Attraction in the Dark: The Magnetism of Speleothems

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No matter how quiet and pristine a cave setting may appear, all speleothems contain assemblages of magnetic minerals. These iron oxide minerals are derived largely from overlying soils, though minor fractions may come from the residuum of dissolved bedrock, reworked sediment carried by episodic floods, geomicrobiological activity, and even windblown dust. Regardless of their origin, these minerals become aligned with Earth's ambient magnetic field before they are fixed within a speleothem's growing carbonate matrix. Here, we describe how the magnetism of stalagmites and flowstone can be used to chronicle high-resolution geomagnetic behavior and environmental change.

KEYWORDS: speleothem magnetism; paleomagnetism; environmental magnetism

INTRODUCTION

Traces of magnetic minerals occur in all natural materials and can be used to gain insights into an enormous diversity of research questions. Studies of magnetic minerals typically fall into one of two broad categories: paleomagnetism (see Box 1) and environmental magnetism (see Box 2). Speleothems have recently made major contributions to both fields.

Speleothems have recently become attractive targets for both paleomagnetic and environmental magnetic studies. Traditionally, geomagnetism researchers have relied on materials with strong magnetic moments (>10^{-9} Am^2) that can be measured accurately using standard magnetic instrumentation: volcanic rocks, lake and marine sediments, soils, and paleosols. However, recent improvements in the sensitivity and spatial resolution of magnetometers have enabled researchers to explore novel, albeit more weakly magnetized, materials such as speleothems, which offer numerous advantages for paleomagnetic studies. For example, certain forms of stalagmites display annual growth layers with thicknesses between one and several hundred micrometers, thereby providing more continuous temporal records than sporadically erupted lava flows. When hiatuses are present in speleothems, they can be identified and avoided using standard microscopic techniques. Studies of these annual growth layers suggest that the geomagnetic alignment of magnetic minerals becomes "locked in" at subannual timescales and experiences little, if any, postdepositional modification (Lascu and Feinberg 2011). This is in contrast to lake and marine sediments, whose magnetizations may be complicated following deposition through bioturbation, compaction, and dissolution/reprecipitation of Fe-bearing minerals in response to varying redox conditions. Perhaps most importantly, stalagmites can be accurately and precisely dated using U–Th methods, which allows their magnetizations and rock magnetic properties to be fixed firmly in absolute time and easily compared to other important datasets derived from speleothems, including measurements of oxygen and carbon stable isotopes and trace elements.

But how do magnetic minerals become incorporated into speleothems? Figure 1 shows a conceptual model of the many processes that deliver magnetic minerals to speleothems. Magnetic minerals formed in the topsoil, such as magnetite, maghemite (γ-Fe_2O_3), and goethite, are transported from the surface to the cave environment by water percolating from the soil via cracks and bedding planes to the point sources for drip water from which speleothems form. After being deposited onto the surfaces of flowstone, stalagmites, and stalactites, grains are permanently fixed within continually growing and seasonally driven carbonate laminae. The magnetic minerals that are incorporated into speleothems via drip points are naturally sorted during transport and are on the order of 10 to 1,000 nanometers (an ideal grain size for paleomagnetic applications because they retain stable magnetizations over geologic timescales). Larger grains (10s to 100s of microns) can be carried into the subsurface by sinking streams and may be deposited along with fine-grained silicates as allochthonous layers on speleothem surfaces during the quiescent stages of water retreat after a flooding episode. Strauss et al. (2013) successfully imaged magnetic grains extracted from stalagmites using scanning and transmission electron microscopy. These images showed the differences in composition, grainsize, and microstructure that differentiate magnetic minerals arriving via drip water from those arriving from more energetic hydrologic processes (e.g., floods and mudspatter) as well as from those that form authigenically, such as needles of goethite formed in situ on silicate minerals (Fig. 2).

The first paleomagnetic study of speleothems was by Latham et al. (1979), who could only measure large ~10 cm^3 speleothem specimens due to the lower sensitivity of magnetometers available at that time. Despite this, the study was noteworthy for demonstrating that actively growing speleothems accurately captured ambient magnetic field directions within a cave. Using flowstone and stalagmite samples from assorted caves in Canada and the United Kingdom, Latham et al. (1979) showed that older speleothems could retain stable reversed magnetizations, thereby opening the potential for using them to study reversals, excursions, and magnetostratigraphy.

