Uranium isotopic ratios and their implication for uranium–uranium dating and groundwater circulation studies: A case study from speleothems of the Demänová caves, Nízke Tatry Mts., Slovakia

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Abstract: Detailed analyses of speleothems collected in caves of the Demänová Valley, Nízke Tatry, Slovakia, have found significant variability in uranium concentrations and initial ^{234}U/^{238}U activity ratios, plus correlation between these variables. They are related to the ages of samples and their differing locations in the caves. Strong variability of initial ^{234}U/^{238}U activity ratios is the reason for serious limitations in applying the ^{234}U/^{238}U Regional Uranium Best Estimate method as an age estimator for speleothems older than the range of the ^{230}Th/^{234}U method (500 ka). The strong variability of uranium contents in speleothems, rocks and waters and their isotopic compositions allowed us to distinguish distinct periods with changes in water circulation paths or distribution/weathering degrees of deposits in surface alimentation areas or in the epikarst zone, that result from the more ready leaching of the ^{234}U isotope from sediments. Periods characterised with higher initial ^{234}U/^{238}U activity ratios are most likely associated with the deposition of "fresh" colluvial deposits and/or soil development (potential uranium source) in the alimentation area. Gradual decrease of initial ^{234}U/^{238}U activity values indicate prolonged stable weathering and uranium leaching conditions. The Demänová results indicate periods of intensive erosion and "fresh" sediment deposition in the alimentation areas at ~120 ka, ~80–70 ka, and during the Holocene.

Keywords: Uranium isotopes, U-series methods, Demänová Cave System, Western Carpathians.

Introduction

Caves in Demänovská dolina Valley (Demänová caves; DC; Fig. 1) are one of the most famous multilevel cave systems in the world. They were formed by allogenic streams sinking into Triassic carbonates. Demänovská ľadová jaskyňa (Demänová Ice Cave; DIC), Demänovská jaskyňa mieru (Demänová Cave of Peace; DCP) and Demänovská jaskyňa slobody (Demänová Cave of Liberty; DCL) are interconnected, the first two being show caves. Detailed studies of cave morphology and correlation of cave levels with river terraces in the Demänovka and Váh river basins were undertaken by Droppa (1966, 1972) and partly modified by Bella et al. (2011). As a result, a scheme of multiphase cave development since the Pliocene was proposed and ages of the cave levels were estimated.

The large study of the uranium series ages of a sample of the DC speleothems presented here is a part of a co-operation between the Institute of Geological Sciences (Polish Academy of Sciences) and Slovak Caves Administration, with the chief aim being to establish a more exact time scale for the sequence of cave development. More than 450 uranium–thorium (U–Th) analyses of speleothems from the DC have been carried out

in total (Table 1; Supplementary Table S1), yielding sample ages and detailed information on the uranium (U) concentrations and isotopic ratios.

Information on ^{234}U/^{238}U activity ratios in speleothems can be useful for two principal reasons:

- The dating range of the U–Th dating method is limited to 500 ka by the 75,700 year half-life of ^{230}Th. The half-life of ^{234}U (245,000 years) allows the dating limit to be extended to 1–1.2 Ma by the ^{234}U–^{238}U method if robust estimation of the initial ^{234}U/^{238}U activity ratio (i.e. at time of deposition) is possible. The U–U method has been successfully applied for dating marine calcite deposits, aided by the limited variation of the ^{234}U/^{238}U activity ratio in seawater (Ivanovich & Harmon 1992; Henderson 2002). In speleothems, much bigger regional and intra-regional variations of the initial ^{234}U/^{238}U activity ratio are usually found due to the widely differing water–rock interaction periods and physico–chemical conditions (e.g., temperature, pCO$_2$, pH) related to climate or hydrology. Sometimes, the homogeneity of U/U ratios obtained from younger speleothems (within the range of the U–Th method) allow a reasonable "Regional Uranium Best Estimate" (RUBE) of the initial ^{234}U/^{238}U activity ratio
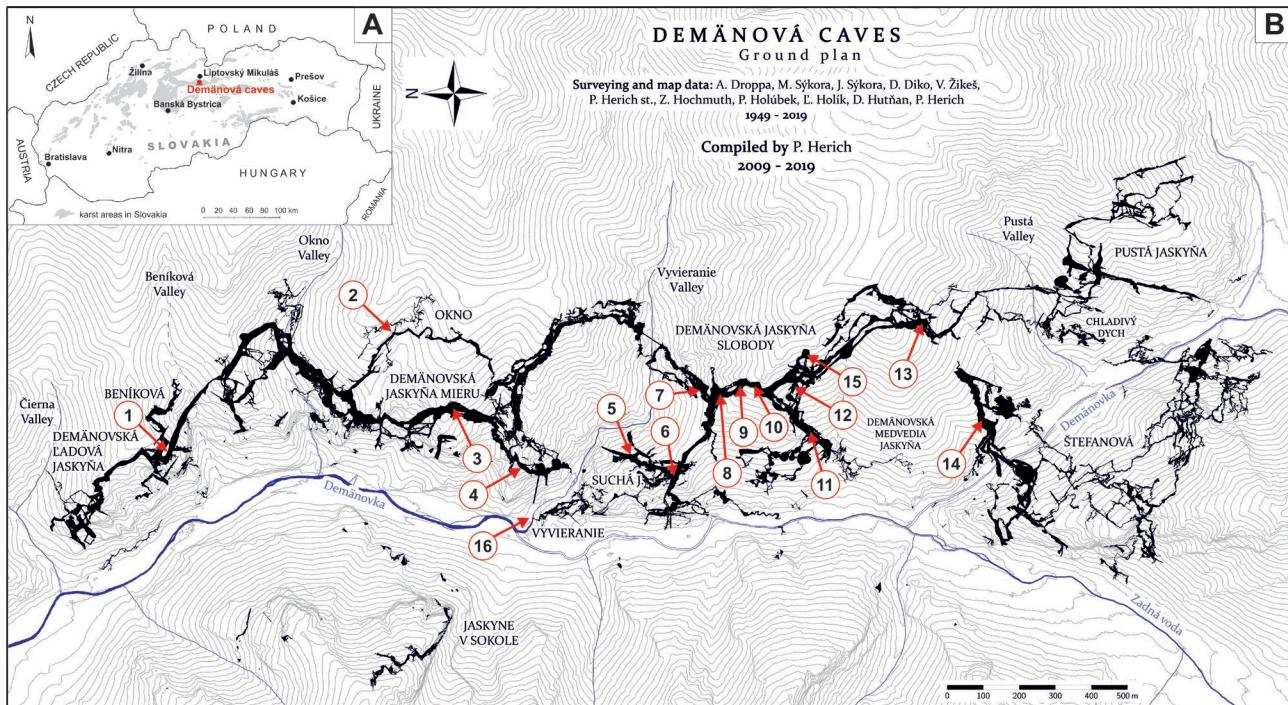
Fig. 1. Location of the study site: **A** — location of the DC in Slovakia; **B** — plan view of the DCS and other caves in the Demänovská dolina (modified after Droppa 1972, and Herich 2017). 1 — DIC (Závrtový dóm Chamber); 2 — Okno Cave (between the Priepasťová chodba Passage and Stĺpová sieň Hall); 3–4 — DCP (3 — Vodopádový dóm Chamber; 4 — Kaskádová chodba Passage); 5–13 — DCL (5 — Hlinená chodba Passage; 6 — Mramorové riečisko Passage; 7 — Suchá chodba Passage; 8 — Veľký dóm Chamber; 9 — between Ráscestie and Veľký dóm Chamber; 10 — Rázcestie (Prízemie) Passage; 11 — Cintorín; 12 — Žulová chodba Passage; 13 — Pekelný dóm Chamber); 14 — Štefanová Cave; 15 — Janáčkov dóm; 16 — Vyvieranie.

Table 1: Samples included in the study.

	Analyses	Excluded samples		Included samples	Cave	Included samples	Sites
Demänová Cave System (DCS)	458	Out of range	51	358	DIC	18	1
					DCP	40	2
		Detrital contamination	49		DCL	295	9
					Štefanová	3	1
					Okno	2	1

of older samples in the same cave or nearby to be made (Gascoyne et al. 1983). The RUBE method has been applied successfully to date speleothems precipitated from hypogene waters (Ludwig et al. 1992 at Devil's Hole, Nevada, and Ford et al. 1993 in Wind Cave, South Dakota), marine sediments (Bender et al. 1979; Ludwig et al. 1991), lacustrine deposits (Kaufman 1971) and soil profiles (Ludwig & Paces 2002). A similar approach was applied for speleothems from the DIC (Hercman et al. 1997).

• The second reason for investigating the variability of initial $^{234}U/^{238}U$ activity ratios over space and time is their use as climatic or hydrologic indicators. In Soreq Cave (Israel), Ayalon et al. (1999) linked higher initial $^{234}U/^{238}U$ activity ratios to periods with increased U weathering in soils due to wetter conditions. Lower values are characteristic of periods with higher relative proportions of carbonate-derived carbon.

Changes of the initial $^{234}U/^{238}U$ activity ratio values in Holocene speleothems from the DC, in combination with changes in carbon isotopic composition, have been interpreted as indicators of changes in the groundwater circulation paths and fracture/soil geometry above the cave (Hercman et al., in press). As ^{234}U is more easily leached in fresh deposits/regolith or soils, disequilibrium between ^{234}U and ^{238}U results. Three principal causes are suggested for the disequilibrium (Rosholt et al. 1963; Kigoshi 1971; Fleischer & Raabe 1978): (1) the ejection of ^{234}U recoil nuclei from the solid grains directly into the water; (2) the recoil creates a damaged zone in the mineral matrix that is afterwards more easily leached by water, and (3) ^{234}U as a slightly lighter isotope is more mobile than ^{238}U.

Tritium is a commonly used radioactive isotope, chiefly for dating but also as a natural tracer in analyses of underground and surface waters; e.g., in the Niedźwiedzia Cave system

(Sudetes, Poland) tritium analyses have shown that there are at least two different water sources (Gąsiorowski et al. 2015). In a similarly fashion, the investigation of long-lived isotopes like U and Th can provide additional information on water sources, circulation and epikarst processes. This information may be especially important in complex karst areas with relatively high infiltration rates, variable flow rates, concentration of flow along fissures, long-distance underground flow, etc. (Ford & Williams 2007).

This study of U isotopic composition and content in DC speleothems and rocks from the Demänovská dolina aimed to: (1) test whether the initial $^{234}U/^{238}U$ activity ratios in the speleothems were stable enough, at least for Middle and Late Pleistocene time, to allow application of the RUBE method to date samples older than the 500 ka limit of the U–Th method, and (2) test whether any variability in U content and isotopic ratios can be used as supplemental climatic/hydrological indicators in the DC or of weathering stages in the alimentation zone (soil and epikarst above the caves).

Materials and methods

The DC are located in the Nízke Tatry (Low Tatra Mts., Western Carpathians, northern Slovakia), chiefly on the eastern side of the Demänovská river valley (Fig. 1). Most of it is contained in a group of eleven caves that historically were explored as separate entities but have been interconnected by further exploration to create the Demänová Cave System (DCS). The total length is more than 41.4 km (Herich 2017). The DIC, DCP, DCL, Vyvieranie and Pustá jaskyňa caves are the largest components of the DCS. Štefanová Cave (>17 km in length) and Okno Cave (>2.6 km; Herich 2017) are other important nearby caves that are probably genetically connected but not yet linked by explorers. Samples of speleothems, stalagmites and flowstones, have been collected for analysis since 1995 in DIC (one site), DCL (nine individual sites), DCP (two sites), Štefanová Cave (one site) and Okno Cave (one site; Fig. 1; Table 1).

All of the samples studied consist of non-porous, dense, well-crystallised calcite with no traces of re-crystallisation or secondary dissolution and cementation. Water samples were collected in two sites inside the cave (Veľký dóm and Janáčkov dóm chambers in DCL; Fig. 1/sites 8 and 15) and from a karst resurgence in the mouth of Vyvieranie side valley (Fig. 1/site 16). Water was collected in 22-liter polypropylene containers. Immediately after sampling, the water was acidified with HNO_3 to avoid radionuclide precipitation onto container walls and spiked with a mixture of $^{232}U–^{228}Th$ dissolved in nitric acid, to determine the proportional chemical recovery of the isotopes to be studied.

Nineteen carbonate rock samples were collected from the cave and Demänovská dolina. Rock samples inside the DCS were collected in the speleothem sampling sites in Veľký dóm Chamber (Fig. 1/site 8), Rázcestie (Prízemie) Passage (Fig. 1/site 10), Pekelný dóm Chamber (Fig. 1/site 13), Suchá chodba (Fig. 1/site 7), Hlinená chodba (Fig. 1/site 5) and Žulová chodba (Fig. 1/site 12) passages. Additional rock samples were collected along the main road in Demänovská valley.

