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PAGES news

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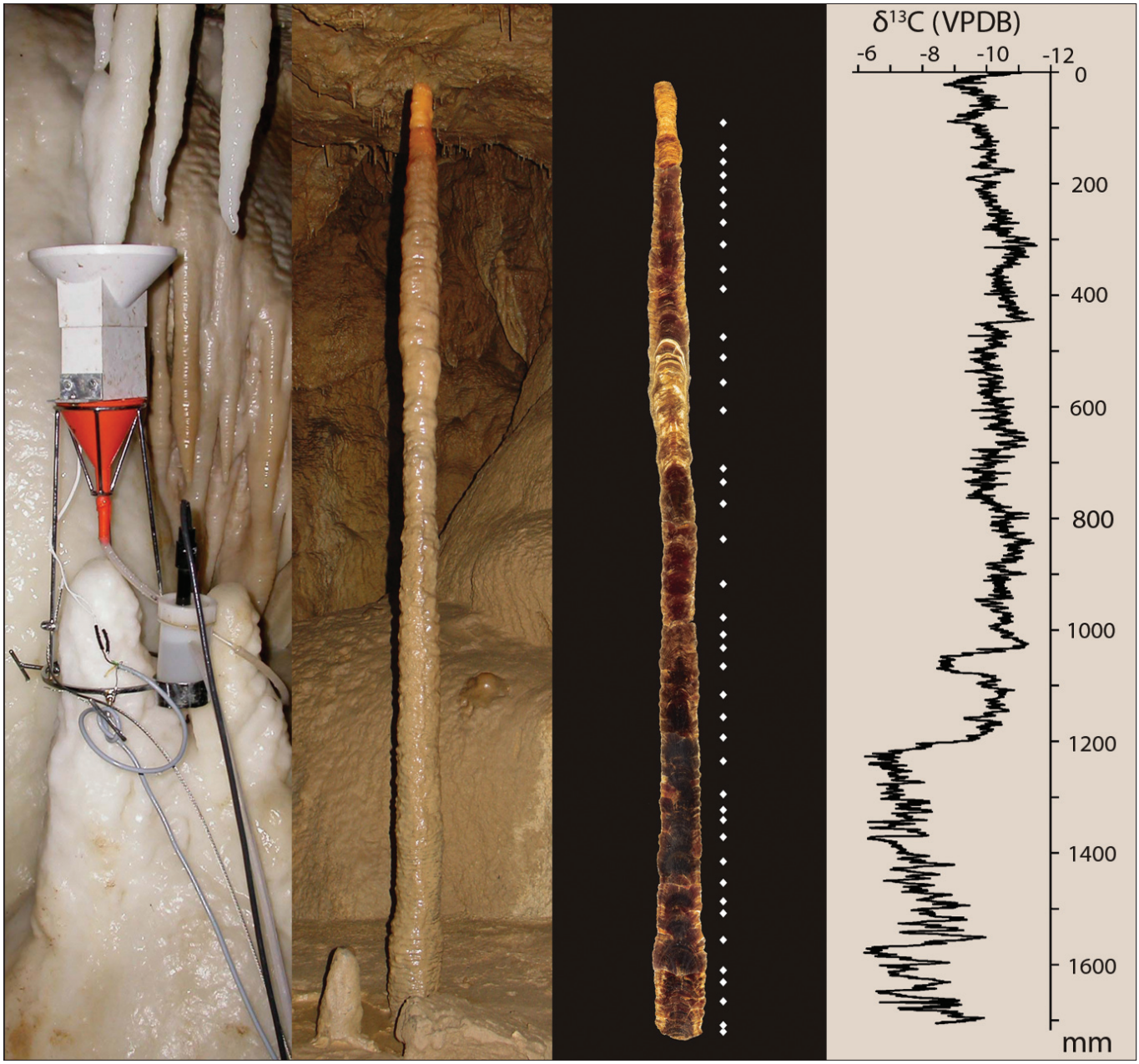
Advances in Speleothem Research

Editors:

Dominik Fleitmann, Christoph Spötl, Louise Newman and Thorsten Kiefer



*From the cave to the curve: **1**) Long-term drip-water monitoring station in Obir Caves (Austria), measuring discharge, drip-water temperature and electrical conductivity, and sampling for chemical and isotopic analyses (photo by C. Spötl); **2**) Stalagmite So-1 from Sofular Cave (Turkey) prior to sampling and **3**) after cutting, diamonds denote uranium-series ages (ranging from 300 - 50 000 yr BP) (photos by D. Fleitmann); **4**) High resolution $\delta^{13}C$ profile of So-1 (Fleitmann unpubl. data).*

Editorial: Advances in Speleothem Research

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"For paleoclimate, the past two decades have been the age of the ice core. The next two may be the age of the speleothem". This recent statement by Gideon Henderson (*Science*, 313: 620-22, 2006) suggests that speleothems (i.e., stalagmites, stalactites and flowstones) have the potential to provide ice core-like records of past climatic and environmental changes.

