Origin and chronology of speleothems in a Lewisian gneiss cave on the western coast of Scotland

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Abstract: Speleothems were recently discovered in a 40m-long coastal cave carved into silicate metamorphic rocks on the island of Iona, the Inner Hebrides, Scotland. Worldwide, calcite speleothems are found commonly in caves formed in carbonate rocks and have been used extensively as a source of data to reconstruct details of past climatic conditions. Such reconstructions have, however, been rare in Scotland, because exposure of carbonate rocks that typically host speleothem-bearing caves is limited. Discovery of speleothems on Iona thus holds great significance, because it offers a climate proxy that, potentially, can be dated accurately. The Iona cave, which lies along the intersection of two faults within gneiss of the Lewisian Complex, hosts stalactites on its ceiling, and abundant wall flowstone from its mid-section to its far end. Whereas the origin of calcium for speleothem formation remains unclear, it is probably sourced from the dissolution of shell sands, known locally as machair. It is suggested here that this machair was deposited above the cave, by wind action, under paraglacial conditions, and then dissolved in meteoric water passing through acidic peat overlying the gneiss. This hypothesis is supported by the fact that, whereas no shell fragments are present in a peat core sampled above the cave, its section contains sand-rich layers and a calcium carbonate content (2–12% by weight) that does not match the mineralogy of the exposed bedrock. U–Th-series ages obtained from an Iona speleothem sample are affected by the presence of unconstrained, non-authigenic thorium and therefore carry significant uncertainties. It began to grow at 4.78 ± 2.25 ka BP (before present; present = 1950 CE), a date that might represent when the cave's roof was raised above sea level in response to postglacial isostatic rebound. Study findings demonstrate that calcium carbonate speleothems can form in silicate rock settings and highlight their potential value as a new archive of Holocene environmental change for western Scotland, and other areas.

Keywords: Speleothems; Iona; Inner Hebrides; Lewisian gneiss; silicate-hosted cave; machair sands; Holocene; U–Th dating; postglacial isostatic rebound.

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Introduction

Because of their unique characteristics, carbonate speleothems — secondary cave deposits — represent valuable archives of palaeoclimate reconstruction data. They have a wide geographical distribution (allowing for global climate reconstructions), are suitable for uranium–thorium (U–Th) dating, contain several climate proxies (e.g., stable carbon and oxygen isotopes, trace elements), have chronological continuity (potentially covering millennia), and have a low sampling cost (Cheng *et al*., 2019). Scottish speleothems have a potentially great importance because climate reconstructions spanning the Holocene are limited. As well as being used to study climate, speleothems in coastal caves can help constrain the timings of relative sea-level changes, because they could not have grown when they were submerged in seawater (Richards *et al*., 1994). This characteristic has a potentially great value in Scotland, considering the significant isostatic rebound and relative sea-level change that occurred following the last glaciation.