The analyses were performed in the U-series Laboratory of the Institute of Geological Sciences (Polish Academy of Sciences, Warsaw, Poland). Two methods of U isotopic measurement were used during the twenty+ year duration of the DC programme: alpha spectrometry for the first fifteen years, and mass spectrometry (ICP-MS) for the past nine years. The individual speleothem and rock samples measured by alpha spectrometry had masses ranging from 0.5 to 2 g (usually <1g). U and Th were separated using an international standard chemical procedure for carbonates (Ivanovich & Harmon 1992) – dissolution in 6 M nitric acid, U and Th separated by chromatography using DOWEX 1×8 resin, the efficiency of separation being monitored by addition of a $^{228}Th–^{232}U$ spike (UDP10030 tracer solution by Isotrac, AEA Technology) before treatment. The alpha activity measurements were obtained on DUO ANSEMBLE or OCTETE PC spectrometers (EG&G ORTEC), equipped with 1200 mm^2 active area, ultra-low background detectors. Each spectrum was corrected for the background and delay time between the chemical separation and the measurement.

Samples for mass spectrometric analysis were collected by drilling and usually weighed 0.1 to 0.5 g. A $^{233}U–^{236}U–^{229}Th$ spike (calibrated by the analysis of uraninite in secular equilibrium) was added to the samples at the beginning of procedures. All subsamples from speleothems were dissolved in nitric acid. U and Th were separated from the carbonate matrix using the chromatographic method with TRU-resin (Hellstrom 2003). Internal standard samples and blanks were prepared and used in all sets of analyses. The mass abundances of ^{236}U, ^{233}U, ^{234}U, ^{235}U, ^{229}Th, ^{230}Th, and ^{232}Th were measured by a double-focusing sector-field ICP mass analyzer (Element 2, Thermo Finngan MAT) in the ICP-MS laboratory in the Institute of Geology of the Czech Academy of Sciences. The instrument was operated at a low mass resolution (m/Δm≥300). A double pass spray chamber with a Teflon nebulizer was used for the sample introduction. The measurements were corrected for counting background and chemical blanks.

100 results have been excluded from the total of 458 analyses. The first group of rejected results (51) were those samples found to be older than 350 or 500 ka (the $^{230}Th/^{234}U$ α spectrometry and mass spectrometry ranges, respectively). The second group (49) were excluded because of significant detrital contamination. For the alpha spectrometry, a value of 20 for the measured $^{230}Th/^{232}Th$ activity ratio was considered the threshold value for "clean" samples. For the ICP-MS measurements, this threshold was set to 200–300 owing to the greater detection capability of the instrument (Hellstrom 2006). No ages were calculated for the rejected samples. For the accepted samples, initial $^{234}U/^{238}U$ activity ratios were calculated from the measured $^{234}U/^{238}U$ activity ratios and their respective $^{230}Th/^{234}U$ ages.

In the water samples, U isotopes were extracted by co-precipitation with iron hydroxide using ammonia and iron

chloride (Narita et al. 1989). The supernate was decanted, and the precipitant then centrifuged with 4000 rpm over 10 min. Residue was dissolved in hydrochloric acid and U and Th isotopes were separated using the standard procedure (Ivanovich & Harmon 1992). Uranium was separated from solution on chromatographic columns (Horwitz et al. 1993) using a DOWEX 1×8 resin and electro-deposited on stainless steel discs. Activities were measured by alpha spectrometry.

Results

The values calculated for the initial $^{234}U/^{238}U$ activity ratios of the DC speleothems cover a relatively wide range, from 0.69 to 4.7 (Fig. 2A). A characteristic multimodal distribution is observed, dominated by values a little below 1 (implying depletion in ^{234}U) and around 1, respectively.

The relationship between the initial $^{234}U/^{238}U$ activity ratios and the sample ages suggests several different groupings in the data. First, two groups can be distinguished based on the ^{234}U enrichment or depletion (Fig. 2B). Periods of higher ^{234}U enrichment are seen around 100 ka and in the youngest samples (Fig. 2B). There are several periods of higher ^{238}U content around 300 ka, 200 ka and 80 ka (Fig. 2C). Plotting the initial $^{234}U/^{238}U$ activity ratios against ^{238}U content shows that there is ^{234}U enrichment in the samples characterized by lower U content (Fig. 2D).

The relationships between the U content and isotopic ratios differ in particular parts of the cave system. For samples from the Ice Cave (DIC), the initial $^{234}U/^{238}U$ activity ratios have changed from ca. 1.6 to 3, displaying multimodal behaviour (Fig. 3A) and clearly lower ^{234}U enrichment in older samples (Fig. 3B). However, there is no correlation between the U concentration and the ages of the samples (Fig. 3C). The U concentration in these speleothems is 0.15–0.48 ppm (Fig. 3C; Supplementary Table S1), falling into the range typical for speleothems. Once again, there is no correlation between initial $^{234}U/^{238}U$ activity ratios and U concentration (Fig. 3D).
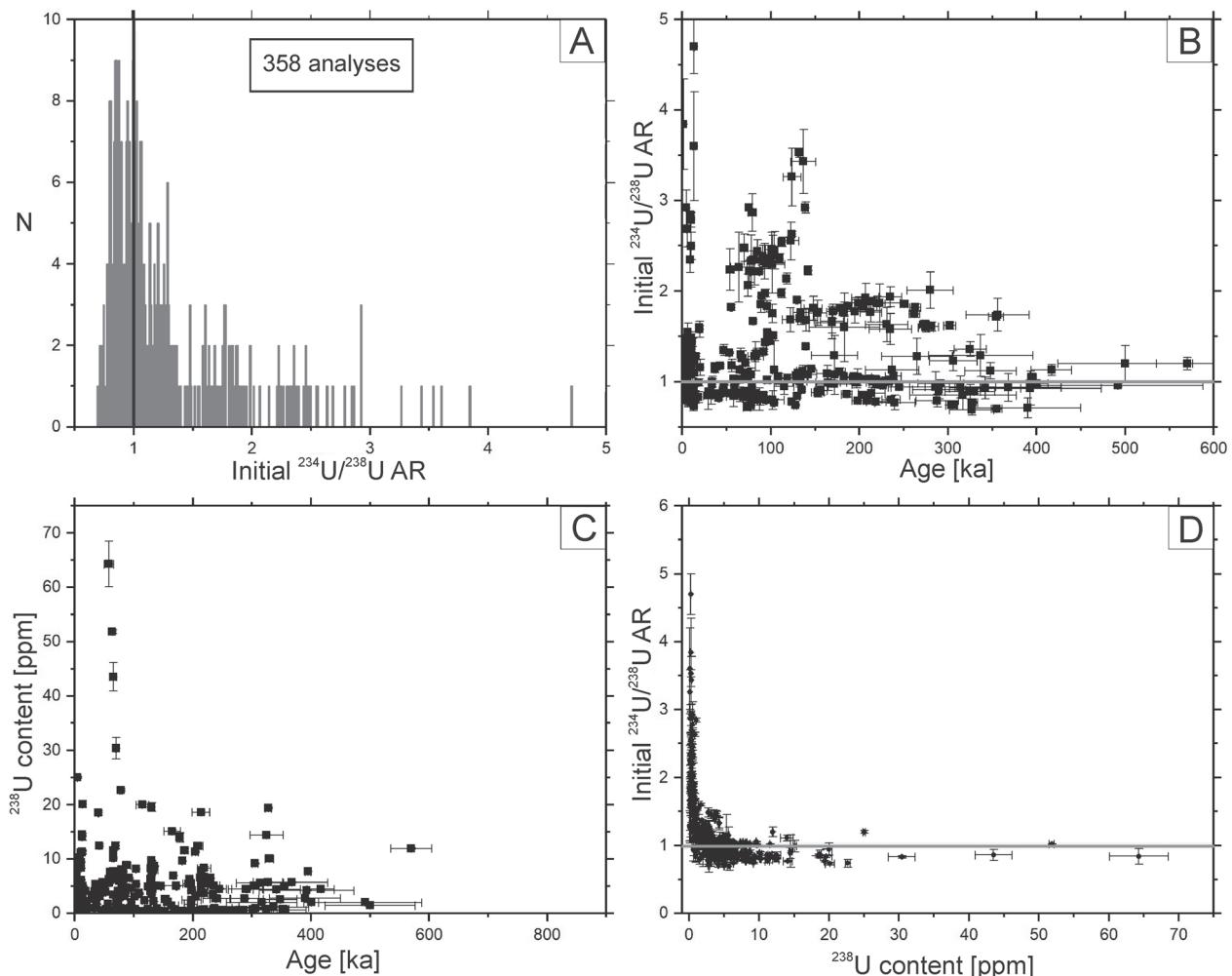
**Fig. 2.** Uranium isotopic composition and content in the speleothems from the DCS: **A** — histogram of the calculated initial $^{234}U/^{238}U$ activity ratios; **B** — calculated initial $^{234}U/^{238}U$ activity ratios vs. sample age; **C** — ^{238}U content vs. sample age; **D** — calculated initial $^{234}U/^{238}U$ activity ratios vs. ^{238}U content.
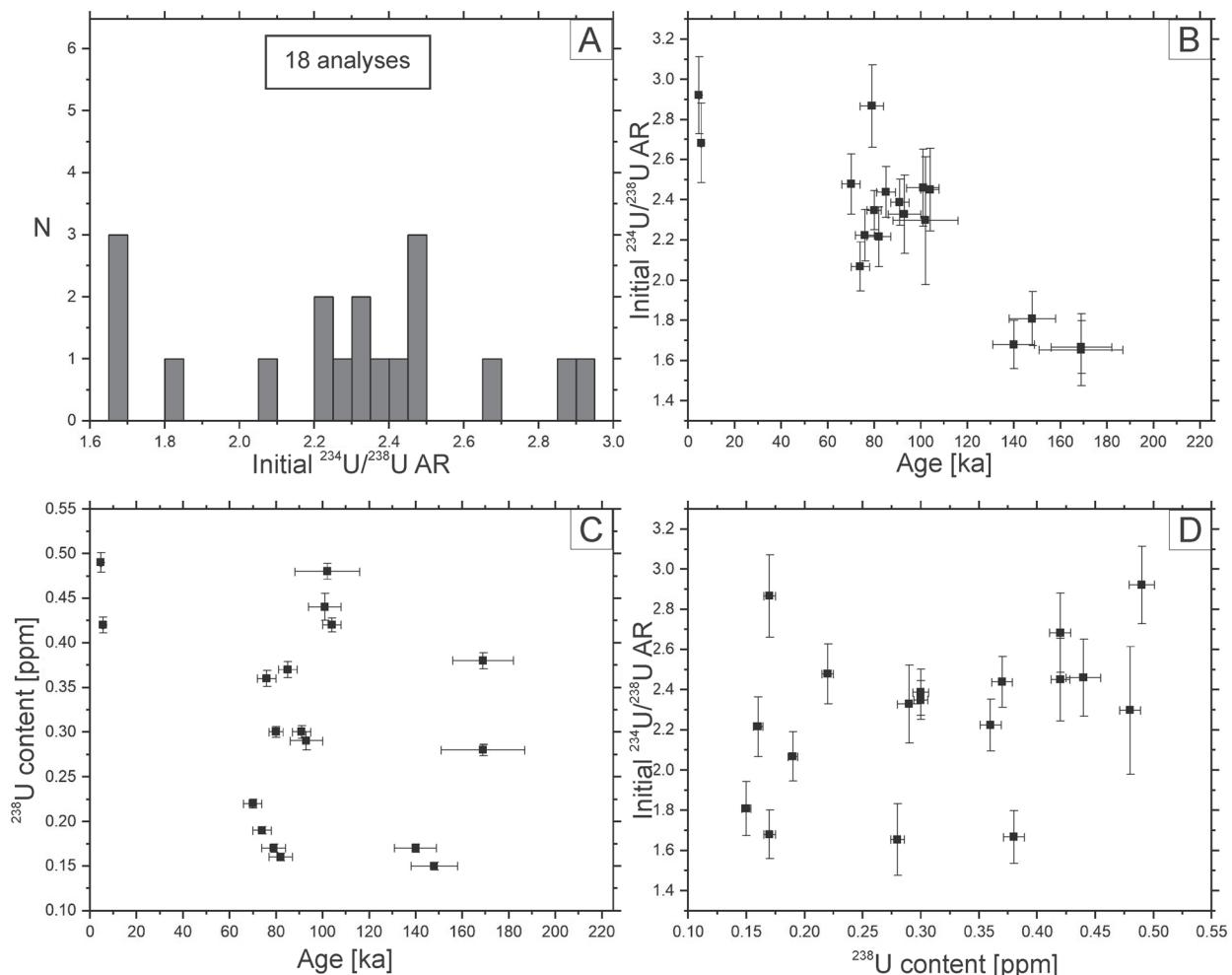
Fig. 3. Uranium isotopic composition and content in the speleothems from the DIC. For explanations see Fig. 2.

These relationships are more complicated in the Peace Cave (DCP; Fig. 4). A division into two distinct sample groups is evident. The first consists of samples with initial $^{234}U/^{238}U$ activity ratio greater than 1 (1.1–2.8), with a weak trend to higher values in the younger samples. The second group has initial $^{234}U/^{238}U$ activity ratio values that are all below 1, i.e. not a typical situation for speleothem samples. The calculated initial $^{234}U/^{238}U$ activity ratios again display multimodality (Fig. 4A) and the samples with initial $^{234}U/^{238}U$ activities below 1 are all younger than 40 ka (Fig. 4B). It was during that time period that all samples with a relatively high uranium content (>2 ppm) were deposited (Fig. 4C). There is a clear relationship between uranium content and initial $^{234}U/^{238}U$ activity ratios (Fig. 4D). U concentrations in the DCP speleothems range from 0.18 to 6.75 ppm (Fig. 4C; Supplementary Table S1).

