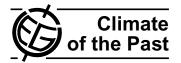
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Coarsely crystalline cryogenic cave carbonate – a new archive to estimate the Last Glacial minimum permafrost depth in Central Europe

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Abstract. Cryogenic cave carbonate (CCC) represents a specific type of speleothem whose precipitation is triggered by freezing of mineralized karst water. Coarsely crystalline CCC, which formed during slow freezing of water in cave pools, has been reported from 20 Central European caves located in Germany, the Czech Republic, Slovakia and Poland. All these caves are situated in an area which was glacierfree during the Weichselian. Whereas the formation of usual types of speleothems in caves of this region usually ceased during the glacials, coarsely crystalline CCC precipitation was restricted to glacial periods. Since this carbonate type represents a novel, useful paleoclimate proxy, data from its Weichselian occurrences in caves in Central Europe were collected, including their C and O stable isotope values, Useries ages and depth below the surface. When using only the CCC data from caves with limited cave ventilation, the permafrost depths of the Weichselian can be estimated to be at least 65 m in the lowlands and uplands. An isolated CCC find indicates that Weichselian permafrost penetrated to a depth of at least 285 m in the High Tatra mountains, Slovakia. A model of the formation of coarsely crystalline CCC assumes its formation especially during periods of permafrost thawing. U-series data confirm that permafrost depth changed and CCC precipitation in deep caves occurred repeatedly in the studied area during marine isotope stages 4, 3 and 2. One important phase of coarsely crystalline CCC formation related to permafrost thawing occurred between 40 and 21 ka BP, and the last phase of its formation was related to the final permafrost destruction between 17 and 12 ka BP.

1 Introduction

The southern limit of the continental glaciation of the central part of Europe during the Last Glacial (Weichselian) roughly followed the line defined by the present-day cities of Hamburg-Berlin-Poznań-Warszawa-Minsk (Poser, 1948; Svendsen et al., 2004). During the coldest phases of the Weichselian, a permafrost zone developed in the glacierfree area along the southern limit of the continental glacier (Frenzel et al., 1992; Vandenberghe, 2001; Czudek, 2005). Permafrost is defined as ground (rock or soil, including ice and/or organic material) that remains at or below 0°C for at least two consecutive years (cf. Permafrost Subcommittee NRC Canada, 1988; Dobiński, 2011). For this glacier-free zone located between the northern continental and the Alpine mountain glaciations, little information has been available about the distribution, duration and thickness of permafrost during the Weichselian. It is usually assumed that continuous permafrost existed in the northern part of this zone during the Last Glacial Maximum (LGM), while discontinuous or sporadic permafrost was probably typical for more southerly located areas in the lowlands (Frenzel et al., 1992; Czudek, 2005).

Caves represent a subsurface environment which has been so far rarely used for the estimation of past permafrost depths. The effects of past freezing temperatures in caves are indicated by several kinds of observations. Typical results of freezing temperatures include cave roof frost shattering, features produced by frost and ice action in unconsolidated cave sediments (i.e. various kinds of size-sorting phenomena and patterned ground phenomena), speleothem damage by frost and ice action, and other phenomena. These features have been reported from several caves in Central and Western Europe by numerous authors (Kyrle, 1929–1931; Kempe, 1989, 2004, 2008; Pielsticker, 1998; Wrede, 1999; Pielsticker, 2000; Kempe and Rosendahl, 2003; Kempe et al., 2006; Lundberg and McFarlane, 2012). The main difficulty in interpreting these observations results from the fact that some of these frost indicators can also be produced by other processes than frost and ice action. A further problem in relating them to surface climate change is represented by the difficulty in their dating. Probably the best evidence for former ice filling of cavities is represented by fragments of speleothems, called "ice attachments", attached by carbonate cement to steep sections of cave walls at sites where they could only be fixed due to the support of ice filling the cavity (or some other type of sediments filling the cavity at one time). These features were thoroughly studied in Germany (see references above), where extensive evidence of past ice filling of cavities has been collected.

Another indicator of past temperatures at the freezing point in caves is represented by cryogenic cave carbonate (CCC). CCC is generally formed by precipitation triggered by freezing of water. Salts are generally poorly soluble in ice, with solubility in the range of micromoles or less (e.g. Gross et al., 1977; Petrenko and Withworth, 1999). Freezing of aqueous salt solutions, therefore, results in a process called salt (brine) rejection (for carbonate solutions, see Killawee et al., 1998). Freezing-induced segregation of solutes leads to a significant increase in salt concentration in the residual (unfrozen) portion of the solution. As freezing proceeds, some dissolved compounds reach their saturation limit and precipitate as cryogenic minerals. Released gases either escape into the atmosphere or are concentrated in bubbles entrapped in the ice. Some other dissolved compounds (e.g. Na⁺ and Cl⁻) form small inclusions of unfrozen, highly concentrated brine. These inclusions are usually distributed along ice crystal boundaries. The resulting ice is a complex multi-phase substance, which contains solid, liquid and gaseous phases (Mulvaney et al., 1988; Souchez and Lorrain, 1991). Depending on the freezing rate and the thickness of the freezing water layer, either fine-grained CCC, or coarsely crystalline CCC (a subject to the present study) can be formed.

Coarsely crystalline CCC can be identified in caves based on its typical mode of occurrence and characteristic crystal and aggregate morphology. Its origin can be unequivocally confirmed by U-series dating fitting to glacial periods in combination with unique C and O stable isotope systematics (Žák et al., 2004, 2008; Richter and Riechelmann, 2008). Whereas the formation of conventional types of speleothems in caves in Central Europe mostly ceases during the glacials, CCC particularly forms during these periods, thus representing a new climate archive providing evidence for freezing conditions in the subsurface. Besides caves, cryogenic carbonates occur commonly in numerous surface and near-surface environments, e.g. carbonates formed at the base of glaciers (subglacial calcite), in freezing groundwater springs (including naled, aufeis), in freezing lakes, in soils of cold environments, etc. These other types of cryogenic carbonates (reviewed in Žák et al., 2004) are not discussed in detail here.

The principal aim of this paper is to review available data on the occurrence of coarsely crystalline CCC in deep caves in Central Europe and to significantly extend the existing CCC age determinations. The transfer of surface climate signal into caves is also discussed, including the influence of cave ventilation. These data, in combination with the depth below the surface of each site, are interpreted from the viewpoint of the Weichselian minimum permafrost depth penetration and periods of permafrost existence and destruction.

2 CCC and its environmental setting

CCC is found in caves in two modes of occurrence, representing two different types of environment of its formation. Fine-grained cryogenic carbonate powder is rather common, formed during rapid freezing of thin water layers in strongly ventilated caves or near-entrance cave sections (e.g. Kunský, 1939; Viehmann, 1960; Ek and Pissart, 1965; Savchenko, 1976; Lauriol et al., 1988, 2006; Pulina, 1990; Clark and Lauriol, 1992; Žák et al., 2004, 2008; Spötl, 2008; Lacelle et al., 2009; May et al., 2011). These powder CCC types are of no use for the estimation of past permafrost depths and are not discussed in this paper.