Speleothems are most commonly found within caves developed in carbonate rocks, and less commonly in those formed in igneous rocks (e.g., Kulkarni *et al*., 2022; Woo *et al*., 2015). For caves in carbonate rocks, the speleothem depositional mechanism is well known. Meteoric water encounters CO_2-rich soil and some CO_2 dissolves in the water. When limestone interacts with this carbonic-acid-rich fluid, it is slowly dissolved, liberating Ca^{2+} ions. When the water containing Ca^{2+} and HCO_3^- reaches a cave, the lower partial pressure of CO_2 in its atmosphere causes CO_2 to be released. The degassing results in deposition of calcium carbonate from the water, forming solid mineral deposits, most commonly calcite and, more rarely, aragonite. Calcite speleothems can also occur within caves in some igneous rock types; in basaltic lithologies, for example, calcium can be released by breakdown of unstable magmatic products, such as Ca-rich plagioclase (White, 2010).
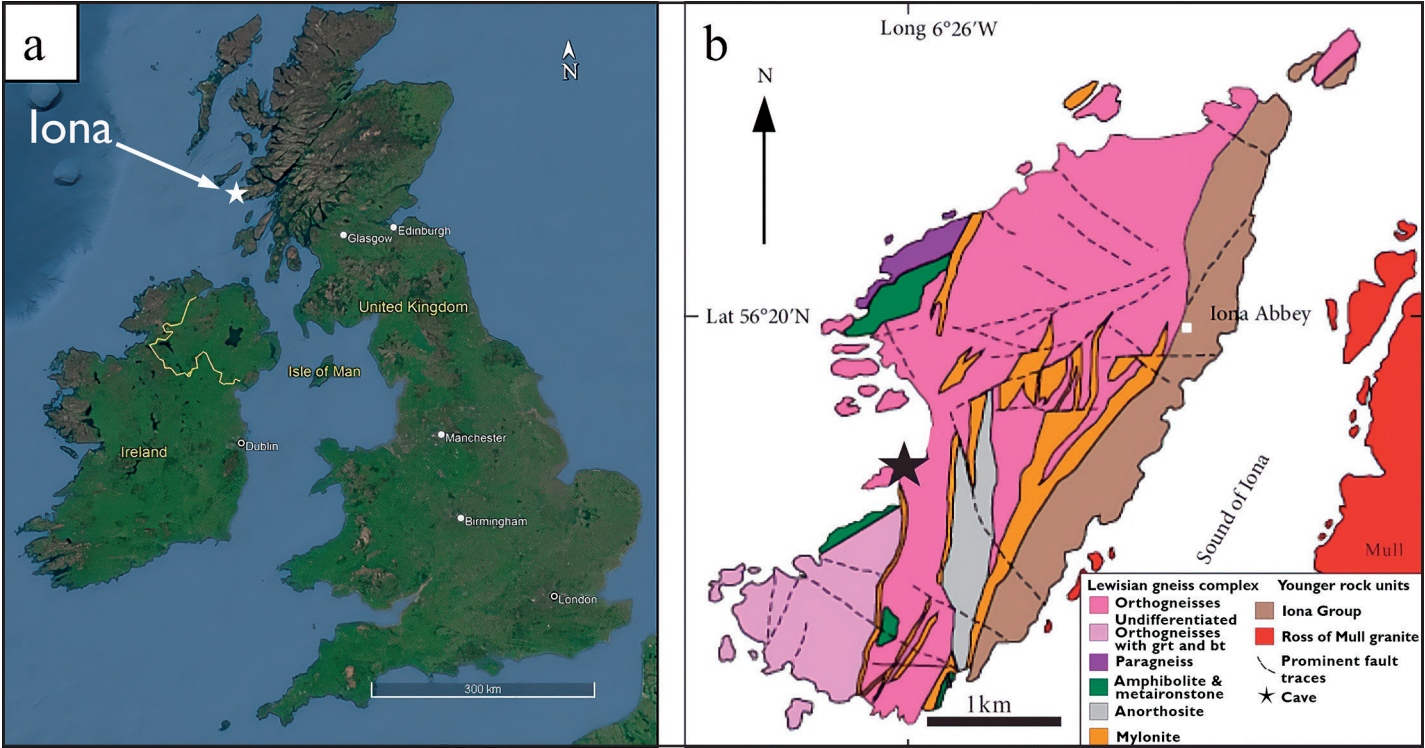
Figure 1: *(a)* Location of the Isle of Iona, NW Scotland (Google Maps, 2023).
(b) Simplified geological map of Iona (after Dempster et al., 2021). The cave's location, on the island's west coast, is marked by a black star.

Speleothems discussed in this Report were discovered recently in a cave within metamorphic silicate rock on Iona, a small island off the west coast of Scotland. Other speleothems described and studied from several caves elsewhere in Scotland (e.g., *Uamh an Tartair* cave in Assynt; Baker *et al*., 2015), all formed within caves in carbonate lithologies, and few are in coastal locations. Given the unusual nature of the Iona cave, this Report describes its geomorphological and geological setting and discusses the possible origin of its speleothems. More generally, this study aims to investigate the formation of speleothems in coastal caves within metamorphic rocks, assess the origin of calcium in such non-carbonate settings, and evaluate the potential for obtaining precise geochronology using U–Th dating of detritus-rich speleothems.

Scientific background and methods
The Iona cave
Iona is an island in the Inner Hebrides and lies 1km west of the larger island of Mull, off the western coast of Scotland (Fig.1a). It holds cultural and historical importance and is often referred to as 'the birthplace of Scottish Christianity'.

St Columba arrived there in 563 AD and established Iona Abbey, making the island a prominent centre of learning during the early medieval period (Jones *et al*., 2022).

The geology of Iona is dominated by ancient gneisses of the Lewisian Complex in the west, overlain unconformably by younger meta-sedimentary rocks of the Iona Group in the east (Fig.1b) (Dempster *et al*., 2021). Zircon Lu–Hf ages date the gneiss complex in Iona to ~2710 Ma (Daly *et al*., 2024).