The biggest data set (N=295) has been obtained from the Liberty Cave (DCL) speleothems (Fig. 5). ^{238}U concentrations in DCL samples covers a much wider range than in the other caves, i.e. from 0.18 ppm to ca. 64 ppm (Supplementary Table 1). The extremely high ^{238}U content of a few samples was surprising for us, so their analyses were repeated as

a check (repeat values are not included in the data plots). There are seven samples with "extremely" high U content (>20 ppm, <2 % of the total sample) and a total of 31 samples >10 ppm U; <10 % of total sample population (Supplementary Table S1). Excluding the seven highest U samples, the final DCL data analysis is similar to the full DC sample set (Fig. 2) because of their statistical dominance. As in the Peace Cave, two groups of samples with different initial $^{234}U/^{238}U$ activity ratios can be distinguished, the first with U concentrations below 1 ppm and high initial $^{234}U/^{238}U$ activity ratios, the second with much higher U concentrations and lower enrichment in ^{234}U (Fig. 5D).

In the host Mesozoic rock samples, ^{234}U and ^{238}U are in the radioactive equilibrium state, as expected. The U content ranges 0.65–3.52 ppm (Fig. 6A). Higher values (typically 2–3 ppm) are recorded in dolomitic strata and lower values in limestone. The speleothem ^{238}U concentration range (0.15–64 ppm), thus extends one order of magnitude higher than in the host rocks that were sampled (Fig. 6A).

The drip water sample collected in the Veľký dóm (DCL; Fig. 1/site 8), as well as a sample from the karst spring, show low U content (around 0.1 and 0.2 mBq/L respectively) and

enrichment in ^{234}U (Fig. 6B). In contrast, drip water collected in Janáčkov dóm (DCP; Fig. 1/site 15) has a U content one order of magnitude higher (around 7.5 mBq/L) and is depleted in ^{234}U. The relationship between the U content and the isotopic composition in the water samples (Fig. 6D) is similar to that for the DC speleothems (Fig. 6C).

Discussion

In general, a relationship between the initial ^{234}U/^{238}U activity ratios and U content and the sample age is observed in the speleothems collected in the DC (Figs. 2–5). Correlation between initial ^{234}U/^{238}U activity ratios and U content cannot be seen in samples from the DIC but there is a strong suggestion of negative correlation with sample age (Fig. 3). The results from DCP and DCL suggest quasi-exponential relationships between the initial ^{234}U/^{238}U activity ratios and the U concentration (Figs. 4–5).

The variability of U concentration and initial ^{234}U/^{238}U activity ratios of the DC speleothems in relation to their ages

is plotted in Fig. 7. The samples have been divided into four "age groups": <20 ka, 20–100 ka, 100–250 ka and >250 ka. The mean and median values of initial ^{234}U/^{238}U activity ratios and U content of these groups agree within one standard deviation but the range of the measured values strongly differs. This is especially true of the maximum values (uppermost triangles). It suggests episodic high U content in deposited speleothems causing high skewness distribution. As a result analytical data for specific samples may strongly differ from the mean value used as an initial value estimator. Negative correlation between initial ^{234}U/^{238}U activity ratios and U content is clear, especially for the maximum values.

The inter-site variability of initial ^{234}U/^{238}U activity in the DC speleothems is summarized in Fig. 8. ^{234}U enrichment was found in most DCL sites (Fig. 1/sites 5–13), except for Rázcestie (Fig. 1/site 10). In the Žulová chodba (Fig. 1/site 12), the oldest flowstone layers (~300–350 ka old) are depleted in ^{234}U, but the younger ones are enriched. Most of the Holocene stalagmites are depleted in ^{234}U in sites 3 and 4 (DCP; Fig. 1). However, six of the Holocene and Weichselian stalagmites from site 4 are enriched (Fig. 8).
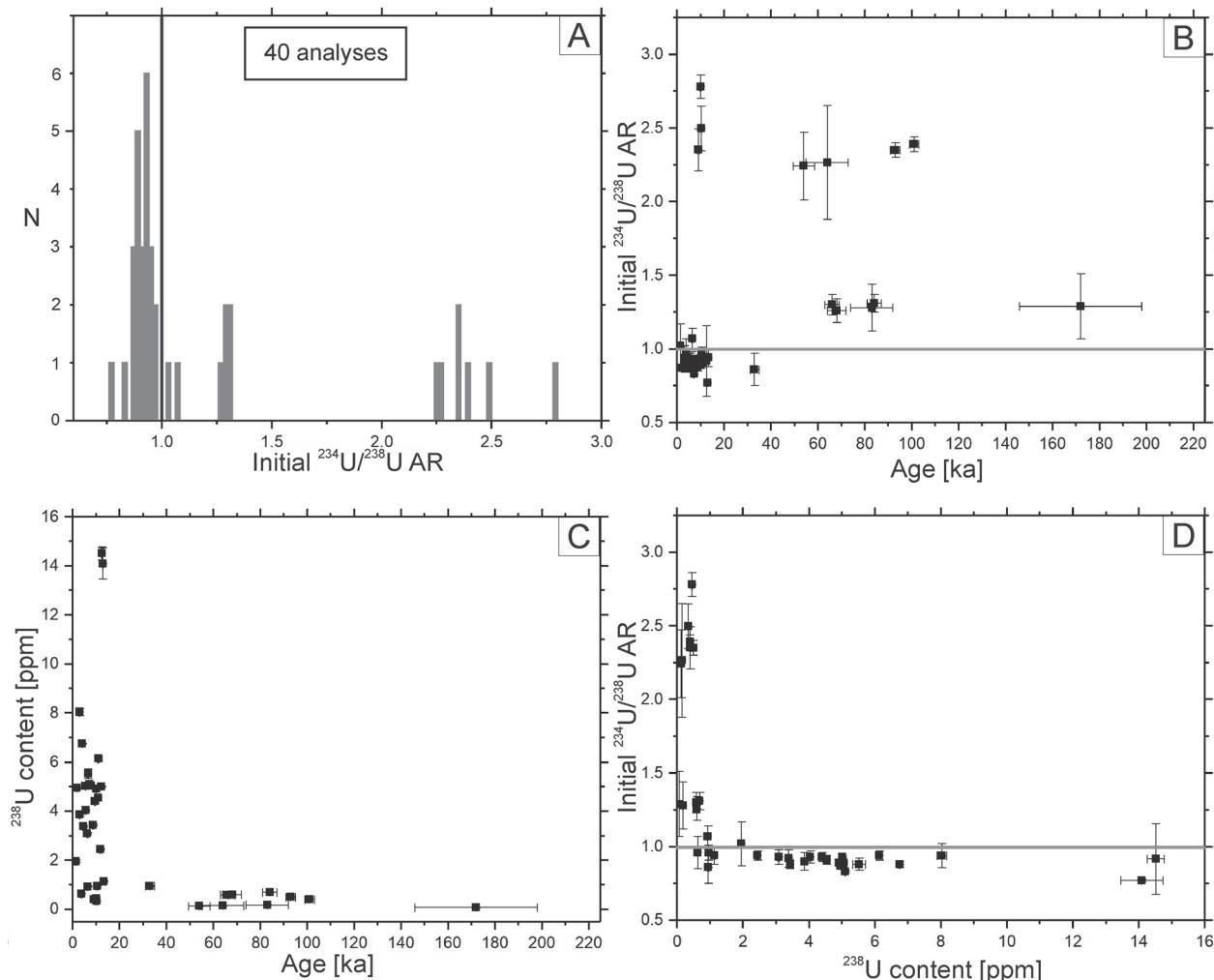
Fig. 4. Uranium isotopic composition and content in the speleothems from the DCP. For explanations see Fig. 2.
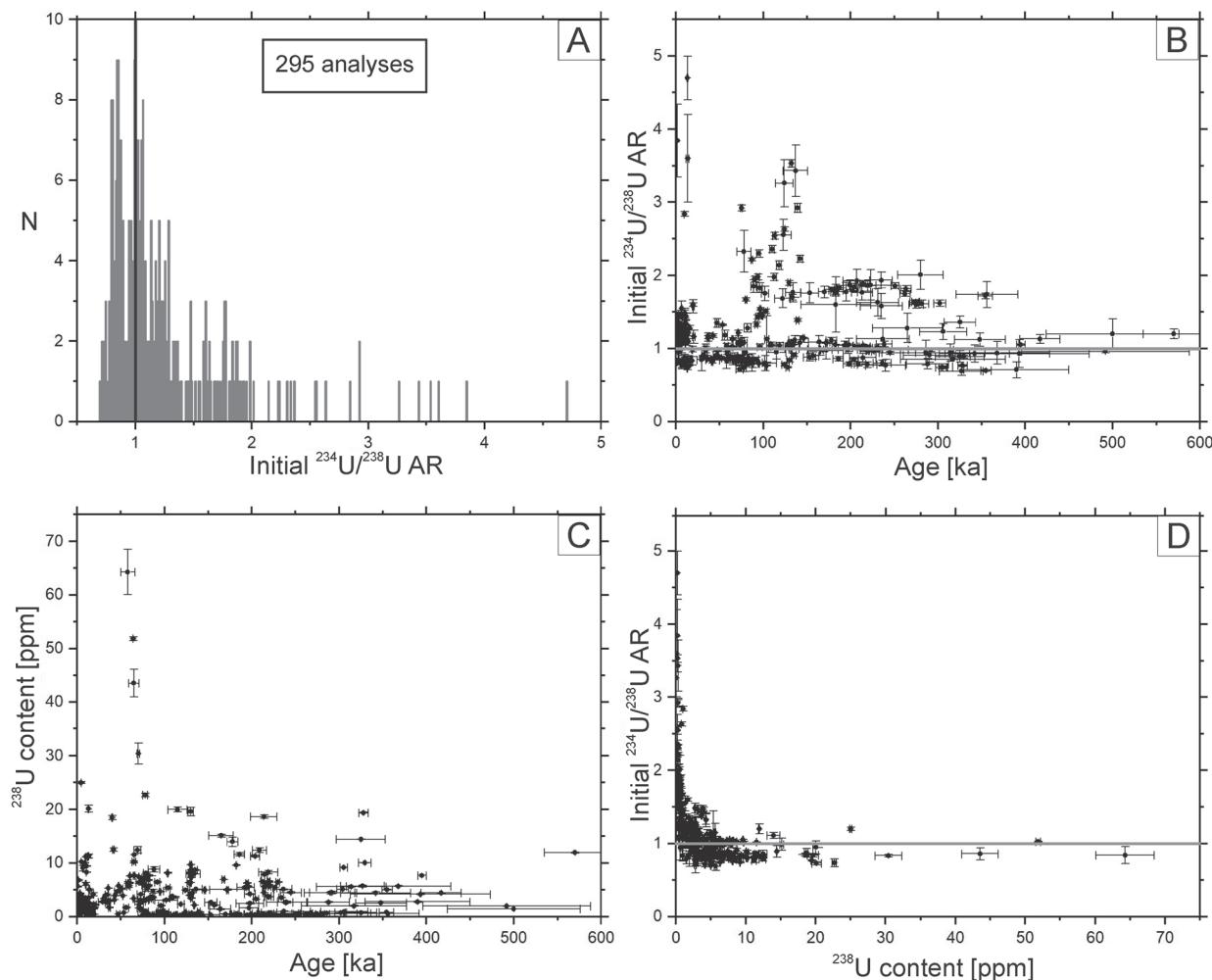
Fig. 5. Uranium isotopic composition and content in the speleothems from the DCL. For explanations see Fig. 2.

Variability in initial U isotopic composition in the speleothem samples cannot be explained by variability of U isotopic composition in the host rocks of the Demänovská dolina, which is much smaller (Fig. 6A). It may be linked with changes in the surface and cave deposits that are the U sources for the percolating meteoric waters. Uranium washed from relatively "fresh" deposits is expected to be enriched in ^{234}U as the more mobile isotope. Preferential removal of ^{234}U from the source deposits causes gradual decrease of the ^{234}U/^{238}U activity ratio in the carrier water and, later, in the speleothems. This suggests that initial ^{234}U/^{238}U activity ratios may be used as proxies for the intensity of deposits weathering that are the sources of leached uranium. The periodicity of changes in ^{234}U enrichment levels suggests periods of more intensive leaching and/or accumulation of "fresh" sediments in the alimentation areas.

Changes in initial U:U ratios and concentrations were found in samples of different ages collected close together in some of the DC sites, or even within a single flowstone/stalagmite. In Veľký dóm (Fig. 1/site 8), U contents and initial isotopic ratios in samples older than ~80 ka were distinct from the younger ones (Fig. 9A). All of the older samples there display

the ^{234}U enrichment typical of speleothems. The younger samples can be divided into two sub-populations, enriched and depleted. This suggests that around 80 ka ago, in the source areas of the waters depositing speleothems in Veľký dóm, "fresh" sediments accumulated as sources for leaching solutions strongly enriched in ^{234}U. With time, preferential leaching caused the gradual decrease of enrichment level. Gradual change of the initial U isotopic ratios is well shown in the data from a single Veľký dóm flowstone (Lab. No. 233–235 in Supplementary Table S1); here, the change from ^{234}U enrichment to depletion took place between 60 and 66 ka (Fig. 9B).

