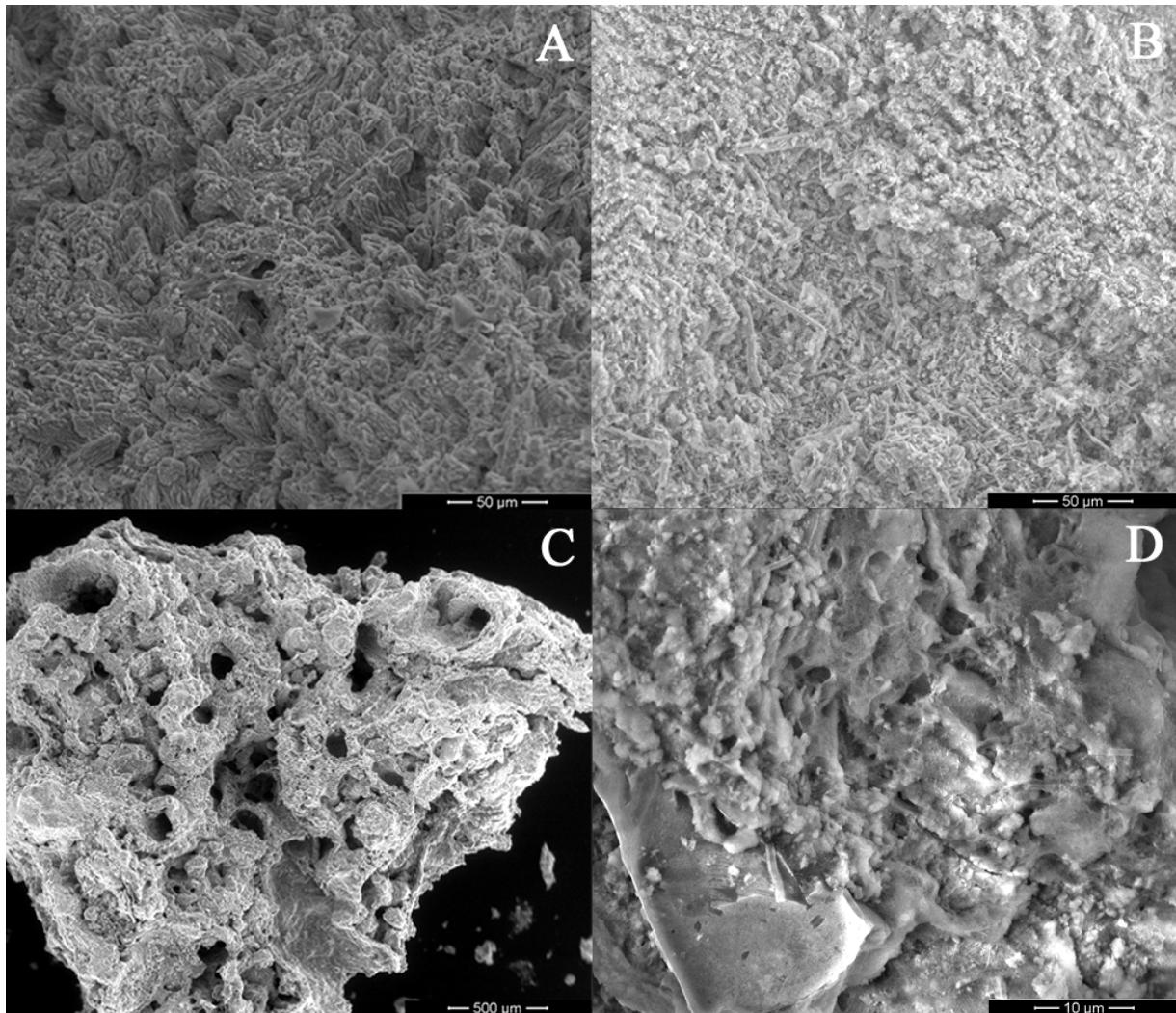


Fig. 9 SEM of speleothem materials: a) carbonate of Krtola Cave A sample; b) Sintrová Cave sample; c) Petráska Cave B sample; d) Petráska Cave B sample, detail.