The second mode of occurrence of CCC is represented by much larger crystals and crystal aggregates (ranging from < 1 mm to about 40 mm), which were formed by segregation of the dissolved load during slow water freezing in pools. The modern formation of these coarsely crystalline CCC types has never been observed in caves. Coarsely crystalline CCC of clearly Holocene origin has been reported only once (Onac et al., 2011), having the form of glendonite-like and up to 4 cm large aggregates found in the ice of the Scărișoara Ice Cave, Romania. The Weichselian coarsely crystalline CCC has been found in 20 caves located in the territories of Germany, the Czech Republic, Slovakia and Poland. These specific carbonate forms have been recorded in caves in this area since the 1950s (Skřivánek, 1954; Tulis and Novotný, 1989; Urban and Złonkiewicz, 1989; Erlenmeyer and Schudelski, 1992; Schmidt, 1992; Durakiewicz et al., 1995), but their formation processes were not fully explained until the papers of Žák et al. (2004) and Richter and Niggemann (2005). Since

then, coarsely crystalline CCC has been discovered in other caves in the periglacial zone of the Last Glacial in Europe (Orvošová and Žák, 2007; Richter and Riechelmann, 2008; Richter et al., 2008, 2009a,b,c, 2010a,b, 2011, 2012; Žák et al., 2008, 2011; Meissner et al., 2010). This specific type of speleothem frequently occurs in caves with limited or practically no ventilation. The only way for these cavities to be cooled down to freezing conditions was, therefore, the occurrence of permafrost.

2.1 Morphology and mode of occurrence of coarsely crystalline CCC

Coarsely crystalline CCC usually occurs in the form of accumulations of loose crystals and crystal aggregates freely deposited on the surface of the bottom of the cavity, being only rarely covered by younger Holocene speleothems and/or clastic sediments. Its formation, therefore, represents one of the youngest events in the evolution of the studied caves. The coarsely crystalline CCC accumulations (several cm thick, at maximum) usually cover an area of up to several square meters, typically located in central parts of larger cavities; they are usually absent in narrow corridors or near cavity walls. CCC of this type never occurs near cave entrances or in a cave zone showing significant seasonal temperature changes. Instead, it is typically found in more remote cave sections.

The morphology of coarsely crystalline CCC is complex and differs from the usual secondary carbonate infillings and speleothems. Its morphological variability led to the development of many local terms used for the description of the main morphological types in individual countries. Generally, coarsely crystalline CCC types can be divided into several morphological, broadly defined endmembers, each with a typical position in the sequence of crystallization, and most probably slightly differing in the environment of their origin (see Sect. 2.2 for stable isotope data). Three most common morphological types include: (i) single crystals and their random or organized aggregates/intergrowth, (ii) crystalline raft-like aggregates, and (iii) fine to coarsely crystalline spherical/globular forms.

The single crystals are usually small (several tenths of millimeter to several millimeters) but relatively well-developed. Their stellate aggregates up to approximately 4 cm in size have also been documented (Urban and Złonkiewicz, 1989). More or less regular rhombohedra and, less frequently, scalenohedra are the most commonly detected crystal forms. The crystals are usually not perfectly developed, and various more or less skeletal, hopper- or lath-shaped, fan-like forms and their various intergrowths are common. They are called "roses", "wreaths" and "twigs" in the local literature (Durakiewicz et al., 1995).

Raft-like aggregates are characterized by their generally flat appearance and by crystal arrangements very similar to classical floating carbonate rafts in non-iced caves (cf. Hill and Forti, 1997, p. 88). The basic building elements of cryogenic rafts usually consist of calcite scalenohedra, sometimes elongated in the direction of the vertical axis. The size of these crystals varies from several tens of micrometers to several millimeters. Crystals of two generations have been observed at some occurrences of cryogenic raft samples (Na Javorce Cave, Czech Republic, Žák et al., 2011).

Spherical or hemispherical forms representing the third type are described using various terms: "Zopfsinter" (Erlenmeyer and Schudelski, 1992), "hemispheres" (Tulis and Novotný, 1989; Forti, 1990), "cave caps" (Hill and Forti, 1997), or "cupula-spherolites" (Richter and Riechelmann, 2008). Coarsely crystalline CCC of this type displays either smooth or finely to coarsely crystalline surfaces, and occurs as separate bodies or their random (sometimes planar) intergrowths/accumulations. Also, it often overgrows other types of coarsely crystalline CCC. Examples of the oriented intergrowth of two spherical bodies have also been frequently documented. Hemispheres (usually approximately half a sphere) with clearly visible crystallinity and more or less exerted crystal edges and corners are usually composed of tightly intergrown, radially arranged crystals. Photographs of typical field occurrences and the most common crystal aggregate morphologies are contained in the Supplement.

Based on the field observations, the model of CCC formation, and stable isotope studies (Žák et al., 2004), it seems highly probable that the sequence of formation of these aggregates starts with sharp-edged aggregate types composed of rhombohedra, while the smooth-surface types are formed later in the precipitation sequence. The coarsely crystalline CCC precipitation sequence is controlled by increasing bulk salinity of the residual unfrozen portion of the solution during progressive freezing and by slightly decreasing temperature. The isotopically most evolved type (showing the lowest δ^{18} O and the highest δ^{13} C values within each site, see Sect. 2.2), formed during the final stage of freezing-induced crystallization, is represented by more-or-less regular hemispheres. While the early rhombohedral and scalenohedral forms were probably already initially deposited as calcite, the smooth-surface types could have been originally formed by unstable carbonate phases like ikaite or vaterite (Killawee et al., 1998; Lacelle et al., 2009). They were probably all converted to calcite during the warmer Holocene climate.