The general characteristics of the U isotopic data that were obtained in the DCS limit the application of the RUBE method for estimating ^{234}U/^{238}U ages for speleothems with ages beyond range of the ^{230}Th/^{234}U method. Using the results with ages within the ^{230}Th/^{234}U range as a guide to initial ^{234}U/^{238}U activity ratios for RUBE dating estimates is problematic in these caves. On the one hand, the mean values of the initial ^{234}U/^{238}U activity ratios in speleothems from different parts of the cave system or of different ages agree within one standard deviation but, on the other, the variations in these values are high
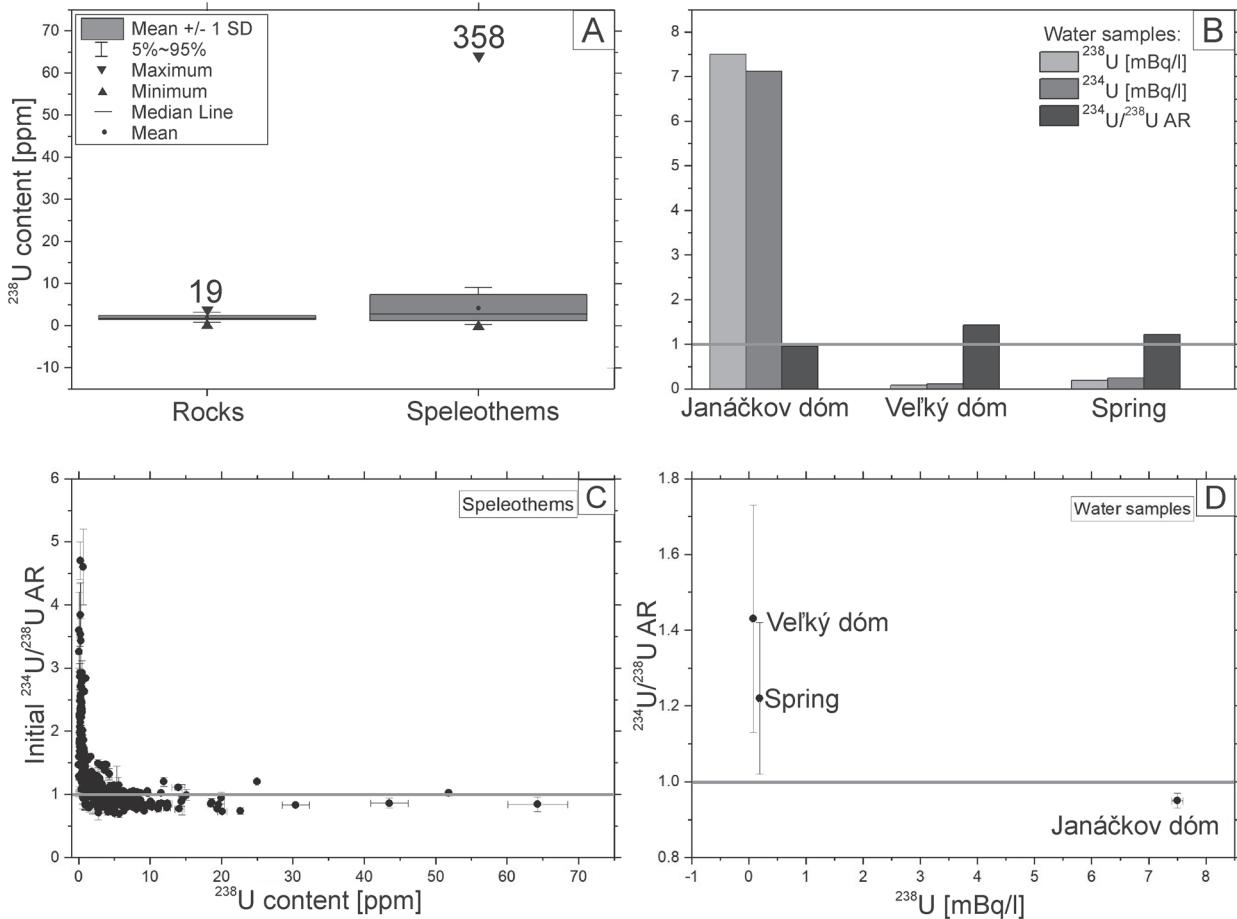
Fig. 6. Uranium content and isotopic composition in different elements of the DC karst system: **A** — uranium content in carbonate rocks and speleothems; **B** — uranium content in drip water samples; **C** — relation between the U content and the initial isotopic composition in speleothems; **D** — relation between the uranium content and the isotopic composition in the water samples.

(Fig. 10). The distribution of initial $^{234}U/^{238}U$ activity ratios is skewed in all of the caves studied; there are a number of samples with values much higher than the average. Use of the mean values for RUBE dating in DCS is definitely questionable. It might be supposed that to estimate ages of samples older than the U–Th range, the characteristic ratios of the oldest nearby and dated samples could serve as acceptable estimators of initial $^{234}U/^{238}U$ activity ratios. However, the high variability of the calculated initial $^{234}U/^{238}U$ activity ratio that we obtained, their multimodal distribution, and their correlation with sample age exclude such an application.

The high variability of initial $^{234}U/^{238}U$ activity ratios in the speleothems, combined with the different $^{234}U/^{238}U$ activity ratios in the water samples, inspired the question whether these differences can be used as natural paleohydrological proxies. U concentration and isotopic composition in a speleothem is controlled primarily by those variables in the source waters from which they crystallized. The U variables in water strongly depend on the physico-chemical parameters such as the pH, redox/oxidation conditions in source rocks and soils, and underground transit times. Under reducing conditions, U occurs naturally in a stable form and in a tetravalent state

(Bourdon et al. 2003). Under oxidizing conditions U isotopes are highly mobile in the environment, forming uranium ions (UO_2^{2+}) as results of weathering (Markich 2002). In aquatic systems, U occurs mostly in the hexavalent form and creates soluble complexes with phosphate and carbonate (near to neutral conditions), fluorides and sulfates (in lower pH), as well as humic substances like humate (Markich 2002; Bourdon et al. 2003). Availability of complexing ions, concentrations and pH conditions strongly determine the proportions of the different U molecular species (Gascoyne 1992).

Under natural conditions U isotopes are normally in a state of radioactive equilibrium, i.e. $^{234}U/^{238}U$ activity ratio=1. At the time of rock weathering, the enrichment in ^{234}U usually occurs in the fluid phase as an effect of physical fractionation of ^{234}U from ^{238}U during water-rock interaction. The chief causes of $^{234}U/^{238}U$ disequilibrium are: (1) preferential leaching of ^{234}U from sites in the crystal lattice damaged by the alpha decay of ^{238}U, (2) alpha recoil of ^{234}U into the fluid phase, and (3) the lower mass of ^{234}U and its higher mobility.

It is recognized that ^{234}U enrichment of water is achieved much more readily by leaching of ^{234}U from soils or sediments than by direct dissolution of bedrock (Ayalon et al. 1999).
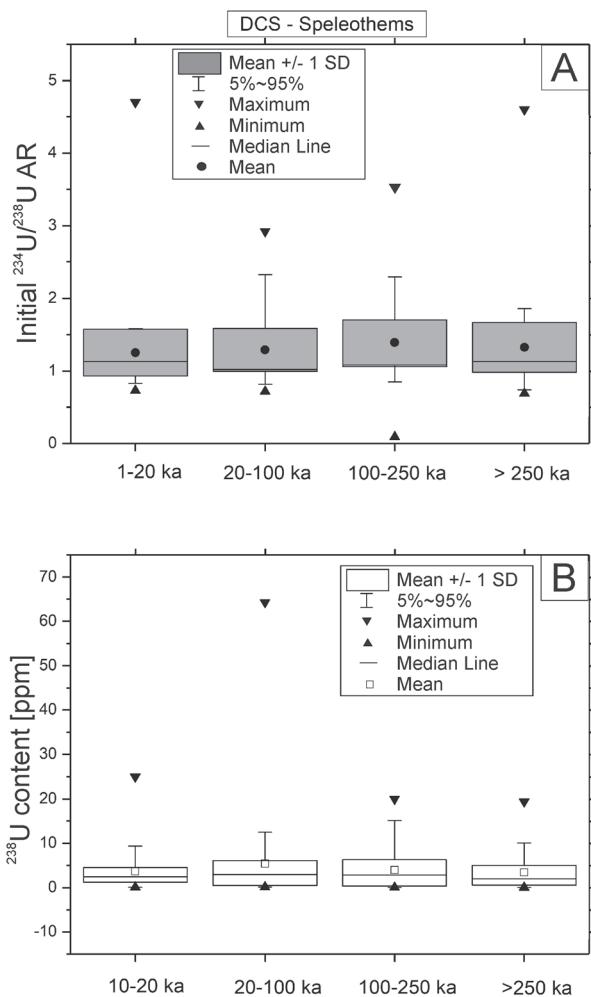
Fig. 7. Statistical relationships between initial $^{234}U/^{238}U$ activity ratios (**A**), uranium content (**B**) and sample age in the DC speleothems.
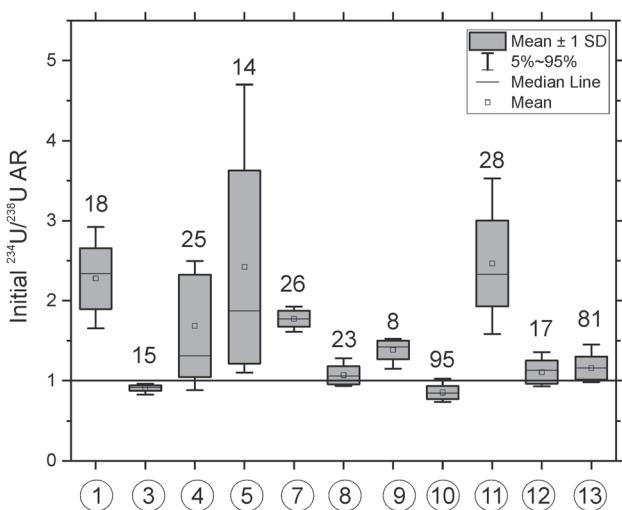
Fig. 8. Variability of initial $^{234}U/^{238}U$ activity ratios of DC speleothems at the different sampling sites. Numbers from 1 to 13 indicate sites as in Fig. 1. Numbers above boxes indicate number of analyses.

Thus, higher values of initial $^{234}U/^{238}U$ activity ratios can be expected in periods when a greater relative proportion of the U derives from leached soil particles or other loose sediments. Lower values are characteristic of periods when a higher relative proportion of the U is derived directly from carbonate rock weathering (Ayalon et al. 1999). Thus, differences in the U isotopic ratios in speleothems deposited at different times can serve as indicators of changes in proportions of ^{234}U being leached from soils/sediments or from the dissolved rocks. In addition, differences in U isotopic composition in samples of the same age can indicate different source water flowpaths with different proportions of U leached from soil/sediment or bedrock dissolution, respectively. Thus, differences in U isotopic composition indirectly indicate the diversity of the alimentation areas, e.g., quantity, type, weathering degree of source deposits, and water circulation paths.

The strong differences in the U isotopic composition in the dripping water samples collected in Veľký dóm and Janáčkov dóm, respectively, suggest different U sources. Dripping water in the Janáčkov dóm (Fig. 1/site 15) is depleted in ^{234}U. This suggests long-time leaching of the source deposits in the alimentation zone. The ^{234}U enrichment in dripping waters in the Veľký dóm and the karst spring suggests relatively "fresh" deposits as the U source. Our investigations found notable differences in U content and isotopic composition: (1) in the different parts of the cave system that were sampled, and (2) over time within some of the individual sampling sites. In some instances, there were changes of U isotopic composition during the process of continuous crystalline precipitation onto an individual speleothem (e.g., in Veľký dóm, Fig. 9B), while in other sites (e.g., Rázcestie, Fig. 8/site 10) the U isotopic ratios, depleted in ^{234}U, were relatively stable over a long time period of the sequence of flowstones and siliciclastic sediments deposition. The oldest flowstones at the bottom of the Rázcestie profile are around 350 ka in age and the youngest are Holocene. All samples were depleted in ^{234}U, suggesting very stable conditions in the groundwater circulation paths and the type and composition of sources in the alimentation area.

In general, the relationships between the U isotopic composition and concentration in different parts of the DC indicate differences in solute composition and proportion, which resulted in different U geochemistry in the precipitated calcites. Two possible mechanisms may be considered: (1) change in the distribution of soil and sediments in the groundwater alimentation areas above cave passages or (2) change in the groundwater percolation paths.