The speleothem-bearing cave, *Uamh a' Chroisean* ("Cave of the Little Cross"; NM 26410 23100), is located on the island's west coast and is hosted within orthogneiss (Figs 2a and 2b). Development of the cave, which is 41.2m long, 0.3–7m wide, and 1.5–8m high, was guided by two prominent fault planes, which now form vertical side walls that are coated in flowstone (Fig.2c). The floor of the cave is frequently inundated by seawater during high tides, making access difficult (Fig.2a). A sample of flowstone (I-1) was collected from the cave ceiling (Fig.2c), 7.06m above present-day Mean Sea Level.
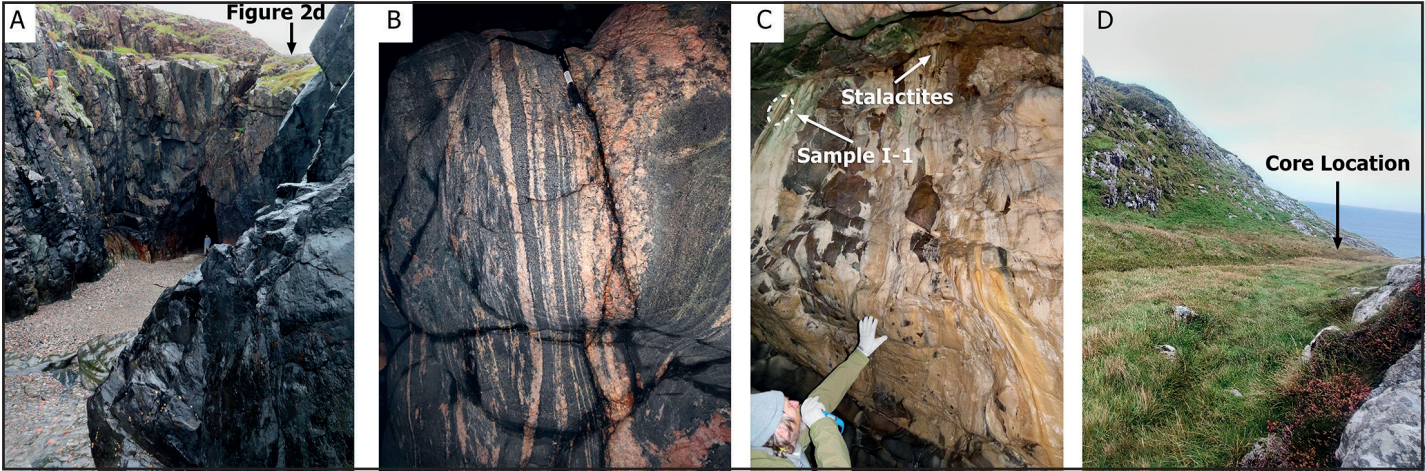