The marked rise in publications on speleothems over the last decade (see Fig. 1) is therefore not surprising and reveals the growing interest among paleoclimatologists in this fairly novel climate archive. Speleothems have several advantages in comparison to other paleoclimate archives (e.g., ice cores, tree rings, ocean and lake sediments). Firstly, caves containing speleothems can be found in almost all parts of the world. Secondly, uranium-series dating permits the dating of speleothems with exceptionally high precision back to ~600 kyr BP. In contrast to radiocarbon dates, uranium-series dates are absolute ages and no calibration is needed. Therefore, precisely dated speleothem records play a key role in dating climatic events and transitions, such as Dansgaard-Oeschger cycles or glacial inceptions and terminations. Thirdly, speleothems grow continuously over long time intervals (10^3–10^5 years) and thus long and highly resolved time series covering several glacial-interglacial cycles can be developed. Fourthly, facilitated by recent analytical and technical advances, a number of geochemical and physical parameters, such as stable isotope ratios, trace elements and annual band thickness, can be routinely measured at decadal- to sub-annual-resolution. These parameters bear information about the world above the cave, including rainfall, temperature, vegetation, soil productivity and glacier extent.

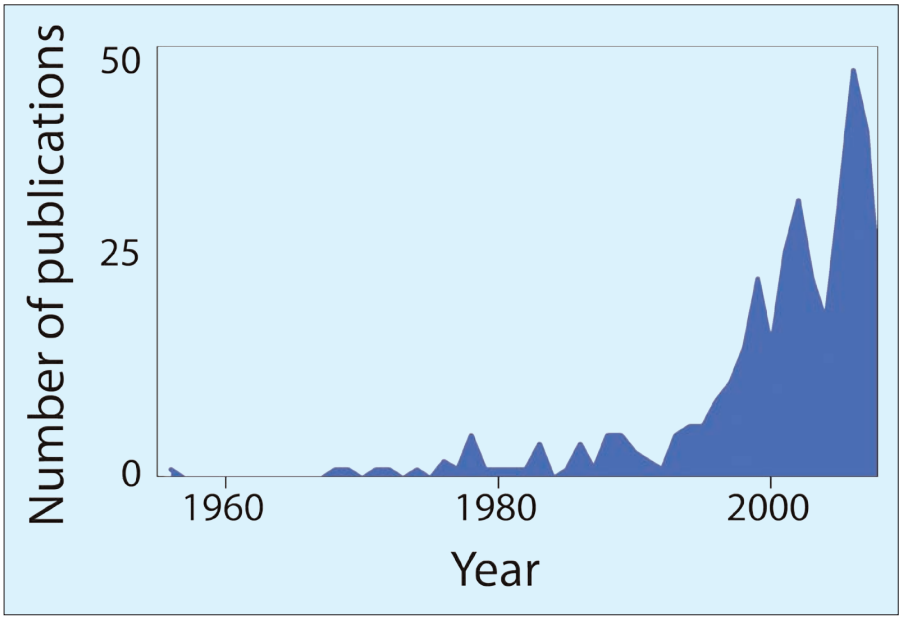

Figure 1: Plot of the number of published papers on speleothems and stalagmites (source ISI's Web of Science, 2008 publications included up to June).

Thus at first glance, speleothems appear to be a perfect climate archive. Yet, with more studies being performed on these deposits, the community is beginning to realize that a variety of climatic, environmental and hydrologic parameters influence the geochemical and physical properties of speleothems and their interpretation as climate proxies is rarely straightforward in a quantitative sense. Each cave has its unique geological and environmental setting, which needs to be understood before stable isotopes, trace elements, annual bands or other proxies can be used with confidence for paleoclimatic and paleoenvironmental reconstructions.

Long-term cave monitoring programs and experimental studies are thus mandatory for caves from which speleothems are collected. Monitoring caves is logistically and technically demanding and few research groups currently have the resources to operate such programs, yet in order to firmly establish (calibrated) speleothem proxy data in the future, comprehensive monitoring is imperative. In conjunction with advanced statistical techniques (e.g., those used in tree-ring studies), these monitoring studies are an important step in advancing this field towards more quantitative climate reconstructions.