The age of the oldest speleothems from passages with active flowing streams in the central part of the DCL (Rázcestie; around 350 ka) suggests that there were no major changes of geomorphological conditions in the valley over the last 350 ka i.e. no periods of rapid valley incision but, rather, alternating phases of sediment aggradation and entrenchment by surface streams. It suggests no important changes of host rock types, their distribution and exposure in the source areas. Thus, it indicates that changes in the sediment/soil distributions above
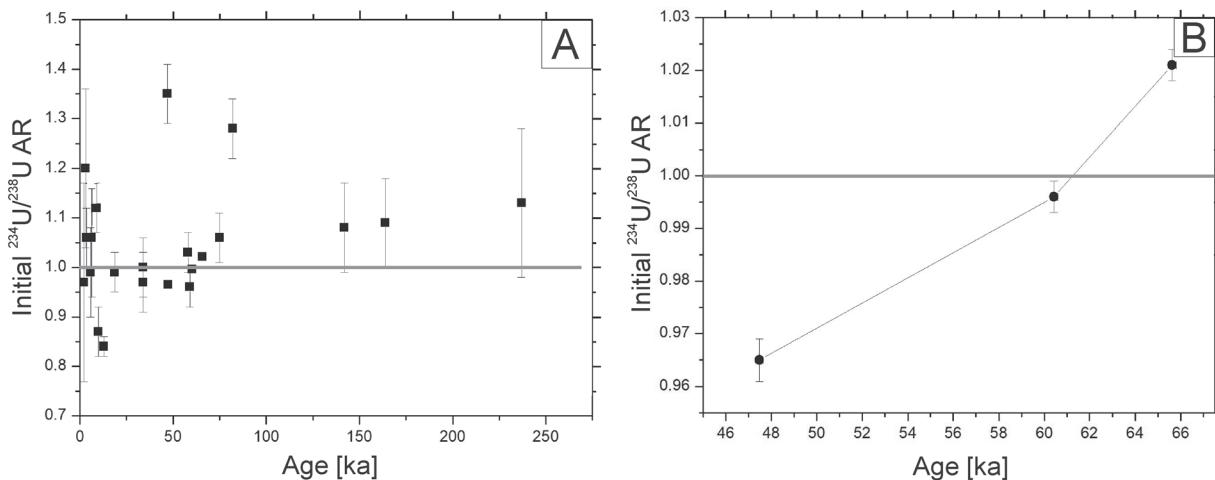
Fig. 9. Variability of initial $^{234}U/^{238}U$ activity ratios in speleothems collected in the Veľký dóm (DCL) (**A**) and results from continuous profile of an individual flowstone (**B**; Lab. No. 233–235 in Supplementary Table S1).
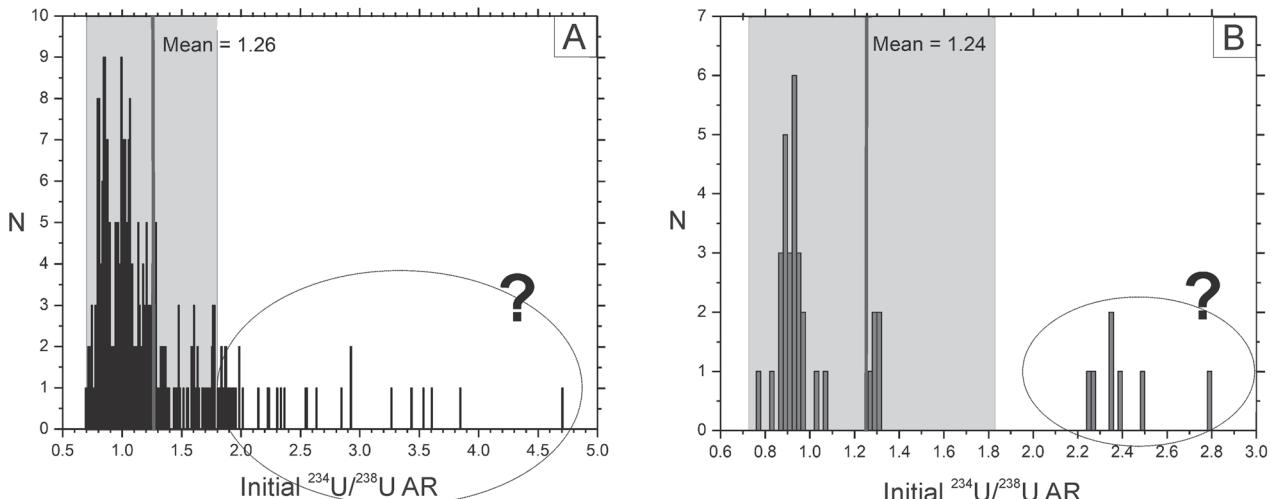
Fig. 10. Mean values of initial $^{234}U/^{238}U$ activity ratios for RUBE estimators for DCL (**A**) and DCP (**B**). Gray rectangles represents one standard deviation range.

cave passages are the most likely to be responsible for variability of U concentrations and U:U ratios recorded here. This implies that the U concentration and isotopic ratios in the water reflect the composition and weathering degree of detrital deposits in their alimentation areas and rocks at the epikarst zone. The periods of ^{234}U enrichment recorded in DC speleothems can be interpreted as periods of intensive erosion and deposition of fresh sediments and/or soils development. The highest parts of the Demänovská dolina, descending to the north from the principal mountains of the Nízke Tatry, were glaciated in the Late Pleistocene and perhaps also at the end of Middle Pleistocene (Vitásek 1923; Volko-Starohorský 1943; Louček et al. 1960; Droppa 1972). This suggests that, at the time of the deglaciations, intensive erosion may have been the means of delivery of "fresh" materials to the DC alimentation areas. Based on this assumption, the results

indicate several periods of such delivery: the interglacials and interstadials at about 120 ka (Eemian, MIS 5e), about 80–70 ka (Early/Middle Weichselian, MIS 5a), and the Holocene (MIS 1; Fig. 2B). The increase in initial $^{234}U/^{238}U$ activity ratios at ~250–200 ka may suggest the effects of older glacial periods (Middle Saalian, MIS 8/7), which is also supported by the increased U content.

Conclusions

Significant differences of U isotopic composition and concentration have been found in the DC speleothems. U concentrations and initial isotopic compositions in them depend on their age and their location within this extensive, multi-level cave system.

Estimation of the initial ^{234}U/^{238}U activity ratios is essential for application of the U–U method for estimating ages of speleothems older than the 500 ka range of ^{230}Th/^{234}U method. The application of the RUBE (Regional Uranium Best Estimate) averaging method, or estimating the initial ^{234}U/^{238}U activity ratios based on the data from younger (U/Th dated) portions of a given speleothem were found to be problematic, essentially inapplicable, in extensive and complex hypergene caves like the DC. Similar problems, likely caused by variability of sediment/soil distribution and composition above the cave may occur in other areas, especially in mountains subject to periodic glaciations or intense periglacial activity.

On the other hand, based on the findings reported above, we conclude that the spatial and temporal variability of the U-content and isotopic ratios in the speleothems can be used as supplemental climatic/hydrological proxies in the DC region. U characteristics (1) reflect changes in U source in space and time, (2) provide information about changes in soil and groundwater circulation paths and/or the distribution/degree of weathering of deposits in the surface alimentation areas, and (3) provide an opportunity to detect specific periods of deglaciation and/or intensive erosion (with deposition of "fresh" sediments in the alimentation areas), estimate their duration and correlate them with other records of regional glaciation. Hence, three periods – at about 120 ka (Eemian interglacial, MIS 5e), around 80–70 ka (Early/Middle Weichselian interstadial, MIS 5a), and during Holocene (MIS 1) – are indicated as probable periods of accelerated erosion with delivery of "fresh" colluvial and other clastic detritus to the alimentation areas, resulting in changes of U isotopic characteristics in growing speleothems in the DC.

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Supplement

Table S1: Results of the U-series dating.

Cave	Nr Lab.	Method	U [ppm]	2σ error	$^{234}U/^{238}U$ AR	2σ error	$^{230}Th/^{234}U$ AR	2σ error	$^{230}Th/^{232}Th$ AR	Age [ka]	2σ error	$^{234}U/^{238}U_{ini}$ AR	2σ error
DIC	N97-1	A	0.28	0.01	1.41	0.03	0.84	0.04	>10,000	169	18	1.6	0.2
DIC	N97-2	A	0.38	0.01	1.42	0.04	0.84	0.03	1,015	169	13	1.7	0.2
DIC	N97-3	A	0.15	0.01	1.54	0.05	0.79	0.03	>10,000	148	10	1.8	0.2
DIC	N97-4	A	0.17	0.01	1.46	0.04	0.76	0.03	>10,000	140	9	1.7	0.2
DIC	N97-5	A	0.17	0.01	2.5	0.09	0.55	0.02	>10,000	79	5	2.9	0.2
DIC	N97-6	A	0.36	0.01	1.99	0.05	0.53	0.02	>10,000	76	4	2.2	0.2
DIC	N97-7	A	0.22	0.01	2.22	0.05	0.5	0.02	>10,000	70	4	2.5	0.2
DIC	N97-8	A	0.49	0.01	2.9	0.05	0.04	0	>10,000	4.7	0.3	2.9	0.2
DIC	N97-9	A	0.16	0.01	2	0.6	0.56	0.02	>10,000	82	5	2.2	0.2
DIC	N97-10	A	0.19	0.01	1.87	0.05	0.52	0.02	>10,000	74	4	2.1	0.2
DIC	N97-11	A	0.3	0.01	2.08	0.05	0.6	0.02	140	91	4	2.4	0.2
DIC	N97-12	A	0.37	0.01	2.13	0.05	0.57	0.02	231	85	4	2.4	0.2
DIC	N97-13	A	0.3	0.01	2.08	0.04	0.55	0.02	662	80	3	2.35	0.1
DIC	N97-14	A	0.48	0.01	1.98	0.04	0.65	0.02	600	102	14	2.3	0.4
DIC	N97-15	A	0.44	0.02	2.1	0.08	0.65	0.03	140	101	7	2.5	0.2
DIC	N97-16	A	0.42	0.01	2.08	0.04	0.66	0.02	>10,000	104	4	2.4	0.2
DIC	N97-17	A	0.29	0.01	2.02	0.07	0.61	0.03	>10,000	93	7	2.3	0.2
DIC	N97-18	A	0.42	0.01	2.66	0.05	0.05	0	>10,000	5.6	0.4	2.68	0.2
Ok	W 1117	A	1.14	0.03	1.11	0.02	0.96	0.02	546	337	60	1.3	0.3
Ok	W 1120	A	0.66	0.02	2.44	0.06	0.95	0.02	210	327	44	4.6	0.6
DCP	W 153	A	14.5	0.3	0.921	0.009	0.11	0	90	12.4	0.3	0.9	0.3
DCP	W 162	A	8	0.2	0.939	0.008	0.03	0	34	3	0.1	0.94	0.09
DCP	W 2021	A	6.1	0.1	0.94	0.09	0.096	0.003	1,768	11	0.3	0.94	0.03
DCP	W 2022	A	4.53	0.09	0.92	0.01	0.095	0.003	365	10.8	0.3	0.91	0.03
DCP	W 2023	A	4.4	0.07	0.93	0.01	0.084	0.003	624	9.5	0.3	0.93	0.03
DCP	W 2024	A	3.43	0.04	0.89	0.02	0.076	0.003	140	8.7	0.3	0.88	0.03
DCP	W 2025	A	3.39	0.04	0.92	0.01	0.066	0.002	468	4.5	0.3	0.92	0.06
DCP	W 2026	A	3.09	0.04	0.93	0.01	0.056	0.002	191	6.3	0.3	0.93	0.05
DCP	W 2032	A	4.04	0.05	0.93	0.02	0.05	0.003	47	5.6	0.3	0.93	0.04
DCP	W 2033	A	3.86	0.07	0.9	0.02	0.027	0.002	42	3	0.2	0.9	0.06
DCP	W 2791	A	2.44	0.04	0.94	0.02	0.103	0.004	44	11.8	0.4	0.94	0.03
DCP	W 2792	A	0.95	0.02	0.96	0.02	0.091	0.004	47	10.4	0.5	0.96	0.05
DCP	W 2793	A	1.13	0.02	0.95	0.02	0.114	0.006	79	13.2	0.7	0.94	0.06
DCP	W 1727	A	20.1	0.7	0.736	0.008	0.114	0.003	1,748	13.2	0.4	0.73	0.02
DCP	W 1728	A	11.2	0.4	0.77	0.02	0.105	0.004	67	12.1	0.4	0.77	0.03
DCP	W 450	A	0.13	0.01	2.1	0.2	0.39	0.03	20	54	5	2.2	0.3
DCP	W 452	A	0.15	0.01	2	0.2	0.44	0.05	24	64	9	2.3	0.4
DCP	W 445	A	0.34	0.01	2.46	0.07	0.09	0.01	55	10.2	0.6	2.5	0.2
DCP	W 446	A	0.4	0.01	2.32	0.06	0.08	0	60	9	0.5	2.4	0.2
DCP	W 1322	A	14.1	0.7	0.777	0.006	0.111	0.002	33	12.8	0.3	0.77	0.01
DCP	W 1512	A	0.69	0.02	1.24	0.04	0.55	0.02	74	84	3	1.31	0.06
DCP	W 1513	A	0.58	0.02	1.25	0.04	0.46	0.02	24	66	3	1.3	0.07
DCP	W 1720	A	0.068	0.004	1.18	0.09	0.82	0.07	36	172	26	1.3	0.3
DCP	W 1779	A	0.94	0.02	1.07	0.03	0.058	0.004	45	6.5	0.4	1.07	0.07
DCP	W 1780	A	0.6	0.02	1.22	0.04	0.47	0.02	87	68	4	1.26	0.08
DCP	W 1852	A	0.95	0.03	0.87	0.02	0.26	0.009	20	33	2	0.9	0.2
DCP	W 1853	A	0.63	0.02	0.96	0.03	0.034	0.004	217	3.7	0.4	1	0.2
DCP	W 1855	A	0.18	0.01	1.22	0.09	0.54	0.05	41	83	9	1.3	0.2
DCP	1058	M	0.398	0.002	2.39	0.05	0.644	0.007	477	101	2	2.39	0.05
DCP	1059	M	0.45	0.03	2.78	0.08	0.088	0.002	549	10	0.3	2.78	0.08
DCP	1060	M	0.493	0.003	2.35	0.05	0.61	0.08	318	93	2	2.35	0.05
DCP	430	M	5.01	0.02	0.934	0.002	0.1075	0.0009	281	12.4	0.2	0.932	0.008
DCP	438	M	4.91	0.02	0.895	0.003	0.088	0.002	566	10.1	0.2	0.89	0.02
DCP	439	M	5.05	0.02	0.89	0.003	0.0701	0.0008	505	7.9	0.1	0.89	0.01
DCP	440	M	5.1	0.02	0.838	0.002	0.0634	0.0009	737	7.1	0.2	0.83	0.01
DCP	431	M	5.02	0.02	0.873	0.003	0.0484	0.0005	130	5.41	0.07	0.87	0.01

Table S1 (continued): Results of the U-series dating.