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Box 1 PALEOMAGNETISM

Paleomagnetism uses a material's magnetic recording to reveal the behavior of Earth's ancient magnetic field, or geodynamo. The field is generated by convection of liquid iron–nickel in Earth's outer core, and although a variety of nondipolar fields are generated within the core, these become attenuated within the mantle, and the field is largely expressed as a dipole at the planet's surface. The geomagnetic pole is generally aligned with the planet's rotation axis and is viewed as a geocentric axial dipole when studied over timescales greater than 10,000 years. On shorter timescales, the geomagnetic pole follows a random walk around the geographic pole in a process called "paleosecular variation". Occasionally, the geomagnetic pole will migrate beyond the limits of normal paleosecular variation and express a geomagnetic "excursion", which is a short-term (≤5,000 years) directional deviation accompanied by a decrease in the overall field strength. Excursions end as the field reestablishes itself to its earlier orientation and field strength. Less frequently, excursions develop into full-scale geomagnetic reversals, a process thought to occur over longer timescales of ≤10,000 years (Merrill and McFadden 1999). When it is not punctuated by excursions or reversals, Earth's magnetic field is oriented in a configuration similar to that of the modern day ("normal polarity") or is oriented in an antiparallel configuration ("reversed polarity"). One of the most important scientific contributions by the paleomagnetic community has been the construction of a geomagnetic polarity timescale, which chronicles nearly all of the geomagnetic reversals from the modern day back to the early Mesozoic and which forms the foundation for magnetostratigraphic dating of rocks and sediments. Similarly, because Earth's magnetic field is expressed as a dipole at the planet's surface, its inclination varies predictably as a function of latitude; hence, these recordings can be used to determine the positions of drifting tectonic plates through time.

Numerous studies have built on the foundations of Latham's work, many of which are reviewed in Lascu and Feinberg (2011). Many of the recent studies on speleothem magnetism use sensitive modern rock magnetometers that employ cryogenic sensors lovingly referred to as "SQUIDs" (superconducting quantum interference devices), which can measure 3-D magnetic moments for ~1 cm^3 geologic samples as low as 10^{-12} Am^2. In these studies, long strips collected from the centers of stalagmites are subdivided into thin (0.5 cm), carefully oriented wafers cut parallel to the stalagmite's horizontal growth surface. Great care must be taken to prevent magnetic contamination of the speleothem during the cutting process. For this reason, nonmagnetic saw blades or wire saws are used, and each sample is carefully sanded and cleaned prior to measurement. Paleomagnetic measurements from these small samples represent an integrated time average, usually between 10 and 1,000 years per sample, depending on their annual rate of growth. Certain specialized magnetometers bring small SQUID sensors very close to a sample surface where they can measure moments as low as 10^{-15} Am^2 over length scales of ~200 μm. These SQUIDs can be rastered across a surface to create a 2-D image of submillimeter-scale magnetizations within a sample, as has recently been done for speleothems (Feinberg et al. 2020; Fukuyo et al. 2021). Scanning SQUID microscope images are incredibly useful for visualizing where magnetic minerals are concentrated within a speleothem. The magnetic information in these images is analogous to aeromagnetic surveys over economic mineral deposits, albeit on a micrometer scale. As is the case with aeromagnetic anomalies, these data must be carefully inverted to determine how magnetic field direction and intensity change along the length of the stalagmite. Efforts are underway to develop reliable inversion protocols that would allow researchers to gather time-series measurements of geomagnetic field changes with near annual resolution.

This steady improvement in our ability to measure speleothem magnetizations has led to a renaissance in their use over the last ten years such that there are more studies to review than is possible in this short contribution. Instead, we highlight two recent case studies of paleomagnetic and environmental magnetic applications to speleothems that demonstrate the power of this approach.