2.2 C and O stable isotope systematics

Precipitation of carbonate minerals accompanied by water freezing exhibits large-scale isotope fractionations, which have been frequently studied (Clark and Lauriol, 1992; Durakiewicz et al., 1995; Fairchild et al., 1996; Killawee et al., 1998; Žák et al., 2004, 2008, 2009, 2011; Richter and Niggemann, 2005; Lacelle et al., 2006, 2009; Orvošová and Žák, 2007; Lacelle, 2007; Spötl, 2008; Richter et al., 2008, 2009a,b,c, 2010a,b; Richter and Riechelmann, 2008; Meissner et al., 2010; May et al., 2011; Onac et al., 2011). In the usual δ^{13} C vs. δ^{18} O plot, data obtained on various types of

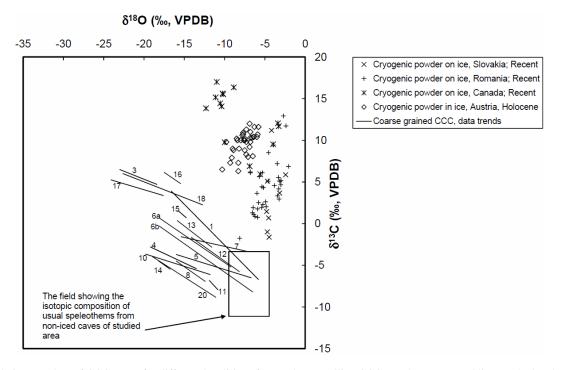


Fig. 1. Stable isotope data of CCC. Data for different localities of coarsely crystalline CCC are shown as trend lines, calculated using linear regression. The regression lines were calculated for all analyses available from a given locality (single crystal bulk analyses, single aggregate bulk analyses, point analyses center to rim in larger crystals and crystal aggregates). The trends are directed towards lower δ^{18} O and higher δ^{13} C values as freezing proceeds. Locality numbers are identical with those in Fig. 1 and Table S1 in the Supplement (Localities 2, 9, and 19 are not covered by published stable isotope data). Data sources: 1 – Apostelhöhle, Germany (Richter et al., 2010b); 3 – Glaseishöhle, Germany (Richter et al., 2009c); 4 – Großen Sunderner Höhle, Germany (Richter et al., 2009a); 5 – Heilenbecker Höhle, Germany (Richter et al., 2009b); 6a – Herbstlabyrinth, Germany, Rätselhalle (Richter et al., 2010a), 6b – the same cave, Weihnachtsbaumhalle (Richter et al., 2011); 7 – Malachitdom Cave, Germany (Richter and Niggemann, 2005; Richter and Riechelmann, 2008); 8 – Ostenberg Höhle, Germany (Richter and Niggemann, 2005); 10 – BUML Cave, Czech Republic (Žák et al., 2011); 11 – Javoříčko Caves, Czech Republic (Žák et al., 2011); 12 – Na Javorce Cave, Czech Republic (Žák et al., 2011); 13 – Novoroční Cave (Žák et al., 2011); 14 – Portálová Cave, Czech Republic (Žák et al., 2011); 15 – Hačova Cave, Slovakia (Žák et al., 2011); 16 – Mesačný tieň Cave, Slovakia (Žák et al., 2011); 17 – Stratenská Cave, Slovakia (Žák et al., 2004); 18 – Cold Wind Cave, Slovakia (Žák et al., 2009); 20 – Jama Chelosiowa–Jaskinia Jaworznicka, Poland (Žák et al., 2004). The data for fine-grained CCC, i.e. the cryogenic cave powder, are shown for comparison. Fine-grained CCC data are from Clark and Lauriol (1992), Žák et al. (2004, 2008, 2011), Lacelle et al. (2009) and Spötl (2008).

CCC cover a wide range from -2 to -24 % VPDB in δ^{18} O and -11 to +17 % VPDB in δ^{13} C. Their variability is obviously much larger than the variability in stable isotope data of speleothems which do not form due to freezing conditions (Fig. 1). Potential isotope fractionations (probably very small or none) related to conversion from possibly originally present unstable carbonate phases to calcite are not known and, therefore, not evaluated.

The C and O stable isotope systematics of individual CCC types depends on the dominance of either disequilibrium (largely kinetic) or equilibrium isotope fractionation, which is controlled by the precipitation rate (cf. DePaolo, 2011). The precipitation rate is controlled by several partly interdependent factors, such as thickness/depth of the layer of freezing water, freezing rate, ventilation of the cave, etc. The kinetic isotope fractionations largely dominate during the formation of the fine-grained cryogenic carbonate powder. Rapid freezing of thin water films and ice sublimation both lead to a faster supersaturation of the carbonate solution and thus to higher precipitation rates. The stable isotope data trends of the CCC powder in the δ^{13} C vs. δ^{18} O diagram are similar to those for speleothems deposited during evaporation of water or during rapid kinetic degassing of CO₂ from the drip water (Fig. 1; cf. Hendy, 1971; Mickler et al., 2006; Dreybrodt and Scholz, 2011).

In the case of the coarsely crystalline CCC formed by slow water freezing in pools, the dominant factor controlling δ^{18} O values is equilibrium oxygen isotope fractionation between water and ice, which leads to progressive lowering of the oxygen isotope values (δ^{18} O_w values) of the residual, yet unfrozen solution (O'Neil, 1968; Jouzel and Souchez, 1982; Souchez and Jouzel, 1984; Lehmann and Siegenthaler, 1991). The stable isotope data trends for coarsely crystalline CCC illustrated in Fig. 1 indicate that the carbonate δ^{18} O

values reflect the $\delta^{18}O_w$ values. To achieve this, the precipitated carbonate needs to be in oxygen isotope equilibrium with H₂O. Hence, the dissolved bicarbonate and carbonate ions (HCO₃⁻, CO₃²⁻) are in oxygen isotope equilibrium with H₂O, which is caused by the buffering reaction (Dreybrodt, 2008; Scholz et al., 2009; Dreybrodt and Scholz, 2011), i.e. the oxygen isotope exchange between H₂O and HCO₃⁻. According to Dreybrodt and Scholz (2011), at 0 °C it takes about 5.8 days for the oxygen isotope equilibration of HCO₃⁻ (approx. 98 % of equilibrium) with H₂O. This implies that the precipitation rate was very slow, because otherwise the oxygen isotope fingerprint of H₂O would be altered by isotope disequilibrium effects, and that the isotope equilibrium fractionation probably occurred during the formation of coarsely crystalline CCC.

The δ^{13} C vs. δ^{18} O plot shows an increase in δ^{13} C values with the decrease of δ^{18} O values (cf. Fig. 1). This indicates that there are no CO₂ reservoirs within the cavities. Therefore, the dissolved carbon ions become enriched in ¹³C due to progressive carbonate precipitation. Stable isotope data trends in this direction in the δ^{13} C vs. δ^{18} O plot are generally rare in nature. Localities of coarsely crystalline CCC found in larger cavities usually exhibit higher δ^{13} C values than those occurring in smaller and more isolated cavities. Since coarsely crystalline CCC is formed under closed (or semi-closed) system conditions and cryogenic powders under open system conditions, the different stable isotope trends of these two types can be explained easily (Fig. 1, cf. Žák et al., 2008).

It should be mentioned that the coarsely crystalline CCC is formed during δ^{18} O changes in the solution, and that carbonate δ^{18} O data as such are not the carrier of paleoclimatic information. Paleoclimatic information of this novel archive is given only by the presence and the age of coarsely crystalline CCC (which is a proof of freezing temperature in the cave).