Cave	Nr Lab.	Method	U [ppm]	2σ error	^{234}U/^{238}U AR	2σ error	^{230}Th/^{234}U AR	2σ error	^{230}Th/^{232}Th AR	Age [ka]	2σ error	^{234}U/^{238}U$_{ini}$ AR	2σ error
DCP	441	M	6.75	0.02	0.885	0.002	0.0367	0.0006	262	4.07	0.07	0.88	0.02
DCP	442	M	4.95	0.02	0.874	0.002	0.0167	0.0005	241	1.83	0.06	0.87	0.02
DCP	W 1523	A	5.5	0.2	0.88	0.02	0.059	0.003	38	6.6	0.3	0.88	0.04
DCP	W 1524	A	1.96	0.06	1.02	0.03	0.013	0.002	68	1.4	0.2	1	0.2
DCL	W 1460	A	0.106	0.008	1.26	0.2	1.08	0.09	25	558	236	2	1
DCL	W 1510	A	0.56	0.03	1.46	0.05	1.01	0.03	35	280	30	2	0.2
DCL	W 1511	A	0.194	0.008	1.47	0.07	0.94	0.04	44	220	20	1.9	0.3
DCL	W 1515	A	0.33	0.02	1.49	0.07	0.8	0.04	82	153	10	1.8	0.2
DCL	W 1518	A	0.248	0.009	3.8	0.2	0.014	0.002	27	1.5	0.2	3.8	0.5
DCL	W 1519	A	0.107	0.006	1.25	0.07	0.1	0.02	335	12	2	1.3	0.3
DCL	W 1530	A	0.31	0.02	1.7	0.07	1.04	0.04	21	281	26	2.6	0.3
DCL	W 1531	A	0.23	0.02	1.33	0.07	0.94	0.05	27	231	24	1.6	0.2
DCL	W 1723	A	0.087	0.008	1.4	0.2	0.86	0.09	22	183	40	1.6	0.4
DCL	W 1831	A	5.01	0.2	1.1	0.02	0.12	0.004	36	13.9	0.4	1.1	0.03
DCL	W 1833	A	0.03	0.003	1.47	0.2	1.1	0.1	277	416	134	1.5	0.6
DCL	W 1856	A	0.078	0.005	2.6	0.2	0.74	0.04	194	124	10	3.3	0.4
DCL	W 2499	A	0.082	0.0007	3.6	0.6	0.12	0.02	46	13	2	3.6	0.6
DCL	W 1506	A	0.24	0.01	4.5	0.2	0.117	0.007	>1,000	13.3	0.7	4.7	0.3
DCL	W 90	A	0.19	0.01	1.7	0.06	0.59	0.01	42	89	3	1.85	0.09
DCL	W 91	A	0.18	0.01	1.57	0.07	0.64	0.02	21	102	4	1.8	0.1
DCL	W 99	A	0.18	0.01	1.64	0.03	0.62	0.01	77	96	3	1.83	0.07
DCL	W 100	A	0.68	0.01	1.47	0.02	0.94	0.01	211	220	9	1.86	0.08
DCL	W 96	A	0.48	0.01	1.49	0.03	0.96	0.02	180	235	13	1.9	0.2
DCL	W 68	A	0.45	0.01	1.5	0.02	0.9	0.01	161	199	7	1.87	0.07
DCL	W 34	A	0.57	0.01	1.42	0.04	0.92	0.02	723	213	12	1.8	0.2
DCL	W 35	A	0.47	0.01	1.48	0.03	0.84	0.02	157	170	7	1.77	0.08
DCL	W 30	A	0.43	0.02	1.53	0.07	0.76	0.03	121	134	9	1.8	0.2
DCL	N 1539	A	0.31	0.01	1.49	0.04	0.71	0.03	313	122	9	1.7	0.2
DCL	W 31	A	0.31	0.01	1.45	0.04	0.89	0.03	236	195	13	1.8	0.2
DCL	W 16	A	0.4	0.01	1.52	0.05	0.92	0.03	308	207	16	1.9	0.2
DCL	705	M	0.788	0.003	1.267	0.003	1.03	0.005	993	354	8	1.72	0.04
DCL	728	M	0.565	0.002	1.272	0.004	1.03	0.02	3,504	360	36	1.7	0.2
DCL	706	M	0.373	0.002	1.474	0.005	0.91	0.01	2,551	204	6	1.84	0.05
DCL	707	M	0.461	0.002	1.626	0.004	0.738	0.005	384	130	2	1.9	0.03
DCL	708	M	0.302	0.002	1.481	0.008	0.74	0.01	274	133	3	1.7	0.04
DCL	999	M	0.507	0.004	1.297	0.004	0.981	0.005	3,440	274	5	1.64	0.03
DCL	1014	M	0.318	0.003	1.495	0.006	0.861	0.008	717	178	4	1.81	0.03
DCL	1015	M	0.358	0.004	1.495	0.006	0.879	0.007	1,915	187	4	1.83	0.03
DCL	1001	M	0.463	0.004	1.296	0.004	0.979	0.006	1,271	273	6	1.63	0.03
DCL	1016	M	0.432	0.004	1.464	0.005	0.868	0.007	5,788	183	3	1.77	0.03
DCL	995	M	0.579	0.004	1.249	0.004	0.948	0.006	2,050	251	5	1.86	0.04
DCL	1013	M	0.299	0.003	1.49	0.005	0.929	0.007	790	214	4	1.89	0.04
DCL	1002	M	0.47	0.004	1.395	0.005	0.984	0.008	2,381	263	7	1.82	0.04
DCL	994	M	0.702	0.006	1.266	0.004	0.998	0.006	2,038	302	7	1.62	0.04
DCL	998	M	0.462	0.004	1.279	0.005	0.984	0.008	983	281	9	1.61	0.05
DCL	1000	M	0.486	0.004	1.281	0.005	0.978	0.009	21,155	275	9	1.6	0.05
DCL	1011	M	0.438	0.004	1.36	0.005	0.978	0.007	1,657	262	6	1.75	0.04
DCL	W 1502	A	11.3	0.4	0.85	0.02	0.113	0.002	98	13	0.2	0.84	0.02
DCL	W 1462	A	0.98	0.04	0.98	0.04	0.021	0.004	27	2.3	0.4	0.97	0.2
DCL	W 1546	A	1.4	0.2	1.06	0.04	0.79	0.03	369	164	12	1.09	0.09
DCL	W 1611	A	0.41	0.02	1.05	0.04	0.74	0.03	112	142	10	1.08	0.09
DCL	W 1717	A	2.8	0.2	1.05	0.03	0.5	0.02	527	75	3	1.06	0.05
DCL	W 1718	A	2.21	0.06	1.06	0.02	0.032	0.002	475	3.6	0.2	1.06	0.06
DCL	W 1724	A	6.1	0.3	0.88	0.02	0.089	0.005	30	10.1	0.5	0.87	0.05
DCL	W 2496	A	1.5	0.06	1.1	0.2	0.057	0.006	184	6.4	0.7	1.1	0.2
DCL	W 2497	A	3	0.2	0.99	0.09	0.052	0.005	95	5.8	0.5	0.99	0.09
DCL	W 2521	A	1	0.02	1.12	0.05	0.079	0.004	21	8.9	0.4	1.12	0.05
DCL	W 2522	A	1.42	0.02	0.99	0.04	0.16	0.005	51	18.8	0.6	0.99	0.04
DCL	W 2729	A	0.98	0.07	1.07	0.05	0.9	0.04	237	237	30	1.1	0.2

Table S1 (continued): Results of the U-series dating.

Cave	Nr Lab.	Method	U [ppm]	2σ error	$^{234}U/^{238}U$ AR	2σ error	$^{230}Th/^{234}U$ AR	2σ error	$^{230}Th/^{232}Th$ AR	Age [ka]	2σ error	$^{234}U/^{238}U_{ini}$ AR	2σ error
DCL	W 2749	A	5.6	0.2	0.96	0.02	0.42	0.02	326	59	2	0.96	0.04
DCL	W 2750	A	2.6	0.1	0.97	0.03	0.27	0.02	823	34	2	0.97	0.06
DCL	W 2751	A	4	0.2	1	0.02	0.272	0.008	641	34	2	1	0.06
DCL	W 2752	A	6.3	0.2	1.02	0.02	0.415	0.008	197	58	2	1.03	0.04
DCL	W 2775	A	0.49	0.02	1.31	0.04	0.36	0.01	97	47	2	1.35	0.06
DCL	W 2776	A	0.312	0.009	1.22	0.04	0.54	0.02	303	82	4	1.28	0.06
DCL	W 2777	A	0.96	0.02	1.16	0.02	0.055	0.003	24	6.1	0.4	1.17	0.08
DCL	W 2778	A	0.47	0.02	1.19	0.04	0.028	0.004	102	3.1	0.4	1.2	0.2
DCL	233	M	11.56	0.03	1.018	0.001	0.4538	0.0009	897	65.6	0.2	1.021	0.003
DCL	234	M	7.59	0.02	0.9963	0.0009	0.4262	0.0008	1,418	60.4	0.2	0.996	0.003
DCL	235	M	5.71	0.02	0.969	0.001	0.353	0.0009	877	47.5	0.2	0.965	0.004
DCL	N 1099	A	2.74	0.05	1.47	0.02	0.09	0	382	10.3	0.5	1.49	0.07
DCL	N 1100	A	4.3	0.07	1.32	0.07	0.03	0	155	3.4	0.2	1.32	0.09
DCL	N 1101	A	4.27	0.07	1.33	0.02	0.03	0	>10,000	3.2	0.2	1.33	0.07
DCL	N 1102	A	5.6	0.2	1.15	0.02	0.01	0	>10,000	0.82	0.09	1.2	0.2
DCL	W 7	A	3.14	0.07	1.45	0.03	0.09	0	67	10	0.4	1.46	0.06
DCL	W 8	A	3.78	0.07	1.46	0.02	0.05	0	261	5.7	0.2	1.47	0.05
DCL	W 18	A	3.95	0.07	1.47	0.02	0.03	0	23	3.05	0.05	1.47	0.03
DCL	W 9	A	3.69	0.05	1.38	0.02	0.01	0	28	1.42	0.03	1.38	0.03
DCL	W 1115	A	0.49	0.01	1.13	0.04	0.91	0.03	42	265	40	1.3	0.2
DCL	W 1125	A	4.4	0.3	0.96	0.02	0.93	0.02	938	290	30	0.9	0.1
DCL	W 1126	A	2.5	0.2	1.03	0.03	0.84	0.02	422	198	16	1.05	0.09
DCL	W 1127	A	5.4	0.2	1.02	0.02	0.83	0.02	928	193	10	1.04	0.06
DCL	W 1116	A	0.62	0.02	0.98	0.03	0.93	0.03	128	286	50	0.9	0.2
DCL	W 347	A	4.8	0.4	0.85	0.02	0.54	0.05	146	87	12	0.8	0.2
DCL	W 346	A	8.8	0.5	0.85	0.02	0.54	0.03	199	88	7	0.8	0.07
DCL	W 224	A	8.1	0.2	0.88	0.01	0.51	0.01	847	79	3	0.85	0.03
DCL	W 252	A	6.3	0.2	0.875	0.008	0.47	0.01	522	71	3	0.85	0.04
DCL	W 350	A	9	0.6	0.802	0.005	0.06	0	65	6.7	0.5	0.8	0.06
DCL	W 403	A	3.24	0.06	0.85	0.02	0.55	0.01	20	85	2	0.81	0.02
DCL	W 404	A	3.5	0.2	0.9	0.02	0.54	0.01	21	83	4	0.87	0.04
DCL	W 348	A	2.8	0.2	0.86	0.01	0.24	0.04	38	29	6	0.9	0.2
DCL	W 203	A	6.8	0.2	0.89	0.02	0.49	0.01	23	75	3	0.86	0.04
DCL	W 253	A	4.14	0.07	0.771	0.008	0.48	0.02	199	73	4	0.72	0.04
DCL	W 419	A	3.68	0.05	0.85	0.02	0.55	0.01	561	86	3	0.8	0.03
DCL	W 328	A	3.1	0.2	0.85	0.02	0.6	0.04	171	102	13	0.8	0.1
DCL	W 979	A	5.3	0.3	0.85	0.02	0.52	0.01	205	80	2	0.82	0.02
DCL	W 980	A	3.1	0.2	0.88	0.02	0.52	0.01	463	79	3	0.95	0.04
DCL	W 417	A	3.1	0.2	0.86	0.02	0.49	0.01	248	73	3	0.83	0.04
DCL	W 329	A	2.8	0.2	0.871	0.009	0.48	0.03	126	72	6	0.84	0.07
DCL	W 402	A	30	2	0.863	0.009	0.48	0.01	845	70	2	0.83	0.02
DCL	W 362	A	64.3	4.2	0.865	0.008	0.41	0.04	136	58	8	0.8	0.2
DCL	W 345	A	43.5	2.7	0.883	0.005	0.44	0.03	633	65	6	0.86	0.08
DCL	W 155	A	10.2	0.2	0.897	0.007	0.45	0.01	730	65	2	0.88	0.03
DCL	W 156	A	6.4	0.2	0.88	0.02	0.56	0.01	319	92	3	0.85	0.03
DCL	W 88	A	6.8	0.1	0.841	0.006	0.52	0.01	905	82	2	0.8	0.02
DCL	N 1513	A	7.6	0.2	0.837	0.005	0.52	0.01	20,028	82	4	0.8	0.03
DCL	N 1514	A	5.98	0.09	0.859	0.006	0.51	0.01	>10,000	78	4	0.83	0.03
DCL	W 89	A	5.93	0.08	0.865	0.007	0.5	0.01	3,242	76	2	0.83	0.02
DCL	W 361	A	12.4	0.7	0.88	0.01	0.46	0.02	1,412	69	4	0.86	0.05
DCL	W 359	A	4.6	0.3	0.91	0.02	0.35	0.02	21	46	3	0.9	0.06
DCL	W 864	A	14	0.9	1.07	0.02	0.81	0.01	494	178	6	1.11	0.04
DCL	W 82	A	6.8	0.2	0.82	0.008	0.01	0	50	1.52	0.04	0.82	0.02
DCL	N 1511	A	22.7	0.4	0.837	0.004	0.5	0.01	575	78	3	0.74	0.05
DCL	W 97	A	5.18	0.06	0.891	0.005	0.37	0.01	81	51	1	0.87	0.02
DCL	N 1512	A	6.27	0.09	0.89	0.07	0.42	0.01	76	60	2	0.87	0.07
DCL	W 225	A	8	0.2	1.001	0.006	0.71	0.02	83	133	8	1	0.06
DCL	W 401	A	7.2	0.3	0.881	0.009	0.5	0.01	55	76	2	0.86	0.02
DCL	W 254	A	18.6	0.3	0.93	0.004	0.85	0.02	1,210	214	15	0.87	0.06