shape and range between 1 and 5 μm in size. The Petráska Cave A sample is highly porous composed of elongated microcrystals of 1–10 μm in size. In places the microcrystals appear to be recrystallized into larger units (boundaries between them are no longer visible). The Petráska Cave B sample is composed of very fine-grained carbonate matter and some kind of microconduits with a circular cross-section are typical (Fig. 9c). The diameter of these microconduits ranges between 10–100 μm . Material of the Petráska Cave B was originally probably composed of microcrystals, which were later transformed into a large compact aggregates, however, remnants of microcrystals can be still traced (Fig. 9d). The Průlezná Cave and Krtola B samples were not studied by SEM due to the lack of material.

At present, aluminium-rich salt efflorescences have been precipitated in cave entrances and their surroundings (alunite and alums; for example in the surroundings of Krtola Cave – Cílek, 2006, the ceiling in the entrance part of U Studánky Cave). Water

dripping from the sandstone unsaturated zone in Klokočské Skály has a pH of between 3.9 and 4.6 and aluminium is the prevailing cation (Bruthans and Schweigstillová, 2009).

4.4. ISOTOPIC COMPOSITION OF SPELEOTHEMS, THEIR RADIOCARBON ACTIVITY AND U-SERIES DATING

The speleothems studied are composed of low-magnesium calcite (Table 1). The isotope composition of C and O of speleothems in the Krtola and Petráska caves can be classed as common Pleistocene and Holocene speleothems of Central Europe (Fig. 10). On the other hand the isotope composition of C and O of speleothems in all the caves in the Klokočské Skály area follows the evaporation trend due to the disequilibrium escape of CO_2 and/or water evaporation, especially at the U Studánky and Průlezná Caves (Fig. 10).

The radiocarbon activity of the sampled speleothems was 20–54 % of modern carbon (pmc)

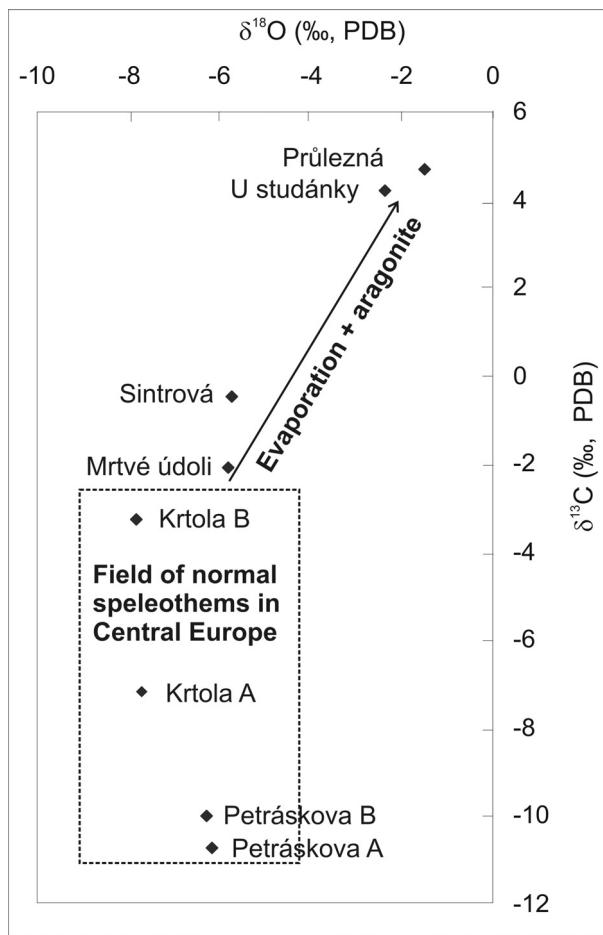


Fig. 10 C and O isotope data of the speleothem samples studied. The field of common Pleistocene and Holocene speleothems of Central Europe follows the data of Šmejkal et al. (1976), Cílek and Šmejkal (1986) and Žák et al. (1987). The evaporation and aragonite trend influenced by rapid non-equilibrium, kinetic CO₂ escape for speleothems follow Cílek and Šmejkal (1986).