2.3 Regional distribution of the known coarsely crystalline CCC sites

Central European caves with occurrences of coarsely crystalline CCC are located in a belt defined by latitudes between $47^{\circ}30'$ and $51^{\circ}30'$ N and longitudes between $7^{\circ}30'$ and $20^{\circ}30'$ E. Details for all studied caves including the references to the original publications are listed in Table S1 in the Supplement. The locations of the caves are also shown in Fig. 2. The entrance elevations are mostly in the range between 175 and 500 m a.s.l. (hilly regions). A small number of the studied caves are located in mountainous areas with entrance elevations between 690 and 2230 m a.s.l.

The studied caves are grouped in several karst regions. In Germany, a group of eight caves containing coarsely crystalline CCC is located in the "Rheinisches Schiefergebirge", with morphologically complex caves formed mostly by corrosion in the phreatic or epiphreatic zone in Devonian limestones. Caves dominated by vadose morphology are less common here (represented by some parts of the Herbstlabyrinth–Advent Cave System). The entrance elevations of these caves range from 175 to 501 m a.s.l. One German cave with coarsely crystalline CCC occurrence (Riesenberghöhle) is located in Jurassic limestones near the city of Hannover, and one mountain chasmal cave (Glaseishöhle), with an entrance elevation of 2230 m a.s.l., is formed in the Late Triassic limestone (Dachsteinkalk) of the Northern Calcareous Alps (Fig. 2).

In the Czech Republic, the studied caves are mostly located in the Bohemian Karst, a small karst region developed in Silurian and Devonian limestones close to the city of Prague. Coarsely crystalline CCC samples were collected from four caves with entrance elevations between 244 and 402 m a.s.l. The caves in this region show a complex morphology (phreatic and/or epiphreatic maze caves are abundant). Several caves bear evidence of hydrothermal processes in the early stages of their evolution. One isolated locality in the Czech Republic (Javoříčko Caves, Fig. 2) with paleofluvial epiphreatic to vadose levels is located in Devonian limestones in Central Moravia, with entrance elevations between 445 and 475 m a.s.l.

In Slovakia, the studied caves are generally located in the mountainous regions of the Western Carpathians and developed in various types of Mesozoic limestone. The entrance of morphologically complex and partly chasmal Hačova Cave, developed in Middle Triassic limestones, lies at 690 m a.s.l. in the Lesser Carpathians, north of Bratislava (Šmída, 2010). Other localities are found in mountain ranges with entrance elevations between 995 and 1767 m a.s.l. (Orvošová and Vlček, 2012). The coarsely crystalline CCC sites in all these caves occur hundreds of meters away from the cave entrances.

In Poland, only a single cave system with several coarsely crystalline CCC sites has been found so far. The extensive Jama Chelosiowa–Jaskinia Jaworznicka Cave System, located close to the town of Kielce, contains at least twelve CCC occurrences. The caves are of complex phreatic and epiphreatic morphology and formed in Devonian limestones below a small hill in a relatively flat country, partly covered by a thin blanket of clastic Early Triassic sediments. The cave entrance elevations range from 257 to 288 m a.s.l.

Cross sections (side projections) of selected localities with the positions of the studied sites are shown in Fig. 3. The figure compiles the localities which contain the deepest known CCC sites and the caves discussed in more detail.

3 Materials and methods

Cave maps and cave sections were obtained from the original publications as well as from local caving clubs. For the construction of cave sections, the surface morphology was derived from detailed topographical maps (scale 1 : 10 000).

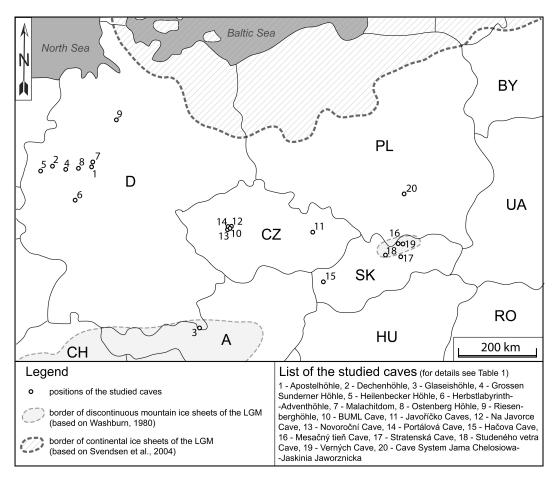


Fig. 2. Locations of the studied caves in Central Europe. The limits of continental and Alpine mountain glaciation during the Last Glacial are taken from Washburn (1980) and Svendsen et al. (2004).

Both the cave maps and surface shape above the caves were in several cases verified or measured more precisely by the authors, using common mapping methods for underground and surface areas. All the known or newly discovered coarsely crystalline CCC sites were then indicated on the cave maps and sections. In order to show the relationships to the surface, the cave sections used are generally side projections (projected profiles), not extended profiles (cf. Palmer, 2007, p. 12). Depths (distances from the closest surface above the site) were then estimated from these cave sections. The studied sites were field documented and photo documented. C and O stable isotope data were collected from the original publications (see description of Fig. 1 for data sources).

²³⁰Th/U-dating was performed both by thermal ionization mass spectrometry (TIMS) and by multi-collector inductively coupled plasma mass spectrometry (MC-ICPMS). TIMS samples were prepared similarly as described in Frank et al. (2000). U-series isotopes were measured with the MAT 262 RPQ TIMS at the Heidelberg Academy of Sciences, Germany, using the double filament technique. MC-ICPMS samples were prepared similarly as described in Hoffmann et al. (2007) and analyzed at the Max-Planck-Institute for Chemistry (MPIC), Mainz, Germany. The mixed ²³³U-²³⁶U-²²⁹Th spike used at MPIC was calibrated against a gravimetric U standard solution and a secular equilibrium standard solution. The gravimetric U standard solution was prepared from a NBL-112a metal bar and also used to calibrate the U-Th spike used at the Heidelberg Academy of Sciences (Hoffmann et al., 2007). The ²²⁹Th concentration of the spike was calibrated against a secular equilibrium standard solution prepared from a 2 Ma old speleothem sample from Wilder Mann Cave, Austria (Meyer et al., 2009), which has been shown to be in secular equilibrium (Hoffmann et al., 2007). The calibration was checked using another secular equilibrium solution (Harwell Uraninite, HU) that was used to calibrate the U-Th spike used at the Heidelberg Academy of Sciences (Hoffmann et al., 2007). Analytical MC-ICPMS techniques involved a standard-sample bracketing procedure to derive correction factors for mass fractionation and Faraday cup to ion counter gain, similarly as described in Hoffmann et al. (2007) and Jochum et al. (2011).