PALEOMAGNETISM OF SPELEOTHEMS

An exciting demonstration of the use of speleothems for understanding short-term geomagnetic field behavior comes from Chou et al. (2018), who examined a 1 m long candle-like stalagmite from Sanxing Cave (southwestern China). The age of the stalagmite was determined using U–Th methods. Paleomagnetic samples were collected at 0.5 cm intervals from the central portion of the speleothem to avoid deflections of the recorded paleomagnetic field directions that are known to occur along the steeply dipping flanks of stalagmites (Ponte et al. 2017). Thus, each specimen represents a multidecadal average of the direction of the Earth's magnetic field. This particular stalagmite grew between 107 ky BP (before present) and 91 ky BP during a geomagnetic excursion called the "post-Blake event." The stalagmite's paleomagnetic information provides a high-resolution view of how the Earth's geodynamo behaves during excursions (Fig. 3).

Perhaps the most eyebrow-raising result of this study is the abrupt, transitory reversal that occurs over only 144 ± 58 years (2σ) (Fig. 3). This rapid field change at 98.3 ka represents one of the fastest recorded variations in the direction of the Earth's magnetic field and would be equivalent to a minimum rate of change of ~0.6° per year. For comparison, the Earth's modern field rarely moves more than ~0.1° per year (Davies and Constable 2020). Subsequent work has shown that during periods of low geomagnetic field strength (e.g., during excursions and reversals), Earth's field direction can swing wildly and approached rates of change as high as 10° per year during the Laschamp Excursion

Box 2 ENVIRONMENTAL MAGNETISM

In environmental magnetic studies, a sample's magnetic recording is of little importance. Instead, it is the physical properties of the magnetic minerals themselves that are used to glean information about the environmental conditions and processes that were active during a rock or sediment's formation. Factors such as the concentration, composition, grain size distribution, and physical distribution of magnetic minerals within natural materials can be incredibly informative. Common magnetic minerals, such as magnetite (Fe_3O_4), hematite (α-Fe_2O_3), and goethite ($FeO{\cdot}OH$), occur across a range of terrestrial and marine settings and often form as a reflection of ambient moisture conditions, temperature, and chemical environment. Slightly rarer, but equally important, Fe-bearing minerals such as greigite (Fe_3S_4), pyrrhotite ($Fe_{1-x}S$, where $x = 0$ to 0.2), siderite ($FeCO_3$), and vivianite [$Fe_3(PO_4)_2{\cdot}8H_2O$] also provide important information about redox conditions and element cycling. There is a terrific diversity of environmental applications, and this approach is one of the fastest growing niches within the international magnetic community. Examples include the estimation of paleoprecipitation rates using magnetic mineral assemblages preserved within paleosols (Maxbauer et al. 2016), unraveling iron speciation in sediments (Slotznick et al. 2020), quantifying the proliferation of biologically produced magnetite in ocean sediments during abrupt global warming events (Wagner et al. 2021), and even documenting the varying influence of sediment sources in North Atlantic marine sediments through time (Hatfield et al. 2017). Readers interested in the vibrant fields of paleomagnetism and environmental magnetism are encouraged to explore the *Elements* issue "Mineral Magnetism" (2009, v5n4) for additional information, as well as Tauxe (2003) and Liu et al. (2012).