Figure 2: *The cave and its surrounding environment.*
(a) General view of the cave, with peat bog (the green vegetation) above the cave. The lack of any vegetation on the shore in front of the cave indicates that the cave floor is in the intertidal zone (photograph by Dr John Faithfull); *(b)* Foliated Lewisian gneiss, the host rock of the cave (photograph by Kang Xie); *(c)* Inside the cave. Flowstone covers the Lewisian gneiss walls, just below the fault fracture visible on the roof of the cave where two stalactites have grown (photograph by Dr John Faithfull); *(d)* Peat bog above the cave and the location of the peat core (photograph by Kang Xie).
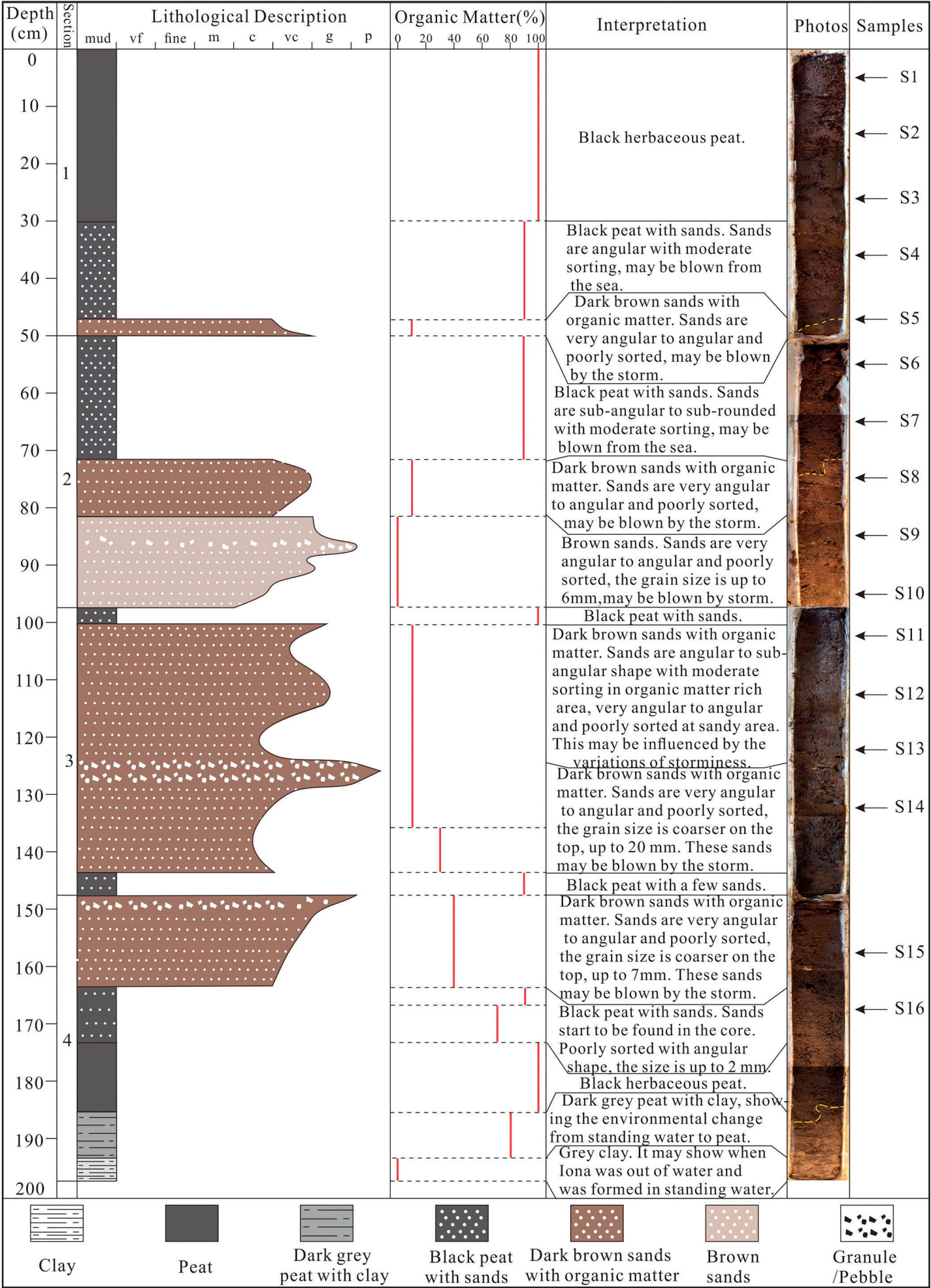
Figure 3: *Graphic log of the Iona core, the location of which is shown on Figure 2d.*
The horizons labelled S1 to S16 are the locations of samples collected for calcium analysis.
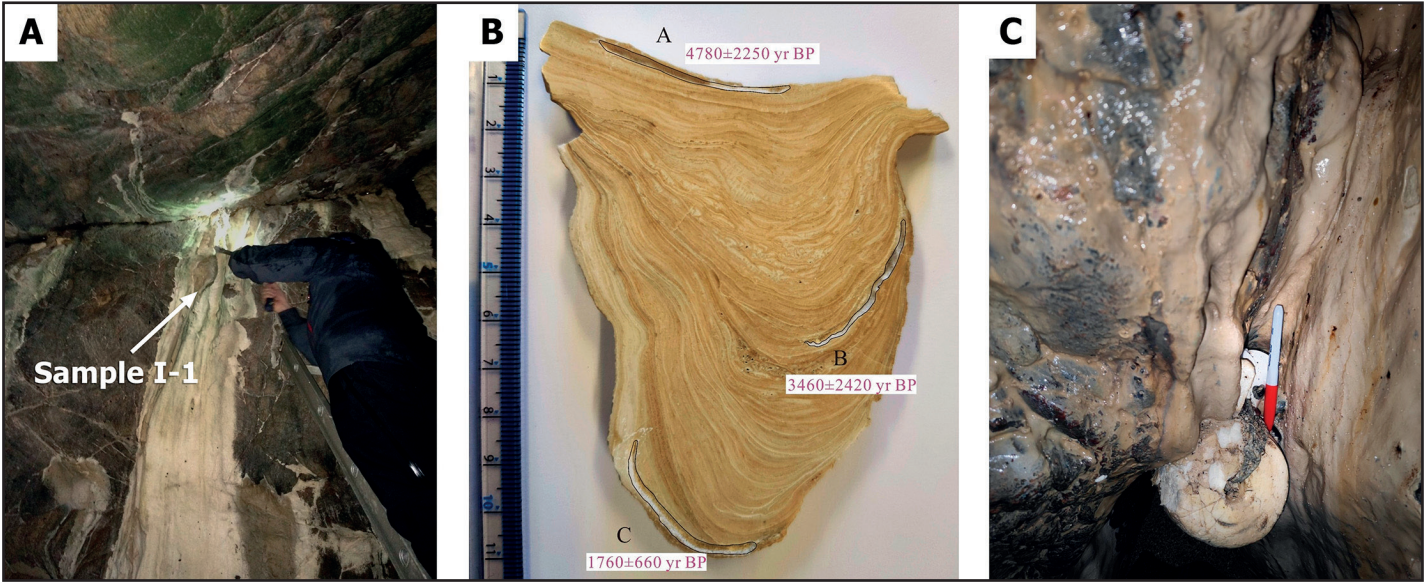
Figure 4: Speleothems formed in the cave.
(a) Location of the 11cm sample I-1 (photograph by Dr John Faithfull);
(b) Sample I-1 slab annotated with U–Th dating results (photograph by Kang Xie). Sample Iona-A is the oldest and Iona-C the youngest;
(c) Plastic float at the end the cave, showing that some flowstone is still forming ('Sharpie' (c. 135mm) as scale bar; photograph by Kang Xie).