All activity ratios reported for both laboratories were calculated using the decay constants from Cheng et al. (2000)

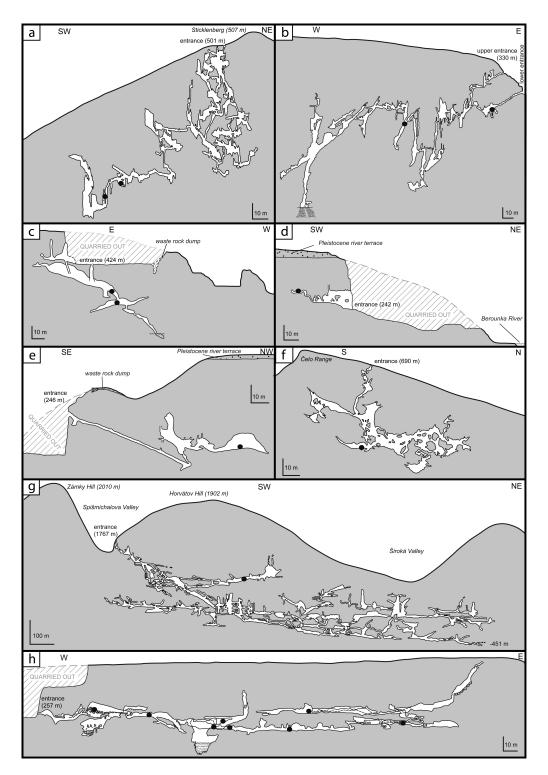


Fig. 3. Cave sections (projected profiles) of selected studied caves. Coarsely crystalline CCC locations are indicated by black dots. (**a**) Apostelhöhle, Germany: mapping by M. Erlenmeyer and co-workers, section after Richter et al. (2010b); (**b**) Na Javorce Cave, Czech Republic: modified and redrawn after Dragoun et al. (2011), mapping by J. Dragoun, J. Vejlupek and J. Novotný; (**c**) Malachitdom Cave, Germany: mapping and section after Erlenmeyer and Schudelski (1992); (**d**) Portálová Cave, Czech Republic: original drawing, mapping by J. Živor and co-workers; (**e**) BUML Cave, Czech Republic: original drawing, mapping by S. Martínek and P. Zbuzek; (**f**) Hačova Cave, Slovakia: original drawing by B. Šmída, mapping by B. Šmída, M. Hačo and co-workers; (**g**) Mesačný Tieň Cave, Slovakia: original drawing by B. Šmída, I. Pap and co-workers; (**h**) Jama Chelosiowa–Jaskinia Jaworznica Cave System, Poland: mapping and section drawing by J. Gubała and A. Kasza, see also Urban and Rzonca (2009).

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and corrected for detrital Th assuming a bulk Earth 232 Th/ 238 U weight ratio of 3.8 for the detritus and 230 Th, 234 U and 238 U in secular equilibrium. All ages are reported in yr BP, i.e. before the year 1950.

4 Results and discussion

Stable isotope data for individual coarsely crystalline CCC locations are shown in Fig. 1. The field relationships, i.e. the depths of the studied sites below the closest surface, are presented in selected cave cross sections (Fig. 3), and summarized in Table S1 in the Supplement. Table S1 also contains all available U-series ages of the coarsely crystalline CCC samples, with the data sources identified. New TIMS and MC-ICPMS U-series data are presented in Tables S2 and S3, respectively, both contained in the Supplement.

4.1 Evidence for cryogenic origin of the coarsely crystalline CCC

All the studied coarsely crystalline CCC samples exhibit a typical mode of occurrence in the field, ranking them among very young events in the evolution of the studied caves (see Sect. 2). The mode of occurrence of the crystals and crystal aggregates (see description in Sect. 2.1) indicates that they were not formed directly at the cavity bottom. In such a case they should be firmly attached to the cave bottom, like usual speleothems, which was not observed for the samples of this study. In the models of coarsely crystalline CCC formation (e.g. Žák et al., 2004, 2008; Richter and Riechelmann, 2008), it is generally supposed that the crystals and crystal aggregates were formed in slowly freezing water pools, located on the surface of the ice fill of the cavity. It can be expected that within a cavity located in the permafrost zone, freezing of the water proceeds simultaneously from the cavity bottom, cavity walls, as well as from the pool surface. The final stages of water freezing accompanied by the most intensive mineral precipitation can in fact occur in a lens of liquid water locked completely in the interior of the ice fill. After final permafrost and cave ice melting, all types of precipitates are mixed and deposited together on the cavity bottom, forming a loose accumulation.

C and O stable isotope data for each locality exhibit a typical trend (Fig. 1) towards extremely low δ^{18} O values and slightly elevated δ^{13} C values as precipitation proceeds (see above in Sect. 2.2). The negative relationship between δ^{18} O and δ^{13} C values is generally rare for common speleothems and is in agreement with the slow equilibrium freezing model (see above in Sect 2.2). The formation of coarsely crystalline CCC at low temperatures is further indicated by their Useries ages, which suggest (with one exception) the formation within glacial or stadial periods. Cryogenic formation of the studied coarsely crystalline CCC is therefore in agreement with the mode of occurrence, C and O stable isotope geochemistry and formation during the glacial. The cryogenic origin of the studied samples can be considered as well demonstrated.

4.2 Ventilation of the studied caves

The typical environment of the occurrence of coarsely crystalline CCC is represented by cave sections remote from the entrances. Most caves with the studied occurrences are morphologically complex maze systems, open to the surface only via rather small entrances and/or very narrow and zigzagshaped corridors. These caves maintain almost constant temperatures throughout the year, which are close to the presentday mean annual air temperature (MAAT). In several cases, the caves hosting coarsely crystalline CCC were discovered as isolated cavities by limestone quarrying, having no open connection to the surface (i.e. all connections were completely filled by pre-Weichselian sediments). In several other cases, the cavities hosting coarsely crystalline CCC were discovered by cavers digging through the pre-Weichselian sediments that originally completely filled the corridors. Cooling of these cavities to subzero temperatures by air circulation was impossible.

Only three localities are exceptions in this context. The chasmal Glasseishöhle (Northern Calcareous Alps) could have also been cooled by air exchange and not by permafrost evolution (present-day seasonal freezing inside the cave reaches a depth of 40 m, the studied site occurs at a depth of 229 m). Recent temperatures slightly lower than the MAAT are also typical for the corridor hosting coarsely crystalline CCC in the Stratenská Cave System, Slovakia. This inclined corridor leads above the studied site through a rock collapse in the direction towards a large collapse structure, which is open to the surface and accumulates dense cold air. Finally, stronger cave winds are also characteristic for some sections of the Hačova Cave, Slovakia, but most of this cave has recent temperatures close to the MAAT. In summary, for 17 of the 20 described sites, the only way to cool the cavities hosting the coarsely crystalline CCC was through the evolution of permafrost.