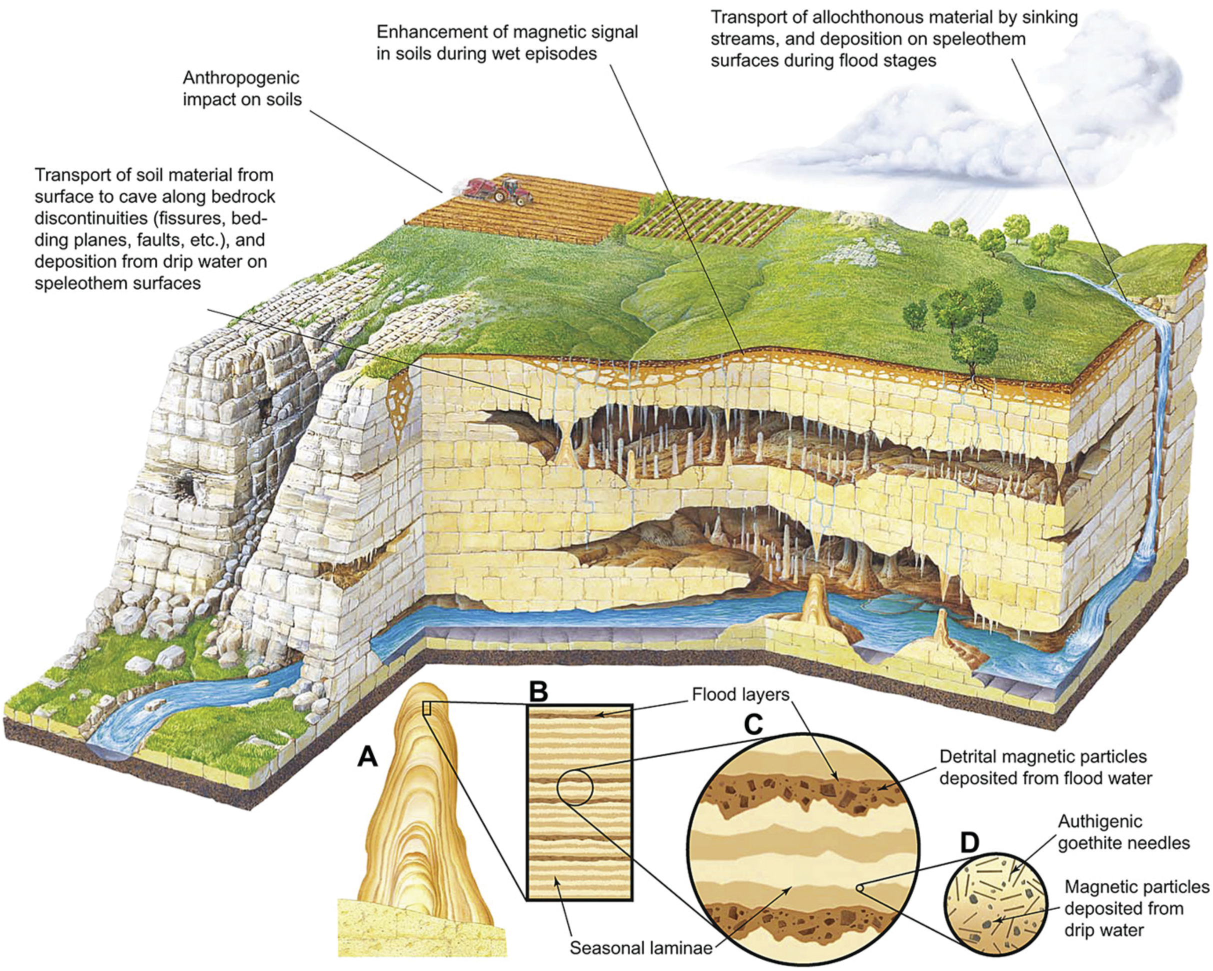

Figure 1 Schematic model of the processes delivering magnetic minerals to speleothems. Magnetic material in the topsoil is transported from the surface to the cave environment by water percolating from the soil via cracks and bedding planes to the point sources for drip water from which speleothems form. Underground streams transport larger detrital magnetic particles, which are deposited on speleothem surfaces during the quiescent stages of water retreat after a flooding episode. (**Inset**) The progressively finer-scale features within a periodically flooded stalagmite. (**A**) Stalagmite at the centimeter scale. (**B**) Growth layers within a stalagmite at the millimeter scale. (**C**) Detail of individual growth layers at the submillimeter scale. (**D**) Detail at the boundary between two seasonal laminae at the submicron scale. Seasonal laminae are on the order of several tens of microns to a few hundred microns, whereas fine-grained magnetic particles transported by drip water or precipitated in situ are on the order of a few nanometers to a couple of hundred nanometers. Adapted from Lascu and Feinberg (2011).

at 39 ka (Davies and Constable 2020). Such geomagnetic instability would have a dramatic effect on modern day satellite communications. Speleothems represent one of the only high-resolution archives capable of preserving this behavior.