Table 4 C and O isotope data of studied speleothem samples and results of radiocarbon dating.

* The cave entrance was buried by colluvial deposits before 1996.

sample	distance of sampled secondary carbonates from cave entrance (m)	sample no.	δ¹³C ‰ PDB	δ¹⁸O ‰ PDB	percent modern carbon (%)	conventional age (years BP)	standard deviation (years)	calculated age for initial activity 70 pmc (kyr BP)	calculated age for initial activity 90 pmc (kyr BP)
Krtola Cave A	31	CU 698	-7.2	-7.7	20.5	12740	170	9.9	11.9
Krtola Cave B	35	CU 703	-3.3	-7.8	26.2	10770	200	7.9	9.9
U Studánky Cave	4	CU 707	4.2	-2.4	37.6	7860	150	5.0	7.0
Sintrová Cave	7	CU 713	-0.5	-5.8	32.3	9070	250	6.2	8.2
Petráskova Cave A	3*	CU 715	-10.7	-6.2	40.5	7260	170	4.4	6.4
Petráskova Cave B	3*	CU 716	-10.0	-6.3	54.4	4880	140	2.0	4.0
Průlezná Cave	3	CU 733	4.7	-1.5	49.4	5660	150	2.8	4.8
Mrvté Údolí Cave	4	CU 734	-2.1	-5.8	33.8	8710	280	5.8	7.9

(Table 4). The uncorrected U-series age (Table 5) of two parts of the Krtola B sample is 13.7 ± 1.0 and 15.8 ± 2.5 kyr. The sample from U Studánky Cave has the uncorrected U-series age 23.7 ± 1.8 kyr.

5. DISCUSSION

5.1. SOURCE OF CARBONATE

In spatial terms, the fine-grained sediment above Krtola Cave (probably loess) closely correlates with the occurrence of speleothems in Krtola Cave (Fig. 4). This fine-grained sediment is thus the probable source of carbonate, which was re-precipitated in the cave as speleothems. At the present time the carbonate is leached out from sediments above the cave (Table 3).

The soil zone and aeration zone of sandstone above the studied caves do not contain carbonates based on the presence of alum efflorescence (chapter 4.3). Carbonate is clearly leached out completely even deep inside the sandstone. If the water in the unsaturated zone comes into contact with even a slight amount of carbonate, the water will be buffered to a pH close to 7 and Al will be immobilized.

The current precipitation of alunite efflorescence in the same caves where carbonate speleothems were precipitated several thousand years ago demonstrates that the chemistry of water in the unsaturated zone changed drastically since the carbonate deposition. After the depletion of the carbonate source in the soil and the unsaturated zone, the water with a pH of about 7 was replaced by water with a current pH close to 4, which is rich in aluminium and sulphate

5.2. AGE OF SPELEOTHEMS

For speleothem dating, it is necessary to know the initial activity of ¹⁴C (or reservoir effect – Goslar et al., 2000) of the carbonate material. The initial activity of the dissolved carbonate in the sandstone

Table 5 Results of U-series dating. Krtola Cave B* is additional material from the same place as Krtola Cave B, but with a higher content of non-carbonate fraction.

Sample	Lab. No.	U cont. [ppm]	$^{234}\text{U}/^{238}\text{U}$	$^{230}\text{Th}/^{234}\text{U}$	$^{230}\text{Th}/^{232}\text{T}$ h	Age [kyr]
Krtola Cave B	W 2162	0.192±0.005	1.316±0.043	0.119±0.008	23	13.7 ± 1.0
Krtola Cave B*	W 2164	0.062±0.004	1.538±0.123	0.136±0.021	21	15.8 ± 2.5
U Studánky Cave	W 2165	0.057±0.002	1.248±0.050	0.198±0.014	24	23.7 ± 1.8

unsaturated zone cannot be measured since the carbonate has now been completely leached out of the soil and the unsaturated zone above the caves. Dissolution of carbonate in rather open than closed system can be expected in the relatively thin sandstone aeration zone.