4.3 Age and model of formation of the studied CCC

In total, 44 U-series ages on coarsely crystalline CCC are currently available (Table S1 in the Supplement, Fig. 4). Of these 44 dates, 41 (i.e. more than 93%) fall within the Weichselian between 104.0 and 11.9 ka BP. The age data are not distributed randomly within the Weichselian, but show several age clusters, each age cluster commonly containing samples derived from different caves of the study area. Ages from the first half of the Weichselian started at approximately 71 ka BP (cf. Fig. 4). The oldest date within the Weichselian (104 ± 2.9 ka BP) falls into a relatively warm interval of marine isotope stage 5c (general introduction to MIS can be

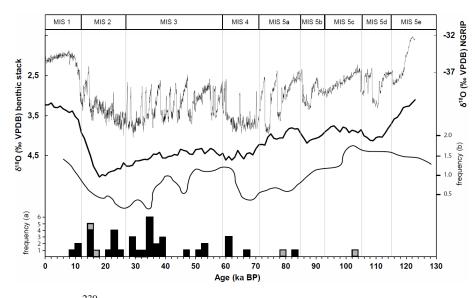


Fig. 4. Frequency distribution of 42 230 Th/U age determinations of coarsely crystalline CCC from Germany, the Czech Republic, Slovakia and Poland for the last 130 ka. Age data from Table S1 in the Supplement are shown as histogram with 2 ka intervals. CCC ages of samples from high-altitude caves showing present temperature below 4 °C are shown as grey squares, other data as black squares (a). The northwestern Europe common speleothem and travertine growth frequency record (b) is after Baker et al. (1993). The stacked benthic O isotope record of Lisiecki and Raymo (2005) (thick line) serves as a reference curve for global glacial–interglacial climate variability. For the Northern Hemisphere climate, the NGRIP record is shown (North Greenland Ice Core Members, 2004; thin line). Time boundaries of marine oxygen isotope stages shown in the uppermost part of the figure are after Wright (2000), with some later modifications.

found in Wright 2000). It was obtained for CCC sample from the Cold Wind Cave, Slovakia, located at a high elevation and a shallow depth below a mountain range. Dates from the first half of the Weichselian are more common for locations with higher elevations or with more northerly positions within the study area (Cold Wind Cave, Slovakia; Jama Chelosiowa– Jaskinia Jaworznicka, Poland; Riesenberghöhle, Germany; see data in Table S1 in the Supplement).

The period with the most frequent occurrence of coarsely crystalline CCC ages between ~ 40 and ~ 21 ka BP corresponds to the younger, climatically unstable part of the MIS 3 and the first part of MIS 2. It is expected that this climatically unstable period induced oscillations in the permafrost depth, creating conditions suitable for coarsely crystalline CCC formation. Age data corresponding to the LGM (~ 26 to \sim 19 ka BP; Clark et al., 2009) are generally scarce, being represented only by ages for the Stratenská Cave, Slovakia. The Stratenská Cave is one of three caves within the studied set which could have also been cooled by air circulation. Another important group of dated samples from several caves is represented by ages between 17.0 and 11.9 ka BP, corresponding to periods of variable climate after the LGM and before the beginning of the Holocene when the permafrost thickness was generally decreasing.

Of the total data set of 44 age determinations, only three (less than 7 %) do not fit in the Weichselian. Two older dates (Apostelhöhle, Germany, 408^{+42}_{-31} ka BP; Cold Wind Cave, Slovakia, 180 ± 6.3 ka BP) fall within the younger parts of

the Elsterian and Saalian glacials, respectively. Findings of coarsely crystalline CCC from earlier glacials are rare, probably because these old accumulations were covered during later cave evolution by clastic sediments or flowstone. From the whole data set, only one date corresponds to Holocene age $(9.191 \pm 0.072 \text{ ka BP})$; Na Javorce Cave, Bohemian Karst). This outlier may be interpreted in two ways. Either the U-series system of the sample was not closed (and the sample post-depositionally adsorbed U during the Holocene; the ²³⁴U/²³⁸U activity ratio of this sample is by far the highest in the whole data set), or the sample age indeed indicates the existence of relic permafrost in the Early Holocene. The first interpretation is supported by the sample character (aggregates of relatively small crystals with large surface area), the second by the position of the sampling site, which is the deepest (65 m below the surface) among the low altitude localities.

A comparison of the U-series ages with the benthic δ^{18} O stack from Lisiecki and Raymo (2005) and with NGRIP climate record (North Greenland Ice Core Project Members, 2004) is shown in Fig. 4. The obtained ages mostly plot near transitions from colder to warmer periods. This is in agreement with the published models of the formation of ice fill in cavities within permafrost (Pielsticker, 2000) and coarsely crystalline CCC formation in these cavities (Richter and Riechelmann, 2008; Richter et al., 2010b). Deeper infiltration of groundwater is practically impossible during permafrost growth since infiltration is eliminated due

to blocking by ice. Groundwater movement is mostly restricted to the near-surface permafrost active layer during these periods.

Deeper circulation of either highly mineralized groundwater within the permafrost or common groundwater in taliks is possible in karst environments (Ford and Williams, 2007, 421–427 pp.; Spektor and Spektor, 2009). Some karst conduits can be kept open by seasonally flowing water, transporting heat to the permafrost zone. Water with higher mineralization can flow within the permafrost at temperatures significantly below 0 °C. In Canada at 79° N, Pollard et al. (1999) documented perennial discharge of mineralized saline groundwater with a temperature of -4 °C. Nevertheless, the available age data show that the periods of permafrost propagation to greater depths were usually not associated with intensive CCC formation.

In contrast, the upper limit $(0 \circ C \text{ isotherm})$ of the relic permafrost is lowered during permafrost thawing. Thawing propagates to depth more rapidly along water-conducting clefts and karst channels. In all the studied cases, the karst porosity of limestone is related dominantly to clefts and corrosion channels; the limestone matrix porosity is relatively low. Because the zone above the upper limit of the deep relic permafrost is water-rich when a karst channel with liquid water penetrates into the top of a cavity, liquid water relatively rapidly fills the whole cavity or a part. The cavity itself, however, is still in the permafrost zone. The water freezes slowly here (cf. Pielsticker, 2000, who came to an identical conclusion from thermal modeling). The duration of the ice fill of the cavity is, therefore, only a short-term event. As the 0°C isotherm moves deeper, the ice melts and coarse crystalline CCC is freely deposited on the bottom of the cavity. CCC age data show that, depending on the oscillation of the surface climate, the permafrost depth oscillated during the Weichselian. An important period of repeated permafrost growth and destruction occurred between 40 and 21 ka BP, and the final permafrost destruction (at the depths of the studied caves) followed between 17.0 and 11.9 (possibly 9.2) ka BP. The basic principles of coarsely crystalline CCC formation in a cavity are depicted on a simple general model in Fig. 5. However, it is clear that more complex scenarios may be constructed for some of the studied caves, with repetition of water freezing periods (see Richter et al., 2010a) or possibly freezing of water streams and lakes in caves of the vadose type (see Richter et al., 2011).