Another important outcome from the Sanxing Cave study is that the magnetic field appears to show centennial- to millennial-scale oscillations during the post-Blake event (see 1–4 and a–f in Fig. 3). Such directional oscillations were hinted at in a previous study of rapidly erupted lava flows that recorded field variations during the last geomagnetic reversal (the Brunhes–Matuyama at ~780 ka) and were coincident with a period of low geomagnetic field strength (Mochizuki et al. 2011). However, the Sanxing Cave record provides a far more continuous and higher-resolution view of these oscillations, and their precise age and duration is much better constrained than is possible for dating of lava flows.

Interested readers are encouraged to view other paleomagnetic studies of speleothems related to excursions (Osete et al. 2012; Lascu et al. 2016), the South Atlantic Magnetic Anomaly (Jaqueto et al. 2016; Trindade et al. 2018), paleosecular variation (Zanella et al. 2018), and general recording processes (Zhu et al. 2012; Font et al. 2014). All of these studies demonstrate that stalagmites and flowstone are reliable recorders of the Earth's magnetic field and act as archives of geomagnetic behavior across a range of timescales. Future paleomagnetic work using speleothems may allow researchers to address important geophysical questions, such as "Do reversals start and end at different times at various locations around the globe?", "Are some reversals faster than others?", "Is oscillatory directional behavior a feature of all excursions and reversals?", and "Can speleothems be used to accurately estimate changes in the strength of the Earth's magnetic field?"

ENVIRONMENTAL MAGNETISM OF SPELEOTHEMS

One of the newest and most exciting advances in the study of speleothem magnetism is the development of the quantum diamond magnetometer (QDM), which is capable of imaging stray magnetic fields in geologic samples at 1 micrometer resolution. While not quite as sensitive as SQUID sensors, a QDM can collect higher spatial resolution images of magnetic fields emanating from a sample by using the response of visible light to crystalline imperfections in diamonds (Glenn et al. 2017). Thus, by collecting