Table S1 (continued): Results of the U-series dating.

Cave	Nr Lab.	Method	U [ppm]	2σ error	$^{234}U/^{238}U$ AR	2σ error	$^{230}Th/^{234}U$ AR	2σ error	$^{230}Th/^{232}Th$ AR	Age [ka]	2σ error	$^{234}U/^{238}U_{ini}$ AR	2σ error
DCL	W 213	A	15.1	0.3	0.994	0.005	0.78	0.03	906	165	14	0.99	0.08
DCL	W 226	A	20	0.5	0.961	0.006	0.65	0.03	4,499	115	11	0.95	0.09
DCL	W 1373	A	7.7	0.2	0.886	0.006	0.448	0.004	345	65.5	0.8	0.86	0.01
DCL	W 1558	A	12.4	0.5	0.88	0.02	0.83	0.02	9,833	209	8	0.79	0.03
DCL	W 1559	A	2.8	0.2	0.9	0.03	0.94	0.03	257	390	60	0.7	0.2
DCL	W 1612	A	3.6	0.1	0.85	0.02	0.116	0.004	23	13.5	0.4	0.84	0.03
DCL	W 2480	A	12.5	0.3	0.82	0.02	0.315	0.005	84	41.6	0.7	0.82	0.02
DCL	W 2481	A	18.5	0.4	0.85	0.02	0.307	0.004	50	40.2	0.5	0.85	0.02
DCL	W 2516	A	11.6	0.3	0.86	0.03	0.8	0.02	21	186	6	0.86	0.03
DCL	W 2527	A	8.7	0.1	0.91	0.02	0.705	0.009	568	135	3	0.91	0.02
DCL	W 2498	A	19.6	0.8	0.84	0.02	0.685	0.009	>1,000	130	3	0.84	0.02
DCL	W 2520	A	8.3	0.3	0.78	0.04	0.84	0.02	888	219	11	0.78	0.04
DCL	W 2523	A	4.94	0.06	0.78	0.02	0.66	0.01	79	124	3	0.78	0.02
DCL	W 2528	A	2.64	0.06	0.88	0.05	0.74	0.02	100	153	7	0.88	0.05
DCL	W 2529	A	2.13	0.04	0.9	0.05	0.75	0.02	98	157	7	0.9	0.05
DCL	W 2483	A	11.3	0.2	0.85	0.02	0.829	0.009	106	204	5	0.85	0.02
DCL	W 2517	A	4.21	0.06	0.79	0.03	0.81	0.01	128	198	6	0.79	0.03
DCL	W 2482	A	5.1	0.1	0.8	0.04	0.86	0.02	>1,000	236	10	0.8	0.04
DCL	W 2495	A	4.4	0.2	0.9	0.2	0.95	0.02	>1,000	342	40	0.9	0.2
DCL	W 2518	A	2.53	0.03	1.12	0.09	0.97	0.02	41	348	29	1.12	0.09
DCL	W 2471	A	2.7	0.1	0.79	0.07	0.9	0.02	154	288	24	0.79	0.07
DCL	W 2478	A	2.7	0.2	0.77	0.08	0.86	0.03	181	240	23	0.77	0.08
DCL	W 2479	A	5.7	0.2	0.69	0.06	0.91	0.02	87	327	25	0.69	0.06
DCL	W 2731	A	1.97	0.09	0.94	0.04	0.93	0.04	1,572	317	60	0.85	0.16
DCL	W 2733	A	3.2	0.3	0.8	0.03	0.084	0.008	36	9.6	0.9	0.8	0.08
DCL	W 2734	A	4.1	0.4	0.84	0.03	0.15	0.01	546	18	1	0.83	0.06
DCL	W 2794	A	0.42	0.01	1.55	0.05	0.169	0.008	93	20	1	1.58	0.09
DCL	W 2795	A	1.32	0.03	1.54	0.04	0.059	0.004	36	6.6	0.4	1.6	0.1
DCL	W 2925	A	4.5	0.2	0.98	0.02	0.92	0.02	233	292	26	0.98	0.02
DCL	W 2927	A	4.5	0.1	0.94	0.02	0.88	0.02	374	245	12	0.94	0.02
DCL	W 2926	A	1.97	0.06	0.96	0.02	0.97	0.02	100	492	96	0.96	0.02
DCL	879	M	8.14	0.06	0.824	0.002	0.594	0.003	258	103.9	0.6	0.765	0.005
DCL	880	M	4.66	0.03	0.841	0.002	0.566	0.004	361	95.2	0.9	0.792	0.007
DCL	881	M	7.42	0.06	0.915	0.002	0.472	0.002	1,814	70.9	0.4	0.896	0.004
DCL	882	M	5.7	0.04	0.884	0.002	0.517	0.002	733	81.6	0.5	0.854	0.004
DCL	884	M	3.33	0.03	0.9	0.002	0.239	0.003	1,567	30.1	0.5	0.89	0.01
DCL	892	M	51.8	0.4	1.02	0.002	0.443	0.002	6,926	64	0.3	1.024	0.003
DCL	893	M	3.16	0.03	0.897	0.002	0.254	0.003	296	32.2	0.5	0.89	0.01
DCL	894	M	5.32	0.04	0.88	0.002	0.275	0.003	936	35.6	0.4	0.867	0.007
DCL	897	M	6.16	0.05	0.874	0.002	0.413	0.002	275	59.3	0.3	0.851	0.004
DCL	898	M	3.9	0.03	0.883	0.002	0.324	0.003	981	43.3	0.5	0.868	0.008
DCL	899	M	3.92	0.03	0.892	0.002	0.377	0.003	1,036	52.5	0.5	0.874	0.007
DCL	929	M	9.61	0.05	1.033	0.002	0.816	0.001	222,407	182.4	0.5	1.055	0.003
DCL	931	M	5.01	0.03	0.889	0.001	0.919	0.002	346	355	7	0.7	0.01
DCL	932	M	9.4	0.05	0.8361	0.0009	0.0928	0.0003	262	10.72	0.05	0.831	0.003
DCL	933	M	8.29	0.05	0.863	0.0009	0.0934	0.0004	218	10.79	0.06	0.859	0.004
DCL	957	M	10.07	0.05	0.957	0.002	0.936	0.004	5,003	330	8	0.89	0.02
DCL	206	M	14.43	0.03	0.954	0.002	0.93	0.01	>100,000	325	28	0.89	0.08
DCL	211	M	7.677	0.008	1.0158	0.0009	0.9789	0	395	395	5	1.05	0.01
DCL	653	M	10.25	0.04	0.8031	0.0009	0.0436	0.0002	220	4.9	0.03	0.8	0.005
DCL	654	M	10.06	0.05	0.8139	0.001	0.0787	0.0006	284	9.02	0.08	0.809	0.007
DCL	655	M	11.18	0.05	0.8187	0.0008	0.0885	0.0007	244	10.19	0.09	0.813	0.007
DCL	656	M	9.19	0.04	0.8878	0.001	0.901	0.003	1,988	305	5	0.74	0.01
DCL	657	M	19.39	0.08	0.9065	0.0009	0.917	0.003	1,304	328	6	0.77	0.01
DCL	658	M	5.09	0.03	0.891	0.002	0.901	0.004	286	304	7	0.74	0.02
DCL	1087	M	0.796	0.004	2.63	0.03	0.736	0.04	3,750	124	1	2.63	0.03
DCL	1090	M	1.002	0.006	2.84	0.03	0.0843	0.0009	664	9.6	0.1	2.84	0.03
DCL	1084	M	0.301	0.002	1.98	0.05	0.679	0.009	344	112	3	1.98	0.05
DCL	1085	M	0.926	0.006	1.2	0.2	0.96	0.02	737	306	27	1.2	0.2

Table S1 (continued): Results of the U-series dating.