Initial activities usually range between 70 and 90 pmc, corresponding to apparent ages between 2750 and 750 yr (Goslar et al., 2000). Similar initial activity (94 pmc) was measured in the carbonate rich water at the base of the unsaturated zone in Ochoz Cave in the Moravian Karst (drip site E, unpublished results).

Considering the initial activity of 70-90 pmc (Table 4), the apparent ^{14}C ages of the Krtola Cave A and B samples are 10–12 kyr before the present (kyr BP), and 8–10 kyr BP, respectively (Table 4). The Sintrová and Mrtvé Údolí caves samples give a radiocarbon age of 6–8 kyr BP, while the Petráskova A and U Studánky caves samples give a radiocarbon age of 4.4–7 kyr BP. The lowest radiocarbon ages are shown with samples from the Petráskova B and Průlezna caves (2–4.8 kyr BP).

These apparent radiocarbon ages probably underestimate the real age of the original speleothems. It is likely that a part of the speleothem material was repeatedly dissolved in participation with biogenic CO_2 , and new ^{14}C was introduced into the carbonate (due to condensation corrosion, biogenic processes in the photic zone, etc.). Except Krtola Cave the speleothems occur within photic zone, close to cave entrances. Therefore speleothems might be affected by significant temperature changes during a year, moisture condensation and algae or other biota activity. It can be postulated that our samples represent a mixture of original and newly generated carbonate. This can be demonstrated if we compare the radiocarbon ages of samples from Petráskova Cave: the weathered sample B shows an apparent age 2 kyr lower than the non-weathered sample A. Dissolution of the carbonate is indicated by presence of microscopic openings in the Petráskova Cave B sample (Fig. 9c). The real age of the onset of speleothem deposition will thus be equal or higher than the aforementioned apparent ages. The radiocarbon apparent ages of speleothems relatively often underestimate the real age by several kyr or more (Goslar et al., 2000).

The basic requirements for age determinations based on U-series disequilibrium methods are (Borsato et al., 2003): 1) that samples contain sufficient amounts of U (>0.05 ppm); 2) that it is not contaminated by detrital Th; and 3) that the speleothem system has remained closed since the time it formed, meaning that U and products of its decay, particularly ^{230}Th , are neither added nor removed from the sample. While the first condition is clearly satisfied with the samples studied, the other two might not be. The open-system behaviour was mostly observed in speleothems that have experienced dramatic environmental changes such as dissolution by undersaturated waters (Borsato et al., 2005), which might be also the case of the speleothems in question. $^{230}\text{Th}/^{232}\text{Th}$ activity ratio is 21–24, which indicates some presence of detrital thorium. The U-series ages were not corrected for the effect of detrital ^{230}Th since $^{230}\text{Th}/^{232}\text{Th}$ activity ratio exceeds 20. If corrected, the corrected ages would be significantly lower than the uncorrected ages presented in Table 5. The U-series data does not show explicit signs of uranium leaching ($^{234}\text{U}/^{238}\text{U}$ between 1.25–1.32). However, uranium leaching is not uncommon even for speleothems, which does not show $^{234}\text{U}/^{238}\text{U}$ ratio close to 1 (K. Žák pers. comm.). Generally the U-series dating results relatively often give apparent ages which are several thousand years or older than the real age, especially when dating fine-grained material (Whitehead et al., 1999; Railsback et al., 2002; Urban et al., 2007b; K. Žák pers. comm.).