Nearshore and the low-lying parts of the west and centre of Iona are covered by machair, a coastal landform/vegetation system that is present only on the northwestern coastlines of Ireland and Scotland and is dominated by wind-blown calcareous shell-sand derived from offshore and nearshore biogenic production (Ritchie, 1976; Hansom and Angus, 2005; Angus, 2006). Ordered from the sea inland, areas of machair generally incorporate a dune cordon, and species-rich grassland (managed by traditional low-intensity agriculture), wetlands, lochs and black lands (peat on rock) (Ballantyne and Gordon, 2021). The *Uamh a'Chroisean* cave is about 130m from the closest machair dunes. A 197.5cm-long core extracted from a peat bog directly above the cave (Fig.2d), using a Russian corer, extended from 28cm below the bog surface to the cave roof (Fig.3).

Speleothem sampling and U–Th dating

Sample I-1 is a stalagmitic flowstone (Figs 2c and 4a). A 1cm-thick slab was cut from the centre of this flowstone, with a length of 11cm and width of 7cm (Fig.4b). Three 80~85mg powder samples for U–Th dating were extracted at the petrology laboratory (Fig.4b), University of Glasgow (UofG), using a dental drill. The powders were used for U–Th dating using a Thermo Neptune Plus multi-collector inductively coupled mass spectrometer (MC-ICP-MS) at the NERC Isotope Geosciences Laboratory (NIGL), British Geological Survey, Keyworth, Nottinghamshire. Ages were calculated assuming an initial $^{230}Th/^{232}Th$ activity ratio of $4.4 \times 10^{-6} \pm 2.2 \times 10^{-6}$ (Edwards *et al.*, 2003).

Water sampling and calcium analysis

To assess the source of calcium, two water samples were collected from a stream above the cave, and two dripwater samples collected from inside the cave. Calcium concentrations were measured at the UofG using an Agilent 5900 inductively coupled plasma–optical emission spectrometer (ICP-OES). In addition, 16 peat samples from the core were analysed to determine their calcium carbonate content (Fig.3). For these analyses, approximately 1g of dried material was placed in a 15ml Falcon tube, treated with excess 1 M hydrochloric acid, and left to react overnight in a fume hood. Tubes were centrifuged, the supernatant liquid discarded, and the residue washed with Milli-Q water. Centrifugation and rinsing were repeated until the pH of the resultant supernatant liquid was neutral. Samples were then dried to constant mass, and the calcium carbonate content was calculated from the difference in dry weight before and after acid treatment.