This PAGES newsletter focuses on several important aspects of speleothem-based paleoclimate research ranging from cave monitoring and studies of established and novel geochemical and physical proxy indicators, to dating issues and the development of long time series. We hope that these examples enlighten the reader on the current status of speleothem research and the strength and further potential of this archive in paleoclimatic and environmental reconstructions.

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Inside PAGES

PAGES Office is moving

In October, the PAGES International Project Office (IPO) will be relocating to new offices within the Oeschger Centre for Climate Change Research at the University of Bern. We are grateful to the University for enabling the IPO to be placed into the multi-disciplinary research environment of the Centre. Our new contact details will be announced soon.

Recent SSC and EXCOM meetings

The PAGES Science Plan & Implementation Strategy (SPIS) dominated discussions at the Scientific Steering Committee meeting in Cape Town, South Africa in May. Other major business included a review of workshop proposals (see www.pages-igbp.org/calendar/ for upcoming PAGES-related meetings), and planning discussions on PAGES OSM and YSM (see below), as well as the election of new SSC members for 2009. The latter will be announced in the first newsletter issue next year. The meeting resulted in plenty of ideas for the IPO, SSC and paleocommunity to bring to life. Minutes from this and the related Executive Committee meeting are available to download at www.pages-igbp.org/people/sscmembers/meetingminutes.html

PAGES 3rd Open Science Meeting

The theme for PAGES 3rd OSM is "Retrospective Views on Our Planet's Future". The meeting will be held in Corvallis, Oregon, USA, from 8-11 July 2009. A preliminary program is available from the meeting website (www.pages-osm.org/) and p. 38 of this newsletter. Further information will be posted on the website as it becomes available. Please have a look and mark you calendars!

PAGES 1st Young Scientists Meeting

The 1st YSM, to be held alongside the OSM from 6-7 July 2009, also has a website (www.pages-osm.org/). We are hoping to secure funding for young researchers, in order to maximize attendance from all parts of the world. If you would like to help and know of a funding source in your country, please contact Thorsten Kiefer (kiefer@pages.unibe.ch). PAGES is excited about this new venture and hopes for a productive 1st YSM.

PAGES/CLIVAR Intersection

The steering panel of the PAGES/CLIVAR Intersection met in June in Trieste, Italy, to review the group's objectives and strategies for the coming years. Concrete plans include a workshop on forward modeling of climate proxies and downscaling of model results, to be held alongside PAGES OSM. All new ideas for the P/C Intersection are formulated in an updated vision document that will be made available online soon.

Another PAGES/CLIVAR project, the Paleoclimate Reconstruction Challenge, received generous support from NOAA and has now officially started. The "challenge" for the community is to reconstruct synthetic (modeled) climate scenarios from synthetic (pseudo) proxies. Dig deeper at www.pages-igbp.org/science/prchallenge/

PAGES Sessions at AGU

Through our working groups Arctic2k, Global Monsoon and Paleoclimate Reconstruction Challenge, PAGES is co-sponsoring 3 sessions at the AGU Fall Meeting in San Francisco (15-19 Dec. 2008):

- C30: Arctic2k - Assessment of Arctic Climate Records of the Last Two Millennia and Their Relevance for Future Warming
- PP20: Asian Monsoon History and Arid-Region Environmental Changes: Global and Regional Significance
- PP24: Advancing Process Understanding in Proxy Climate Records

We encourage you to contribute to or attend these sessions. The deadline for abstract submission is 10 Sep. 2008.

Next issue of *PAGES news*

The next issue of PAGES newsletter will contain a special section on polar paleoscience and will be guest edited by Julie Brigham-Grette (University of Massachusetts, USA; juliebg@geo.umass.edu) and Ross Powell (Northern Illinois University, USA; ross@geol.niu.edu). If you are interested in contributing a science highlight to this special section, please contact Julie or Ross directly. Equally welcome are your "open" contributions, unrelated to the polar topic. The next deadline for manuscript submissions to the open section is 30 Sep. 2008. Guidelines for these contributions can be found at www.pages-igbp.org/products/newsletters/instructions.html

PAGES Calendar 2008

01 - 03 Oct 2008 - Bernin (Grenoble), France
Past Interglacials Workshop
www.pages-igbp.org/calendar/

29 - 31 Oct 2008 - Shanghai, China
Symposium: Global Monsoon and Low-Latitude Processes
www.pages-igbp.org/science/monsoon/

10 - 13 Nov 2008 - Venice, Italy
EPICA Open Science Conference: Quaternary Climate from Pole to Pole
www.epica2008.eu/

24 - 28 Nov 2008 - Santiago, Chile
4th Alexander von Humboldt International Conference - The Andes: Challenge for Geosciences
www.dgf.uchile.cl/%7ergarreau/EGU-Andes/