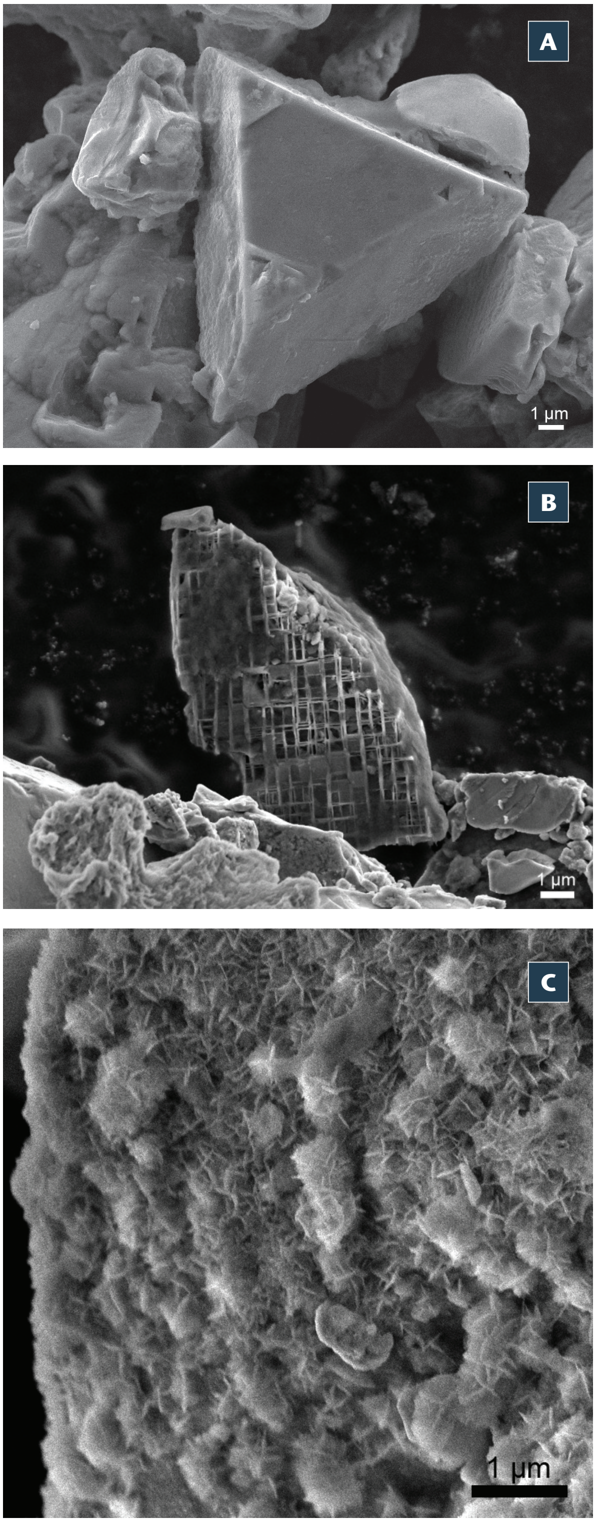

FIGURE 2 Three scanning electron microscope images of magnetic minerals isolated from stalagmites. (**A**) Titanomagnetite allochthonous grain from a flood deposit in Spring Valley Cavern (Minnesota, USA). (**B**) Exsolved magnetite and ulvöspinel intergrowth as an allochthonous grain from a flood deposit in Niagara Cave (Minnesota). (**C**) Needles of goethite crystals covering a clay platelet from Crevice Cave (Missouri, USA). Scale bar is 1 µm in each panel. IMAGES ADAPTED FROM STRAUSS ET AL. (2013).

images of geologic materials through a synthetic diamond in a carefully controlled environment, it is possible to image magnetic fields, which in turn highlights where magnetic minerals are located and how strongly magnetized they are.

Fu et al. (2021) provide a tantalizing demonstration of how these new magnetometers can be effectively applied to the study of stalagmite magnetism. FIGURE 4 shows the flanks of a stalagmite from Pau D'Alho Cave (Brazil). Images collected via a QDM from a condensed section of laminae are shown in the expanded portion of FIGURE 4, where dark blue regions correspond to areas showing strong magnetizations due to an enrichment in magnetic minerals. The ages of prominent dark layers were determined from the center of the stalagmite using U–Th methods by Novello et al. (2016) and traced to the stalagmite flank by Fu et al. (2021) to create a time series of magnetization within the speleothem.

The spatial resolution of the QDM allows researchers to quantitatively measure the magnetization of individual annual lamina within a stalagmite. In the study by Fu et al. (2021), we see that the distribution of magnetic minerals is not homogeneous throughout the stalagmite but instead occurs as discrete 10–100 µm layers. By contrast, most recent environmental magnetic studies on speleothems required bulk samples that were 5 mm thick in order to ensure a strong enough magnetization to be detectable using cryogenic three-axis rock magnetometers. Such a thickness for a single sample would encompass the entire measurement area of the data shown in FIGURE 4. The QDM's high spatial resolution allows geophysical researchers for the first time to collect magnetic data from speleothems at the same scale, or better, than the geochemical paleoenvironmental proxies of stable isotope measurements of carbon and oxygen.

This approach offers much more than simple pictures of a sample's magnetization. The relative ease of collecting QDM images allows researchers to image samples after exposing them to diagnostic laboratory fields that are intended to strongly magnetize or demagnetize the sample's magnetic mineral assemblage. In this way, the stability of a speleothem's magnetization can be explored, and inferences can be drawn about the composition and magnetic stability of a particular set of minerals at the scale of individual growth laminae. For example, FIGURE 4 shows a magnetic stability term called the "coercivity index" plotted as a profile across the speleothem. This index represents the percent of magnetization in an area with coercivities between 17 mT and 70 mT. A sample's coercivity is the magnetic field that is required to reverse half its magnetization and is inversely related to grain size. The fact that all of the layers in FIGURE 4 have nearly the same coercivity index suggests that the grain size and composition of magnetic minerals throughout the speleothem remain relatively constant and that peaks in the magnetic maps represent increases in magnetic mineral concentration.