Cave	Nr Lab.	Method	U [ppm]	2σ error	234U/238U AR	2σ error	230Th/234U AR	2σ error	230Th/232Th AR	Age [ka]	2σ error	234U/238U_ini AR	2σ error
DCL	1086	M	1.467	0.009	1.2	0.2	1.005	0.02	930	500	76	1.2	0.2
DCL	1093	M	6.32	0.04	0.74	0.01	0.662	0.005	10,108	128	2	0.74	0.01
DCL	1094	M	0.804	0.005	1.67	0.02	0.536	0.005	1,212	80	1	1.67	0.02
DCL	1095	M	1.69	0.01	1.6	0.02	0.165	0.002	3,678	19.5	0.2	1.6	0.02
DCL	1088	M	0.731	0.005	1.36	0.08	0.99	0.01	1,625	325	18	1.36	0.08
DCL	1089	M	0.331	0.002	1.8	0.08	0.87	0.02	2,283	183	8	1.8	0.08
DCL	1091	M	4.44	0.03	1.13	0.06	0.992	0.006	11,026	417	23	1.13	0.06
DCL	1092	M	11.94	0.07	1.2	0.07	1.01	0.003	45,679	570	35	1.2	0.07
DCL	W 114	A	0.38	0.01	1.3	0.05	0.94	0.03	41	235	24	1.6	0.2
DCL	W 170	A	0.31	0.02	2.6	0.2	0.79	0.05	143	137	14	3.4	0.4
DCL	W 192	A	0.25	0.01	2.1	0.08	0.73	0.04	104	123	9	2.6	0.3
DCL	W 201	A	0.28	0.05	2.4	0.5	0.6	0.3	41	78	45	3	2
DCL	W 178	A	0.28	0.02	2.1	0.2	0.54	0.04	79	78	8	2.3	0.3
DCL	914	M	0.277	0.002	2.556	0.009	0.527	0.006	255	75	2	2.92	0.04
DCL	915	M	0.225	0.002	2.12	0.009	0.692	0.007	268	113	2	2.54	0.05
DCL	916	M	0.245	0.002	2	0.01	0.68	0.01	429	110	3	2.36	0.05
DCL	917	M	0.193	0.002	1.83	0.02	0.785	0.009	233	142	3	2.23	0.05
DCL	918	M	0.295	0.002	1.99	0.02	0.615	0.009	632	95	3	2.3	0.05
DCL	919	M	0.216	0.002	1.755	0.007	0.606	0.008	287	94	2	1.98	0.04
DCL	635	M	0.271	0.003	2.3	0.02	0.79	0.01	787	139	3	2.92	0.06
DCL	347	M	0.241	0.0009	2.75	0.002	0.777	0.007	453	132	2	3.53	0.05
DCL	638	M	0.284	0.003	1.82	0.02	0.71	0.01	433	118	3	2.14	0.06
DCL	348	M	0.234	0.002	1.742	0.006	0.593	0.008	290	90	2	1.95	0.04
DCL	637	M	0.296	0.003	1.95	0.02	0.58	0.02	2,397	87	0.7	2.22	0.02
DCL	219	M	4.96	0.03	1.047	0.002	0.803	0.003	291	172	2	1.08	0.01
DCL	220	M	4.98	0.02	1.094	0.002	0.751	0.003	770	146.1	0.8	1.142	0.008
DCL	221	M	2.16	0.007	1.035	0.003	0.713	0.002	239	134.1	0.6	1.051	0.005
DCL	222	M	4.45	0.02	1.042	0.001	0.708	0.002	3,354	132.1	0.8	1.061	0.006
DCL	223	M	3.98	0.02	1.047	0.002	0.699	0.002	4,028	128.7	0.4	1.068	0.004
DCL	224	M	3.452	0.008	1.064	0.002	0.707	0.003	2,640	131	1	1.092	0.008
DCL	W 85	A	5.58	0.09	0.976	0.009	0.94	0.02	874	314	40	0.9	0.2
DCL	W 86	A	4.19	0.08	0.97	0.02	0.96	0.03	238	393	80	0.9	0.2
DCL	W 135	A	5	0.1	0.98	0.02	0.79	0.04	273	172	20	1	0.2
DCL	W 87	A	5.65	0.09	0.978	0.009	0.96	0.02	954	368	60	0.9	0.2
DCL	W 120	A	5.1	0.1	1.08	0.02	0.72	0.02	44	136	7	1.12	0.06
DCL	W 358	A	5.4	0.2	1.14	0.02	0.62	0.01	475	104	3	1.1	0.4
DCL	W 421	A	3.37	0.03	1.11	0.008	0.45	0	156	65	0.8	1.13	0.02
DCL	W 363	A	1.83	0.04	1.22	0.02	0.1	0	157	11.1	0.4	1.23	0.04
DCL	W 418	A	1.83	0.02	1.35	0.02	0.1	0	25	10.8	0.2	1.36	0.03
DCL	W 420	A	1.75	0.03	1.25	0.02	0.05	0	32	5.7	0.2	1.25	0.05
DCL	W 360	A	2.68	0.07	1.17	0.02	0.02	0	26	1.7	0.2	1.2	0.2
DCL	W 1443	A	2.2	0.1	1.15	0.05	0.174	0.02	45	21	2	1.16	0.09
DCL	W 1444	A	1.71	0.05	1.15	0.03	0.293	0.008	128	37	1	1.17	0.04
DCL	W 1467	A	2.76	0.07	1.27	0.02	0.026	0.002	29	2.8	0.2	1.28	0.09
DCL	W 1468	A	2.32	0.09	1.23	0.03	0.02	0.002	21	2.2	0.2	1.2	0.2
DCL	W 1475	A	2.5	0.1	1.15	0.03	0.12	0.004	186	13.9	0.5	1.22	0.05
DCL	W 1476	A	3.08	0.08	1.18	0.02	0.086	0.003	152	9.7	0.3	1.19	0.04
DCL	W 1477	A	2.88	0.08	1.13	0.02	0.294	0.008	547	37	1	1.14	0.04
DCL	W 1503	A	2.95	0.06	1.12	0.02	0.076	0.003	95	8.6	0.3	1.13	0.04
DCL	W 1504	A	2.88	0.07	1.15	0.02	0.072	0.003	46	8.1	0.3	1.15	0.04
DCL	W 1509	A	3.28	0.07	1.16	0.02	0.089	0.004	28	10.1	0.4	1.16	0.05
DCL	W 1514	A	3	0.1	1.25	0.02	0.022	0.002	27	2.4	0.2	1.2	0.2
DCL	W 1516	A	25	0.2	1.197	0.005	0.0424	0.0005	112	4.7	0.06	1.2	0.02
DCL	W 1585	A	0.9	0.03	1.06	0.04	0.52	0.03	81	78	4	1.08	0.07
DCL	W 1587	A	1.49	0.05	0.8	0.03	0.059	0.005	26	6.6	0.6	0.79	0.07
DCL	W 1588	A	2.32	0.05	1.03	0.02	0.67	0.02	179	119	4	1.04	0.04
DCL	W 1589	A	3.45	0.07	1.01	0.02	0.64	0.02	266	109	3	1.01	0.03
DCL	W 1590	A	1.9	0.1	0.85	0.03	0.103	0.007	66	11.8	0.7	0.84	0.05
DCL	W 1595	A	1.59	0.04	1.06	0.02	0.48	0.01	484	71	2	1.07	0.04

Table S1 (continued): Results of the U-series dating.

Cave	Nr Lab.	Method	U [ppm]	2σ error	$^{234}U/^{238}U$ AR	2σ error	$^{230}Th/^{234}U$ AR	2σ error	$^{230}Th/^{232}Th$ AR	Age [ka]	2σ error	$^{234}U/^{238}U_{ini}$ AR	2σ error
DCL	W 1596	A	1.4	0.03	1.08	0.02	0.5	0.02	96	74	2	1.1	0.04
DCL	W 1598	A	1.62	0.05	0.99	0.03	0.84	0.03	63	197	11	0.99	0.06
DCL	W 1599	A	2.01	0.05	1.02	0.03	0.59	0.02	148	96	3	1.02	0.04
DCL	W 1600	A	3.79	0.08	1.04	0.02	0.64	0.02	1,157	109	3	1.06	0.03
DCL	W 2649	A	0.95	0.02	1.44	0.05	0.099	0.004	>1,000	11.3	0.5	1.44	0.05
DCL	W 2651	A	1.72	0.02	1.08	0.04	0.095	0.004	26	10.8	0.4	1.08	0.04
DCL	W 2661	A	1.89	0.02	1.18	0.04	0.063	0.002	167	7.1	0.3	1.18	0.04
DCL	W 2662	A	1.44	0.02	1.2	0.05	0.043	0.002	33	4.7	0.2	1.2	0.05
DCL	W 2663	A	1.11	0.02	1.18	0.06	0.037	0.003	25	4.1	0.3	1.18	0.06
DCL	W 2664	A	0.79	0.02	1.24	0.07	0.033	0.002	17	3.6	0.3	1.24	0.07
DCL	W 2665	A	1.32	0.02	1.19	0.09	0.027	0.002	>1,000	2.9	0.2	1.19	0.09
DCL	W 2666	A	0.786	0.009	1.36	0.09	0.027	0.002	20	3	0.2	1.36	0.09
DCL	W 2470	A	2.3	0.1	1	0.2	0.031	0.004	34	3.4	0.4	0.95	0.12
DCL	W 2468	A	2.3	0.1	1.1	0.1	0.048	0.005	58	5.4	0.4	1.09	0.1
DCL	W 2438	A	0.94	0.03	0.83	0.07	0.12	0.01	157	15	2	0.83	0.07
DCL	458	M	0.875	0.003	1.266	0.004	0.749	0.005	215	139	2	1.39	0.02
DCL	459	M	0.872	0.002	1.362	0.003	0.609	0.006	240	97	2	1.47	0.02
DCL	190	M	0.707	0.002	1.345	0.003	0.618	0.003	279	99.1	0.8	1.45	0.03
DCL	462	M	0.914	0.003	1.331	0.004	0.595	0.008	1,165	94	2	1.43	0.03
DCL	193	M	0.77	0.002	1.41	0.004	0.61	0.004	790	96.5	0.8	1.54	0.02
DCL	463	M	0.74	0.002	1.384	0.003	0.634	0.008	1,505	103	2	1.51	0.03
DCL	464	M	0.542	0.002	1.254	0.003	0.586	0.007	224	92	2	1.33	0.03
DCL	194	M	0.74	0.002	1.275	0.003	0.395	0.003	170	53.5	0.4	1.32	0.01
DCL	1105	M	0.75	0.005	1.269	0.008	0.133	0.004	281	15.6	0.5	1.28	0.03
DCL	1106	M	1.479	0.009	1.219	0.004	0.103	0.003	671	11.9	0.3	1.23	0.03
DCL	1107	M	1.183	0.007	1.215	0.005	0.082	0.001	338	9.4	0.2	1.22	0.01
DCL	1108	M	1.386	0.008	1.235	0.004	0.084	0.002	332	9.5	0.2	1.24	0.03
DCL	1109	M	1.243	0.007	1.242	0.002	0.056	0.002	585	6.3	0.2	1.25	0.04
DCL	1110	M	1.466	0.009	1.256	0.005	0.046	0.001	1,551	5.1	0.2	1.26	0.03
DCL	1111	M	1.468	0.009	1.249	0.002	0.055	0.002	667	6.2	0.2	1.25	0.02
DCL	1112	M	2.09	0.02	1.278	0.007	0.087	0.004	562	9.9	0.4	1.29	0.05
DCL	1113	M	2.24	0.02	1.278	0.003	0.049	0.001	293	5.5	0.2	1.28	0.02
DCL	1114	M	2.31	0.02	1.302	0.007	0.023	0.002	215	2.6	0.2	1.3	0.05
DCL	784	M	5.41	0.03	0.979	0.002	0.868	0.004	11,414	227	3	0.96	0.01
DCL	785	M	7.86	0.04	0.999	0.003	0.859	0.006	11,334	215	4	1	0.02
DCL	786	M	3.71	0.02	1.003	0.003	0.866	0.008	8,694	220	6	1.01	0.03
DCL	787	M	5.69	0.03	0.997	0.003	0.865	0.003	4,445	220	2	0.994	0.009
DCL	788	M	4.46	0.02	1.008	0.002	0.86	0.003	2,356	214	2	1.02	0.01
DCL	789	M	3.53	0.02	0.999	0.002	0.881	0.004	4,656	234	3	1	0.02
DCL	790	M	5.59	0.03	1.007	0.002	0.86	0.003	13,961	214	2	1.01	0.01
DCL	791	M	6.44	0.03	0.988	0.002	0.863	0.002	19,528	220	2	0.977	0.009
DCL	792	M	6.65	0.03	0.983	0.002	0.861	0.003	23,823	220	2	0.969	0.009
DCL	793	M	6.47	0.04	1.005	0.002	0.878	0.003	9,717	230	3	1.01	0.009
DCL	794	M	6.92	0.03	0.99	0.002	0.781	0.003	18,223	168	2	0.98	0.01
DCL	795	M	6.54	0.03	1.016	0.003	0.859	0.004	28,486	212	3	1.03	0.01
DCL	796	M	3.25	0.03	1.016	0.002	0.849	0.005	4,724	205	3	1.03	0.02
DCL	797	M	6.26	0.04	1.03	0.002	0.836	0.003	4,753	195	2	1.05	0.01
DCL	798	M	7.94	0.05	1.011	0.002	0.698	0.003	488	130.7	0.8	1.016	0.006
DCL	799	M	8.22	0.05	1.022	0.002	0.694	0.003	868	129	1	1.032	0.007
DCL	800	M	9.7	0.06	1.013	0.002	0.697	0.004	4,416	130	2	1.019	0.008
DCL	801	M	8.72	0.05	1.016	0.002	0.698	0.003	867	130	1	1.024	0.007
DCL	802	M	8.2	0.1	1.029	0.003	0.701	0.006	2,057	131	2	1.04	0.02
DCL	609	M	5.58	0.03	0.998	0.002	0.834	0.003	31,050	196	2	0.997	0.01
DCL	610	M	5.89	0.03	1.008	0.002	0.868	0.004	22,258	220	3	1.02	0.02
DCL	611	M	3.77	0.03	1.014	0.003	0.868	0.006	10,431	219	4	1.02	0.02
DCL	612	M	2.66	0.02	1.031	0.003	0.9	0.01	4,922	238	9	1.06	0.04
DCL	613	M	5.73	0.05	1.031	0.003	0.838	0.003	13,800	194	2	1.05	0.02
DCL	614	M	6.38	0.04	1.03	0.002	0.699	0.003	1,142	129.8	0.9	1.043	0.007
DCL	615	M	7.32	0.05	1.028	0.002	0.702	0.003	1,191	130.8	0.8	1.04	0.006

Table S1 (continued): Results of the U-series dating.

Cave	Nr Lab.	Method	U [ppm]	2σ error	$^{234}U/^{238}U$ AR	2σ error	$^{230}Th/^{234}U$ AR	2σ error	$^{230}Th/^{232}Th$ AR	Age [ka]	2σ error	$^{234}U/^{238}U_{ini}$ AR	2σ error
DCL	616	M	7.68	0.04	1.035	0.002	0.704	0.003	1,541	131.4	0.8	1.05	0.006
DCL	662	M	3.14	0.02	1.17	0.03	0.316	0.003	2,987	42.8	0.4	1.17	0.03
DCL	548	M	2.29	0.01	1.21	0.04	0.488	0.003	2,479	71.3	0.5	1.21	0.04
DCL	W 1547	A	1.7	0.2	1.15	0.04	0.41	0.02	286	56	3	1.18	0.07
ST	668	M	2.822	0.009	0.967	0.002	0.0461	0.0008	470	5.178	0.1	0.97	0.02
ST	759	M	1.964	0.006	0.994	0.002	0.972	0.004	367	400.7	12.4	0.98	0.03
ST	873	M	0.125	0.0004	1.699	0.007	0.408	0.006	280	55.26	1.03	1.82	0.03

Explanations: Calculations use the decay constant for: ^{238}U of Jaffey et al. (1971); ^{234}U and ^{230}Th of Cheng et al. (2013) and ^{232}Th of Holden (1990). Reported errors are equal 2σ; AR — aktivity ratio; DCL — Demänová Cave of Liberty; DCP — Demänová Cave of Peace; DIC — Demänová Ice Cave; Ok — Okno Cave; ST — Štefanová Cave; A — alpha spektrometry; M — mass spektrometry (ICP-MS).

Supplementary references

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