4.4 Comparison of the coarsely crystalline CCC formation frequency with other climatic records

Comparison of the formation frequency of coarsely crystalline CCC with other climatic records is not straightforward. For well ventilated caves the temporal delay between a climate change at the surface and the related change of the cave parameter, e.g. cave air temperature, should be small. In case of caves with limited ventilation located within the permafrost zone the transfer of a surface climatic signal to the underground can be significantly delayed. This is due to the reason that the flux of heat into the permafrost zone -i.e. the propagation of the climate signal from the surface into the karst system – is limited by the thermodynamic properties of the soil and karst system. Thus, the permafrost thawing at depth can be significantly delayed with respect to surface climate. This is evidenced by local permafrost lenses surviving from the Last Glacial until today in NE Poland (Honczaruk and Śliwiński, 2011; Szewczyk and Nawrocki, 2011). This may cause contemporary growth of speleothems and CCC within the same cave system; speleothems may grow in shallow parts situated close to the surface (in already ice-free zone) and CCC may be formed in deeper parts (still lying in permafrost zone). The coarsely crystalline CCC from deep caves therefore represents a new paleoclimate archive indicating primarily the periods of existence of permafrost, its destruction phases, and its minimum depth.

As discussed in Sect. 4.3, the studied coarsely crystalline CCC are not related to the coldest and driest periods, but mostly to the warming trends following them. It seems probable that during the coldest phases of the Weichselian equivalent to MIS 4 and 2, the abrupt climatic oscillations known as Dansgaard–Oeschger events (cf. Genty et al., 2003) were not able to melt the whole permafrost zone, and that permafrost lenses survived in the study area during the whole MIS 4 and 2.

Baker et al. (1993) compiled over 500 U-series ages of common speleothems and travertines from northwestern Europe and compared their growth frequency with surface climatic changes. Since the growth of common speleothems and spring travertines requires significant groundwater supply and enhanced biogenic CO₂ production in the soil zone, the formation of these secondary carbonate types and the formation of coarsely crystalline CCC should be, at least partly, complementary. The growth frequency of speleothems and travertines after Baker et al. (1993) is shown in Fig. 4 (line b). It exhibits in general a very low cumulative growth frequency starting from the transition of MIS 5/4 at approximately 71 ka BP, with minor revival of the growth frequency between 62 and 49 ka BP. The lowest growth frequency were found between 46–44 and 35–22 ka BP. The majority (more than 86 % of samples) of the studied coarsely crystalline CCC samples were formed in several intervals between 71 and 10 ka BP, which is in general agreement with the low formation frequencies of common speleothems and travertines in this time interval. The lowest formation frequency of common speleothems and travertines at 36-34 ka BP (Baker et al., 1993) coincides well with the highest occurrence of coarsely crystalline CCC (six samples in the 36–34 ka BP interval, cf. Fig. 4).

Several authors used carbon and oxygen isotope time series, respectively, of precisely dated stalagmites to test the coherence of continental climatic record with the wellestablished Dansgaard–Oeschger oscillations known from

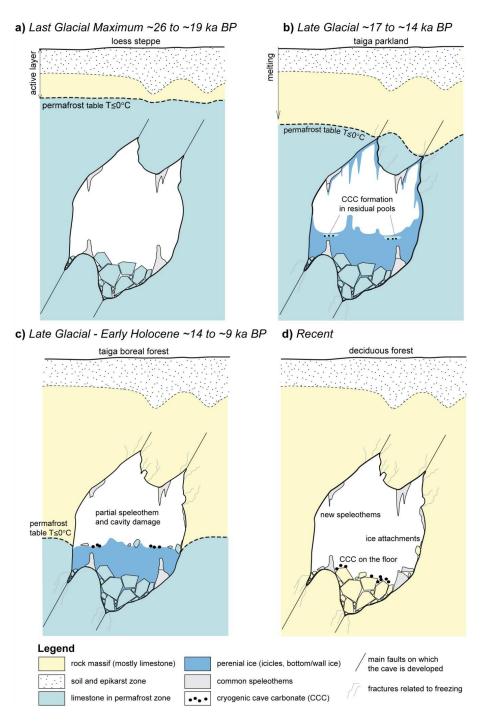


Fig. 5. A model of formation of coarsely crystalline CCC during a single permafrost destruction (thawing) after the Last Glacial Maximum. See text for further details.

ice cores and marine records (Genty et al., 2003; Boch et al., 2011). Genty et al. (2003) showed the coherence of a δ^{13} C time series from a stalagmite of Villars Cave located in southwest France with marine and ice-core records in the period between 83 and 32 ka BP and documented that the δ^{13} C signal reflects the Dansgaard–Oeschger events. The longest and the most important growth hiatus of the stalagmite between

 67.4 ± 0.9 and 61.2 ± 0.6 ka BP was interpreted as an extremely cold phase in southwestern France. First, more frequent occurrence of CCC in the studied Central European region (4 dates in the interval of 68–60 ka BP in Fig. 4) shows that the first extensive evolution of Weichselian permafrost in Central Europe occurred just in relation to this period equivalent to MIS 4, especially during its youngest parts. The

coarsely crystalline CCC ages higher than 68 ka BP were obtained on samples derived from caves located in mountainous settings, which exhibit low temperatures of cave interiors even today. The only exception is represented by the age of 83.5 ± 1.4 ka BP from the Apostelhöhle located at low elevation (Richter et al. 2010b). Since some of the calcite aggregates reported from this cave show δ^{13} C and δ^{18} O data in the range of common, non-cryogenic speleothems (see trend No. 1 in Fig. 1), this dated sample can be of non-cryogenic nature, but later being mixed with CCC.

Similarly, Boch et al. (2011) published combined stalagmite δ^{18} O record for the Northern Alps which is based on stalagmite time series from different caves which are located between 780 and 1964 m a.s.l. The combined δ^{18} O record covers the period 120–63.7 ka BP and is coherent with the Greenland Dansgaard–Oeschger pattern. The younger part of MIS 4, when the formation of common speleothems in the Northern Alps largely ceased, represents the incipient stage of formation of the studied coarsely crystalline CCC samples in Central European non-mountain settings.

An important cluster of coarsely crystalline CCC ages (5 ages obtained from several different caves in the time interval from 14 to 15 ka BP) occurs after the LGM and corresponds quite well to the chronological position of the rapid warming between Greenland Stadial 2a and Greenland Interstadial 1e at 14.7 ka BP (Lowe et al. 2008). Two younger coarsely crystalline CCC ages in the interval of 12–10 ka BP indicate that the latest permafrost lenses survived until this period, locally at depths of > 60 m below surface possibly up to 9.2 ka BP, as indicated by the single CCC datum from the Na Javorce Cave.