08 - 12 Dec 2008 - Victor Harbor, Australia
Australasian Quaternary Association (AQUA) Biennial Conference
www.pages-igbp.org/calendar/

6 - 11 Dec 2008 - Christchurch, New Zealand
Open LUCIFS Workshop: Compiling Records of Holocene Erosion and Sediment Transport
www.pages-igbp.org/calendar/

16 - 18 Feb 2009 - Dalat City, Vietnam
Climate Variability in the Greater Mekong River Basin: paleo-proxies, instrumental data, historical records and model projections
www.pages-igbp.org/calendar/

05 - 09 Mar 2009 - Chandigarh, India
3rd LIMPACS Conference - Holocene Lakes: Climatic Instability and Salinization
www.himclimate.in/html/limpacs

Absolute chronologies from the ocean: Records from the longest-lived, non-colonial animals on Earth

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Although there is an extensive network of annually dated, terrestrial-based proxies in the northern hemisphere for the last millennium (e.g., NRC, 2006), such records are scarce in the marine realm. Most existing annually resolved, marine-based proxy records (corals) are biased to the tropical oceans. In part, research in the mid- to high-latitude oceans has been hindered by a lack of suitable high-resolution marine archives for sclerochronological studies. Sclerochronology is the broad study concerning the accretionary hard tissues of organisms (e.g., mollusks, corals, fish) and can be regarded as the aquatic counterpart of dendrochronology. Sclerochronological techniques are used to develop a master chronostratigraphy within biogenic carbonates, which is then used as a template for growth and geochemical analysis.

The long-lived bivalve mollusk *Arctica islandica* (Linnaeus, 1767), common in the shelf seas of the temperate to sub-polar North Atlantic, has enormous potential as a high-resolution marine archive. This stationary benthic clam is highly suitable for environmental and climate studies because (1) it is extremely long-lived (up to 3-4 centuries; Schöne et al., 2005; Wanamaker et al., 2008a), (2) it produces annual increments in its shell (Jones, 1980; Fig. 1), (3) regional increment series can be cross-matched, demonstrating a common response to environmental forcing(s) (Schöne et al., 2003; Fig. 2), (4) fossil shells can be cross-matched and floating shell chronologies can be constructed after radiocarbon dating (Scourse et al., 2006), (5) live-caught shells can be cross-dated with fossil shells to assemble very long, absolutely dated growth records (Marchitto et al., 2000), (6) master shell chronologies can be created that are as statistically robust as tree ring chronologies (Witbaard et al., 1997; Helama et al., 2007), (7) *Arctica islandica* is widely distributed in the mid- to high-latitudes of the North Atlantic throughout the Holocene (see Scourse et al., 2006), and (8) the geochemical signature (^{14}C, $\delta^{18}O$, $\delta^{13}C$) from the shell material and master shell-growth chronologies can be used to reconstruct ocean circulation, hydrographic changes, and ecosystem dynamics (Weidman and Jones, 1993; Weidman et al., 1994; Witbaard et al., 2003; Schöne et al., 2005; Wanamaker et al., 2008b). The generation of annually resolved marine proxy data from the extra-tropical Atlantic Ocean, using *A. islandica* and other suitable archives (e.g., Halfar et al., 2008) to document and interpret environmental change, will be a major research goal for the next decade of marine paleoclimate research.