Results

Sample description

Speleothems are widespread from the middle to the end of the cave. Most are flowstones, but two stalactites occur on the cave's ceiling approximately 24m from its entrance (Fig.2c). This 11cm long flowstone exhibits more than one thousand clear laminae (25–150µm thick), each separated by thin and dark layers (Fig.4b). Laminae colour varies; some are brown whereas others are white, suggesting periodic changes in water chemistry, potentially linked to seasonal or hydrological variability (Fig.4b). Flowstone is still actively forming in the cave, as demonstrated by ongoing deposition around a solitary plastic float (from a fishing boat), found at the rear of the cave, (Fig.4c).

Analytical results

The U–Th-series ages for the three analysed samples of I-1 (Fig.4b) yielded high uncertainties (average 51.5%, 2σ) (Table 1). These elevated uncertainties are attributed to a high input of detrital material containing non-radiogenic Th.

Sample Name	Sample weight (mg)	Date uncorrected (ka BP)	Absolute Uncertainty (±2σ)	Date corrected (ka BP)	Absolute Uncertainty (±2σ)
Iona-A	80.25	7.99	0.06	**4.78**	**2.25**
Iona-B	81.05	6.89	0.04	**3.46**	**2.42**
Iona-C	84.06	2.77	0.02	**1.76**	**0.66**

Table 1: U–Th ages for the I-1 speleothem.
The locations of the three dated samples within the speleothem slab are shown in Figure 4b.
The 'corrected' dates were calculated assuming an initial $^{230}Th/^{232}Th$ activity ratio of $4.4 \times 10^{-6} \pm 2.2 \times 10^{-6}$ (Edwards et al., 2003).

Stream water samples collected from above the cave contained 14 and 9 ppm Ca, whereas the two dripwater samples from within the cave contained 41 and 45 ppm Ca. These compositional differences show that the water acquires Ca between the peat bog surface and interior of the cave. Table 2 shows sample details and analytical results of 16 samples from the peat core collected above the cave. Values of $CaCO_3$ are variable, with the highest (12%) from clay and the lowest (2%) from clayey sand.

Discussion

Formation of the cave

The cave occupies the intertidal zone of a small bay on the Atlantic coast, with the roof situated ~10m above Mean Sea Level. It developed along two ancient fault planes that defines its vertical side walls. The gneiss within this fault-bounded zone was fractured and thus more vulnerable to marine erosion, which progressively removed the weakened rock and excavated the cavity. During and around high-tide periods, the cave floor is inundated by seawater, indicating that interaction with the coastal environment is ongoing. As such, the cave can be classified as a fissure cave, primarily formed by exploitation of fault-guided fracturing and later modified by the effects of marine erosion related to tidal inundation and, possibly, storm surges.

Despite the cave's floor being flooded at high tide, speleothems have grown on its walls and ceiling where they remain apparently unaffected by the effects of direct seawater contact. The cave's semi-enclosed geometry and limited ventilation might help maintain high humidity and stable temperatures, both of which favour calcite precipitation.

Source of calcium for the speleothem

Growth of the speleothem deposits in the cave is continuing (Fig.4c), indicating that their calcium source remains present. Calcium concentrations in cave dripwaters are substantially higher than those in the stream waters above the cave, suggesting that the source lies between the peat-bog's surface and the cave ceiling — namely, the overlying peat bog and/ or the host rock. The cave is developed in orthogneiss, which consists predominantly of K-feldspar, quartz, partly sericitized plagioclase, hornblende (altering to chlorite), and biotite (Daly *et al*., 2024). Thus, the orthogneiss does not include a significant level of soluble calcium-rich minerals, making it an unlikely contributor of calcium to the speleothems. Even though no shell fragments were observed in the peat core (Fig.3), the most plausible source of calcium is dissolution of shell material from wind-blown machair sands by acidic porewaters in the peat.

Supporting evidence includes sand-rich layers in the peat core (Fig.3), interpreted as residues of dissolved machair deposits, and calcium carbonate contents of 2–12 wt% in the peat samples, which are inconsistent with the host rock mineralogy and strongly suggest a machair-derived carbonate source (Table 2).