4.5 Estimation of past permafrost thickness

Discontinuous permafrost typically starts to develop in areas with MAAT below approximately 1 °C (King, 1984). Continuous permafrost is typical for areas with subzero MAAT. Its depth penetration can be roughly calculated based on MAAT using appropriate equations (e.g. French, 2007; Anderson and Anderson, 2010). Nevertheless, these model calculations represent only a rough estimate since the real permafrost thickness depends also on many other parameters such as thermal conductivity and diffusivity of earth materials, vegetation cover, quantity and duration of snow cover, and topography including the aspect, which influences the intensity of insulation or existence of water bodies at the surface (French, 2007).

An important factor for permafrost depth penetration is the duration of the climatic period with subzero MAAT. Construction and destruction (thawing) of deep permafrost is usually substantially delayed with respect to the evolution of the surface climate. Full thermal equilibration of the upper crust to the new surface climate conditions may take several thousand years (Šafanda and Rajver, 2001). Temperaturedepth profiles measured in deep boreholes in the area of past permafrost still exhibit thermal relics of the Weichselian permafrost in Europe even today (Šafanda and Rajver, 2001; Šafanda et al., 2004). A deep relic zone with temperature close to 0 °C starting at a depth of 357 m below the surface and continuing down to the borehole base (450 m) was reported by Honczaruk and Śliwiński (2011) and Szewczyk and Nawrocki (2011) in Cretaceous sediments of northeastern Poland. The temperature is controlled by water/ice transitions here.

Today, in Europe, permafrost exists at higher elevations in the Alps and sporadic permafrost patches are also assumed to exist in the highest parts of the Western Carpathians (High and Low Tatra mountains) in the territory of Poland and Slovakia (Brown et al., 1997; Keller et al., 1998; Dobiński, 2005). The present-day permafrost in European mountain regions is generally more irregular than the permafrost which developed in lowlands during glacial periods. In the Alps, the lower limit of occurrence of the present sporadic permafrost is at approximately 2100 m a.s.l., and discontinuous permafrost starts here at approx. 2400 m a.s.l. (e.g. Keller et al., 1998). Sporadic or discontinuous permafrost is probably also present in the Tatra Mountains at elevations above approximately 1930 m a.s.l., but has so far been indicated only by ground-surface temperature measurement and ground-penetrating radar on the northern mountain slopes (e.g. Gadek and Grabiec, 2008). The present-day climaterelated change of permafrost in Europe was studied by Harris et al. (2003, 2009).

The deepest cavities hosting coarsely crystalline CCC in the "Rheinisches Schiefergebrige" (Germany) and in the Bohemian Karst (western part of the Czech Republic) are at similar depths of 60 to 65 m below the surface. Data do not allow an estimate whether the permafrost was discontinuous or continuous. The depth of 65 m can be considered as the minimum permafrost depth during the latter half of the Weichselian in these areas. Since the formation of CCC occurs predominantly during permafrost destruction, which certainly proceeds both from above and from below, the maximum permafrost depth was certainly greater. It should also be noted that the permafrost depth is typically lower in karst areas than the surrounding non-karst areas (Ford and Williams, 2007). One site in the Northern Calcareous Alps exhibits CCC at a depth of 229 m below the entrance (entrance elevation of 2230 m a.s.l.).

The permafrost depth map of the Last Glacial Maximum by Frenzel et al. (1992) indicates for Central Europe the permafrost zone thickness in the range between 0 and 200 m. Based on unpublished modeling data (V. T. Balobayev and L. I. Schipiciny in Czudek, 2005), the maximum Weichselian permafrost depth in the Czech Republic was estimated to be in the range from approximately 50 m to a maximum of 250 m, which is in agreement with the depth estimate presented in this paper. Pons-Branchu et al. (2010) supposed the absence of a continuous permafrost zone in northeastern France during the period of 55.36 ± 0.95 until 53.34 ± 0.49 and at ca. 45.85 ± 0.49 ka BP, based on the formation of common speleothems in this region. None of the CCC ages obtained by this study fits these intervals.

In the eastern part of the Czech Republic (Moravia), only a single locality with coarsely crystalline CCC occurrence is known at the relatively shallow depth of 30 m below the surface (Javoříčko Caves). The same cavity also hosts other indications of former ice fill (e.g. destroyed speleothems and "ice attachments"). In the large cave systems of the slightly more southerly located Moravian Karst, no CCC site is known. This absence is possibly due to the existence of abundant underground water streams in this karst region, which could produce extensive taliks. Relatively thick permafrost in the northern part of Moravia is indicated by the observation of Růžičková and Zeman (1992), who described post-cryogenic structures down to a depth of 220 m in the Blahutovice borehole. Unfortunately, this observation is not accompanied by dating and it is not clear when the observed features did form.

All the sites in Slovakia are located in mountainous areas. Greater depths for the coarsely crystalline CCC sites can be expected here since even today some of these caves exhibit relatively low temperatures. The deepest studied site in Mesačný Tieň Cave in the High Tatra mountains is located at a depth of 285 m (surface elevation approx. 1800 m a.s.l.). The present-day temperature of the cave interior varies between 1.0 and $3.5 \,^{\circ}$ C (B. Šmída, personal communication, 2011; no precise temperature measurements have been performed to date). This deep occurrence of coarsely crystalline CCC is in agreement with earlier estimates of Weichselian permafrost depth for the High Tatra mountains, with potential Weichselian permafrost depths down to 400 m below the surface (Dobiński, 2004).

5 Conclusions

Coarsely crystalline CCC formed by expulsion of the dissolved load during slow water freezing in caves during the Last Glacial (Weichselian) has been found in 20 caves located in the territories of Germany, the Czech Republic, Slovakia and Poland. The caves were located in the glacierfree zone between the northern continental glaciation and the Alpine mountain glaciation during the Weichselian. The coarsely crystalline CCC formed mainly in several restricted time periods, mostly between 40 and 21 ka BP and 17.0 and 12 ka BP. Its formation in deep caves mostly occurred during periods of permafrost thawing related to surface withinglacial warming periods (interstadials), when liquid water penetrated into empty cavities within the deeper relic permafrost and froze slowly. The studied occurrences indicate the minimum permafrost depth during the younger half of the Weichselian in the studied area to be more than 65 m in the lowlands and uplands. In the High Tatra mountains, a permafrost depth of more than 285 m below surface is suggested.

Supplementary material related to this article is available online at: http://www.clim-past.net/8/1821/ 2012/cp-8-1821-2012-supplement.pdf.

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