The U-series apparent ages (Table 5) are in all cases higher than the apparent ages based on the ^{14}C method (Table 4) for the same samples. As mentioned above, the U-series apparent ages are likely to overestimate the real age (due to open-system conditions), while the radiocarbon ages will likely underestimate the real age due to contamination by younger carbon. The real age falls probably between both values. The Krtola Cave B and B* subsamples show an apparent U-series uncorrected age of 13.7 ± 1.0 kyr and 15.8 ± 2.5 kyr, respectively, while the apparent ^{14}C age is 7.9–9.9 kyr BP. The difference is about 3 kyr between both methods. On the contrary, the sample from U Studánky Cave has the apparent U-series age of 23.7 ± 1.8 kyr, while the apparent ^{14}C age is 5.0–7.0 kyr BP. The difference between both

dating methods is 15 kyr. It seems highly unlikely that the apparent age of about 22-26 kyr (U Studánky Cave, U-series) is real, as the study area was subjected to permafrost in this period, and calcite, if precipitated, would have a completely different isotopic composition (Žák et al., 2004). A much larger difference between the results of the U-series and ¹⁴C methods was found for the speleothem sample in a very shallow environment (U Studánky Cave), compared to the deep part of the cave (Krtola Cave B).

We are not aware of other dated speleothems in sandstone caves in the Czech Republic. Speleothems in sandstone caves in the Beskidy Mts., Poland were dated by means of ¹⁴C on organic material (Urban et al., 2007a) and a combination of U-series and ¹⁴C methods on calcite material (Urban et al., 2007b). In both cases the speleothems are Holocene in age.

The speleothems in BPPLA probably originated as a result of carbonate mobilization at the end of the Last Glacial period. Carbonates were fixed during the Last Glacial and released when the permafrost melted and the temperature increased. Carbonates of such an origin started to precipitate in the form of scree cementation in central Bohemia as early as 13 kyr BP based on U-series dating (Žák et al., 2003).

5.3. RETREAT RATE OF CAVE WALLS AND DEPOSITION RATE OF CAVE SEDIMENTS

Preserved calcite coatings on smooth surfaces in the caves demonstrate that these cave walls (Figs. 7, 8) did not retreat more than a few millimetres since the deposition of the calcite coating. The negligible retreat of the cave walls during the Holocene accords well with the retreat < (2-5) mm/kyr of the sandstone rock overhangs in the Česká Lípa area, based on the position of Mesolithic finds (Cílek, 2007). A predominant part of the cave ceilings and walls is at present case hardened. Case hardening is assumed to be a product of a relatively warm Holocene climate (Bruthans et. al., in rev.).

Archaeological evidence (overburden thickening of the archaeological find horizons from inner parts of caves towards cave entrances, sedimentary textures) indicates that most of the cave sediment was transported to the caves from the surface. The deposition rate of cave sediments in caves during the Holocene is at least two orders of magnitude faster than the retreat rate of cave walls based on radioisotope dating. There is evidence of a recent ceiling collapse in Krtola Cave, but most of the caves display rounded smooth ceilings, which probably did not retreat more than few millimetres during Holocene.

5.4. ORIGIN OF CAVES AND ROCK OVERHANGS

Various hypotheses have been published on cave evolution. Caves have been compared to tafone (Vitek, 1987), but unlike tafone the caves penetrate much deeper into the rock. Mertlík (2006) and

Adamovič and Mikuláš (2010) proposed that caves originated from large calcareous concretions, which lose the carbonate cement due to leaching by groundwater and the subsequent excavation of loose sand. There are traces of structures, which may be interpreted as former concretions in some caves, but these concretions are mostly only a few decimetres in size and even the largest do not exceed 1 m in the study area. This hypothesis does not explain why cave spaces are much larger than concretions and how the individual concretions were linked together to form caves.

The results of radioisotope dating accord well with the idea of Cílek (2006) and Cílek and Žák (2007), who proposed that caves originated in the glacial period, by freeze/melt cycles by water infiltrating into the fractures and bedding planes in the sandstone during permafrost conditions. The major drawback with the above-mentioned hypotheses is that in the long term the infiltrated water will freeze up in the sandstone massive, sealing the fractures, and there will be thus too few freeze/melt cycles to disintegrate the sandstone. Also the very common blind upward terminations of cave passages, which are not guided by any fracture, cannot be explained since the sandstone pore space will be impermeable for water under permafrost conditions.