In this case study of a tropical stalagmite, Fu et al. (2021) use the consistency of magnetic grain sizes throughout the speleothem and the lack of coarse detrital material to argue that these magnetically enriched layers are associated with periods of aridity, when speleothems grow more slowly or are halted altogether. While this may be true for tropical speleothems, at higher latitudes there are karst system processes active that may deliver magnetic minerals to speleothems during wetter periods. For example, a recent study using scanning SQUID microscopy imaged magnetic layers within a mid-latitude stalagmite from Minnesota (USA) that corresponded to historical extreme precipitation events that periodically flooded the cave system (Feinberg

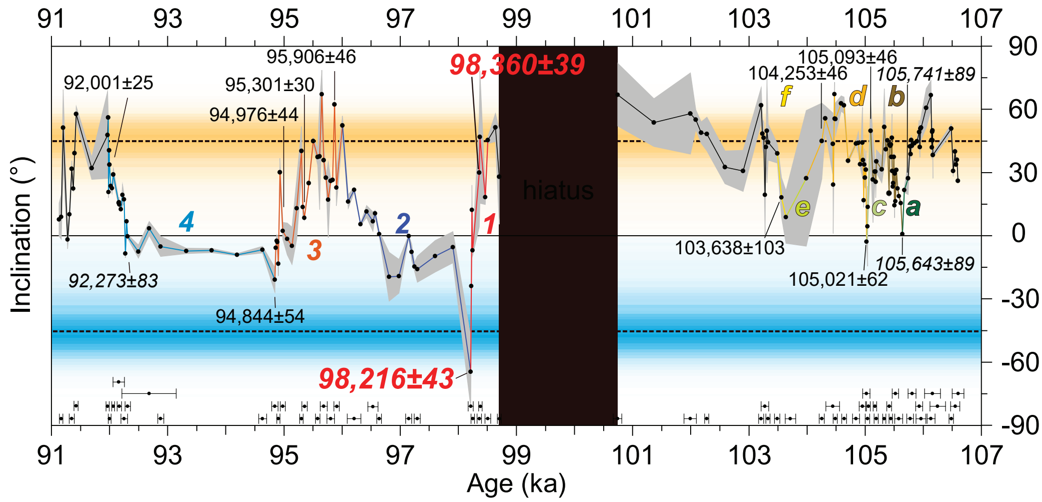

Figure 3 Paleomagnetic record obtained from a 1 m long stalagmite from Sanxing Cave (China). Timeseries of post-Blake excursion events showing paleomagnetic inclination (angular dip from horizontal) from the Sanxing stalagmite plotted with 95% confidence limits. Zones in yellow and blue indicate the highest probability inclinations expected at the cave's latitude for normal and reversed geomagnetic polarities, respectively, and take into account variability from paleosecular variation (see Box 1: Paleomagnetism). Select U–Th ages are labeled with 2σ errors and are shown at the bottom of the figure. The ages marked in bold italics show the interval over which the Earth's magnetic field transitioned rapidly from a normal to reversed polarity state. Colored numbers 1 to 4 (left side of plot) and letters 'a' to 'f' (right side of plot) show centennial- to millennial-scale oscillations during the post-Blake event. Figure adapted from Chou et al. (2018).