Factors influencing dating results with high uncertainty

Accurate and precise U–Th dating assumes an initial zero concentration of ^{230}Th (Wendt *et al*., 2021). The high uncertainties in the dating results presented here are due largely to unconstrained input of non-authigenic ^{230}Th. In the Iona speleothem, this Th probably comes both from the Lewisian gneiss host rock and from the peat bog. Lewisian gneiss provides a potential source of detrital material enriched in both non-radiogenic ^{232}Th and inherited radiogenic ^{230}Th. The standard detrital correction for U–Th dating assumes an initial ^{230}Th/^{232}Th activity ratio of $4.4 \times 10^{-6} \pm 2.2 \times 10^{-6}$ (Edwards *et al*., 2003), which might underestimate the true detrital ^{230}Th contribution in samples influenced by old U-bearing metamorphic rocks, resulting in derivation of inaccurate U–Th ages, which appear misleadingly old. Whereas peat bogs are not major thorium reservoirs, clays within them can contain minor amounts of ^{232}Th and non-authigenic ^{230}Th. Such detrital components could be transported into the cave and incorporated within growing speleothems. Thus, the dating of speleothems in caves within Lewisian gneiss must be approached with caution and with awareness of the potential for detrital contamination.

Given the uncertainties in the U–Th age determinations, only a rough estimate of the Holocene isostatic rebound rates can be provided. Based upon the oldest age recorded for the speleothem, considering the associated analytical uncertainty (4.78±2.25 ka BP), and knowledge of its height above present-day Mean Sea Level (7.06m), averaged rates of isostatic rebound range from 1 to 3mm/year. This range of values is similar to those calculated from isostatic curves for the nearby areas of Arisaig (western Scotland, mainland) and the island of Islay for the Late Holocene (Lin *et al*., 2021; Shennan *et al*., 2006, 2018; Smith *et al*., 2019).

Sample number	Sediment type	Depth in the core (cm)	Sample weight (g) (before dissolving)	Sample weight (g) (after dissolving)	Calcium carbonate (g)	CaCO$_3$ content (weight by percent)
S1	Peat	5	0.82	0.73	0.09	12%
S2	Peat	15	0.86	0.77	0.08	11%
S3	Peat	26	0.98	0.89	0.09	10%
S4	Peat with sands	36	1.59	1.50	0.09	6%
S5	Sands with organic matter	47	4.41	4.30	0.11	3%
S6	Peat with sands	55	0.85	0.79	0.07	8%
S7	Peat with sands	65	1.24	1.14	0.09	8%
S8	Sands with organic matter	75	4.17	4.07	0.10	2%
S9	Sands	85	3.11	3.04	0.07	2%
S10	Sands	95	2.80	2.73	0.07	3%
S11	Sands with organic matter	102.5	3.79	3.72	0.07	2%
S12	Sands with organic matter	112.5	2.69	2.62	0.06	2%
S13	Sands with organic matter	122.5	3.15	3.02	0.12	4%
S14	Sands with organic matter	132.5	0.95	0.85	0.10	12%
S15	Sands with organic matter	157.5	2.38	2.29	0.09	4%
S16	Peat with sands	167.5	0.92	0.83	0.10	12%

***Table 2**: Nature and calcium carbonate content of peat samples obtained using a Russian corer; see also Figure 3.*

Conclusions and further work

Unexpectedly, a recently discovered cave (*Uamh a' Chroisean*) in meta-silicate rock on the western coast of the island of Iona, western Scotland, is decorated with speleothems. Development of this cave was mainly by the effects of marine erosion, guided along fault planes. The most likely calcium source for speleothem formation is former high-level machair sands that have undergone dissolution beneath the overlying peat bog. High U–Th age uncertainties make the dating results derived from the Iona speleothem samples inconclusive. Large errors are evident, primarily due to detrital input from the Lewisian gneiss host rock. Nevertheless, despite the uncertainties, the derived rates of isostatic rebound are similar to those estimated in nearby areas.

These findings demonstrate that, under appropriate geochemical conditions, speleothems can and do form in caves within non-carbonate metamorphic rocks. Such sites might serve as valuable archives that can provide information to inform palaeoclimate reconstruction and relative sea-level studies, particularly in regions such as Scotland, where traditional carbonate-hosted speleothems are rare. This work also highlights the importance of adapting detrital correction strategies when applying U–Th dating in such environments. Work continues to obtain carbon and oxygen stable isotope analyses of the *Uamh a' Chroisean* samples, to assess the suitability of these speleothems as proxy data sources to aid palaeoclimate reconstruction.

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