The remarkable similarity between the sandstone caves studied (Figs. 2, 3) and some hydrothermal karst caves (for example Sátorkö-Puszta Cave in Hungary) implies that a fluid convection mechanism may be responsible for enlarging these caves both upward and to the sides (Bruthans et al., 2009b; Fig. 11). Instead of water the air circulation might effectively transport the heat needed for freeze/melt cycles of the frozen sandstone (in the permafrost zone) during the Last Glacial. This model can explain the very common rounded shape of cave spaces and the existence of upward pockets. In the summer time the relatively warm air entered the caves, where it was cooled down by the sides of the passages releasing the heat. Cave walls could experience several tens of freeze/melt cycles per year. Sand grains from decomposed sandstone could be transported out of the caves by periodic trickles during the thaw. This hypothesis is supported by measurement of airflow in caves during summer, which the same as that is proposed by the model (Fig. 11). The model, however, is unable to explain how cavities evolved until they reached the size in which the air mass could start to circulate.

Since 2009 the sandstone erosion processes have been observed and studied in detail in Střeleč quarry, including the physical modelling of sandstone erosion in situ as well as under controlled laboratory conditions (Bruthans et al., in rev.). In terms of its properties the sandstone exposed in Střeleč quarry is very similar to the sandstone in which caves and rock overhangs are formed and the observed processes may thus be applicable to these natural outcrops. The study revealed that sandstone not protected by case