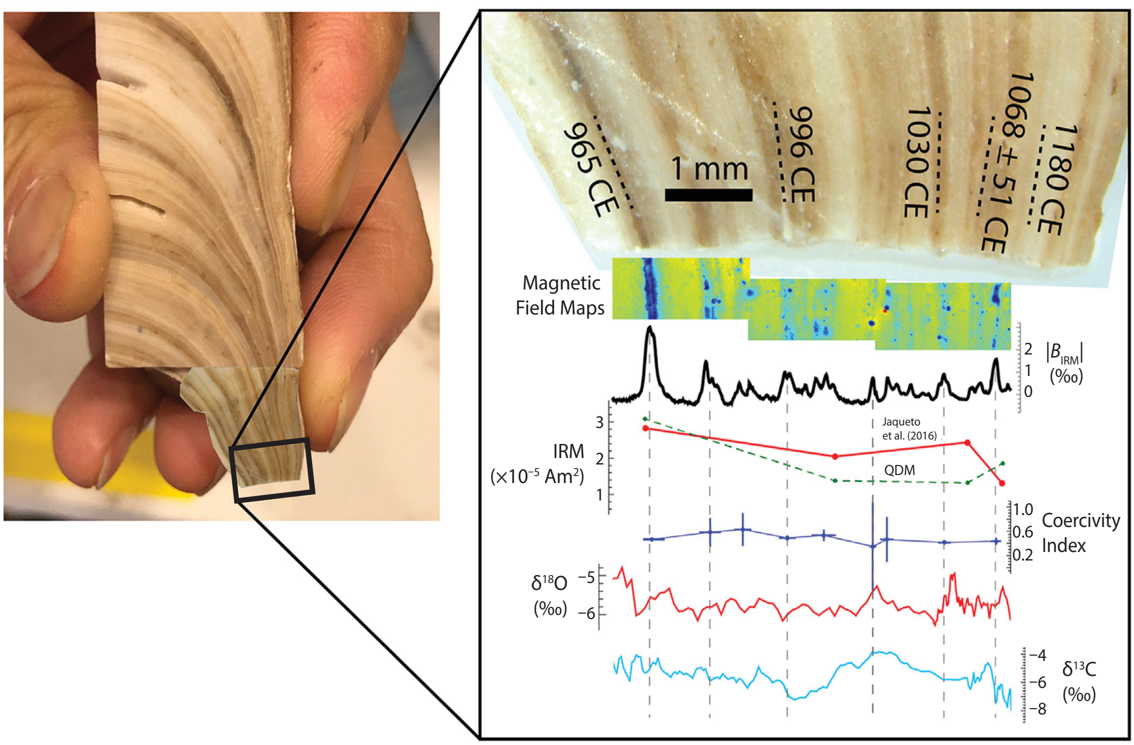

Figure 4 Images of stalagmite ALHO6 from Pau D'Alho Cave (Brazil). (**left**) Appearance of a hand specimen slice. (**right**) Expanded view showing age dates, magnetic field time series, and $\delta^{18}O$ and $\delta^{13}C$ data. The ages are based on U–Th dating of laminae traced or are interpolated from the central portion of the stalagmite. The magnetic field maps were created using a quantum diamond magnetometer (QDM) that measures the magnetic field (B) created by a laboratory-induced isothermal remanent magnetization (IRM); dark blue regions correspond to areas showing strong magnetizations due to enrichment in magnetic minerals. The B_{IRM} curve is a time series of mean magnetic field intensity created by vertically integrating the maps above. The IRM curves are the bulk samples of Jaqueto et al. (2016) and the QDM magnetic field values, respectively. The coercivity index is a proxy for the mean magnetic grain size throughout the speleothem. Oxygen and carbon isotopic data are from Novello et al. (2016). This series of plots suggests that magnetic mineral compositions remained constant throughout the stalagmite's growth and that zones of high magnetization in this tropical speleothem are associated with periods of aridity. Adapted from Fu et al. (2021).

et al. 2020). Flood layers in this 500 year record represent a valuable source of data for exploring how land use changes can alter an area's hydrogeologic response to extreme precipitation and an increase in flooding frequency. Similar flood layers do not occur in all speleothems, and more research is required to understand the conditions that favor their formation.

Similarly, other environmental magnetic studies have observed a long-term correlation between precipitation and magnetic mineral concentrations in speleothems (Bourne et al. 2015; Zhu et al. 2017; Chen et al. 2019; Regattieri et al. 2019), but it is likely that, as researchers gain ever finer views of magnetic minerals inside speleothems, our understanding of the environmental processes that deliver them to the stalagmite will become more nuanced across a range of climates, karst systems, and time scales.

CONCLUSIONS

The paleomagnetic and environmental sensitivity of the magnetic minerals present in speleothems provides us with remarkable records of past changes in geomagnetic field behavior and local and regional climate. Understanding the processes that drive these changes is relevant in our own time: the stability of the Earth's magnetic field influences the efficacy of modern satellite communication, and anthropogenic activity is altering hydrogeologic systems and precipitation patterns. In this light, the improved spatial and temporal resolution of tools available for the study of speleothem magnetism will help illuminate our understanding of geodynamo behavior and environmental systems well into the future.

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