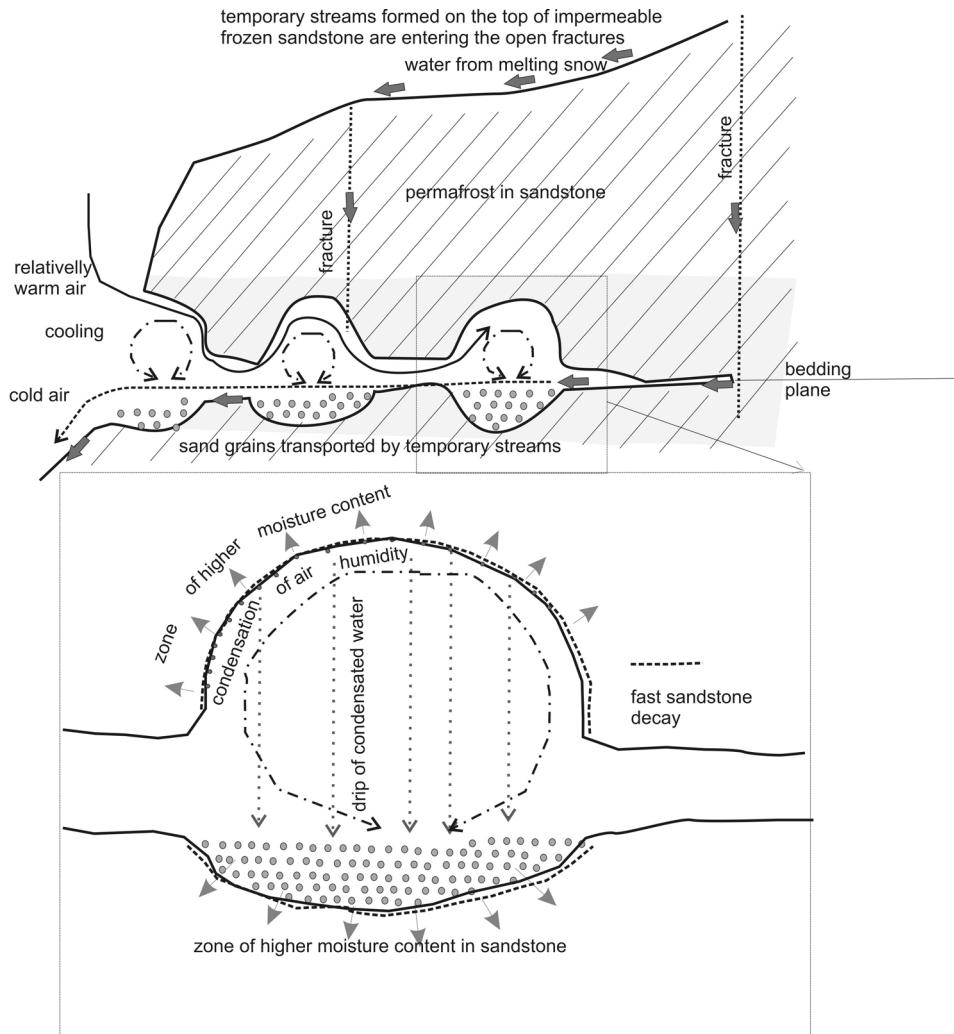


Fig. 11 Model of cave enlargement by freeze/melt cycles. Heat is transported by circulating air.

hardening is easily erodible by seeping groundwater without any previous weathering (seepage erosion; see Bruthans et al., in rev, for details). Based on these observations, the caves and rock overhangs were probably most intensively forming at the end of the Last Glacial. The groundwater might have accumulated above the receding permafrost, flowed laterally, and seeped out to the surface in a position high above the valley floors. Cavities could be potentially formed very fast (over a few centuries or even less) based on the rapid erosion rate observed in the Střeleč quarry (Bruthans et al., in rev.). Once the impermeable permafrost melted, the groundwater started to seep deeply into the sandstone, the caves and rock overhangs dried out, and their enlargement ceased. The exposed rock surfaces were hardened in a relatively warm climate and caves and rock overhangs were preserved in their present shape. At the present time, most of the cave wall and ceiling surfaces are protected from erosion by case hardening. In several caves, however, the sandstone is still soft

and small trickles of water have been observed eroding sandstone.

The above-mentioned erosion processes, which might have occurred during the Last Glacial or at the end of the Last Glacial, can explain why there is just one site out of 170 caves and rock overhangs with archaeological finds which contains Palaeolithic finds. Sandstone caves might be much older than the Last Glacial, but most of their fills were eroded during or at the end of the Last Glacial period (Cílek and Zák, 2007). The seepage erosion model seems to be most probable explanation of cave and rock overhangs origin. It fits well with radioisotope dating results and archaeological evidence. Unlike other models it is supported by results of physical modelling using real sandstone.

6. CONCLUSION

In several sandstone caves in the Bohemian Paradise calcite speleothems have been found which have enabled radiometric dating. Speleothems cover

part of the cave ceilings and walls to a thickness of about 1 cm. They are formed by low Mg calcite and consist of calcite moonmilk and coraloids. In Krtola Cave carbonates occurs in zone cementing cave sediments, which are up to 15 cm thick.

The stable isotope composition (C and O) of speleothems in the Krtola and the Petráskova caves correspond to values characteristic for cave speleothems in Central Europe. The isotopic composition of speleothems in the Klokočské Skály area indicates evaporation and fast carbon dioxide escape during carbonate precipitation.

The apparent radiocarbon age of speleothems in Krtola Cave indicates deposition at least 8 kyr BP. The minimum age of speleothem deposition is between 5 and 8 kyr BP in the Sintrová, Mrtvé Údolí and U Studánky caves. Based on the age of the speleothems and archaeological finds in cave sediments the minimum age of the onset of cave and rock overhang development is the end of the Last Glacial. The dated cave walls did not retreat even a few mm in the last 5-8 kyr and are thus virtually not evolving under recent climatic conditions. Most of the cave ceilings and walls are at present indurated by hardened surfaces, which protect the sandstone from erosion and the caves and rock overhangs are predominantly dry.

Sandstone caves probably intensively evolved either during or at the end of the Last Glacial period. There are two different erosion mechanisms which might have formed/reshaped the caves and rock overhangs at that time: A) In the case of permafrost conditions: Repeated freeze/melt cycles affecting sandstone pore space followed by the transport of fallen sand grains by minor temporary trickles. We expect that heat was transmitted by air circulating between the cave and the surface; B) Seepage erosion of sandstone during the melting of permafrost, prior forming of case hardening.

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