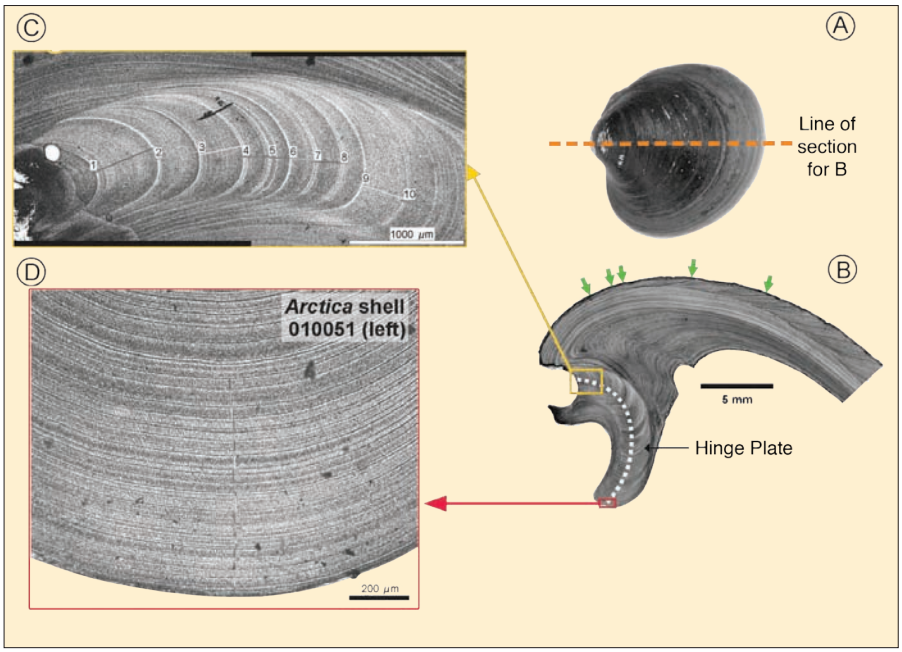

Figure 1: Idealized preparation of an Arctica islandica *shell for sclerochronological studies (from Scourse et al., 2006).* ***A****) External valve face, showing line of section through the left valve used to generate acetate peel replicas;* ***B****) Acetate peel replica cross-section of valve showing annual growth band increments along shell margin (arrows) and within the hinge plate. The axis of growth through the hinge plate used to generate increment data is indicated by the dashed white line;* ***C****) Numbered annual growth bands are measured from the earliest growth bands to the most recent along the axis of growth. Juvenile early bands are wide and reflect the ontogenetic growth curve of the individual;* ***D****) Narrow senescent late bands from the outer part of the hinge plate axis.*

Some *A. islandica* research highlights

Weidman and Jones (1993) first showed the potential for using the radiocarbon content from the annually banded shells of *A. islandica* to monitor ocean circulation in the northwestern Atlantic (Georges Bank). Interestingly, the oceanic response to atmospheric-bomb testing during the

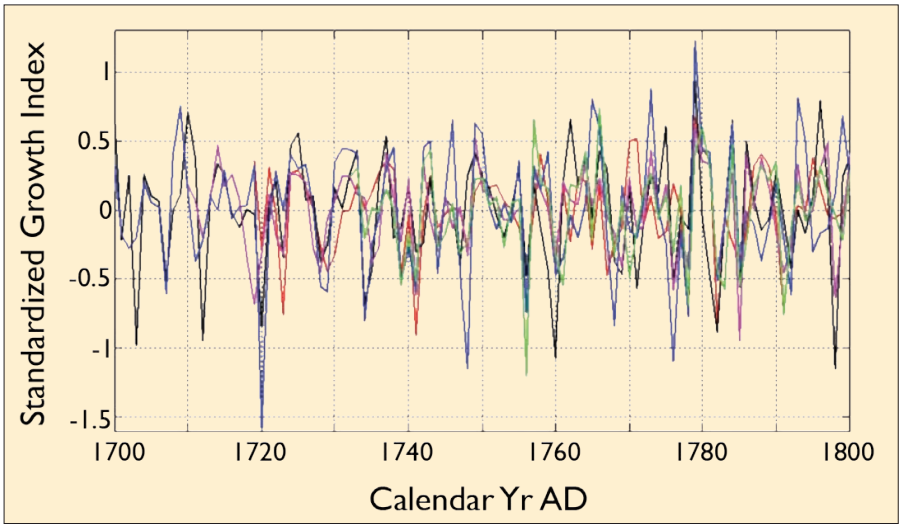

Figure 2: Incremental growth series measured from five dead-collected A. islandica *shells from the Irish Sea (off the Isle of Man) showing a high degree of synchronicity. Data are from P.G. Butler (unpublished). Each shell growth record was normalized with a natural logarithmic function, then detrended with a 15-year spline to remove the ontogenetic growth trend.*

1950s on Georges Bank was attenuated compared to the tropical Atlantic. This result highlighted important differences in ocean mixing processes between the low- and mid-latitudes, and it likely indicated that there was a deepwater source for the waters of Georges Bank. Further, it was illustrated that the oxygen isotopes from the shell material of *A. islandica* were precipitated in isotopic equilibrium with the ambient seawater ($\delta^{18}O_{water}$) (Weidman et al., 1994). Therefore, sub-annual to annual seawater temperature records could be constructed if the $\delta^{18}O_{water}$, which is related to salinity, could be constrained. It was later shown that absolutely dated master *A. islandica* shell chronologies could be accurately constructed from live-caught and fossil material (Marchitto et al., 2000). Recently, it was demonstrated that fossil *A. islandica* shells could be successfully cross-matched using initial 'range finding' radiocarbon measurements and a rigorous comparison among shell growth series (Scourse et al., 2006). In the North Sea, shell growth records from *A. islandica* were used to infer changes in zooplankton cycles and productivity, which seemed to be in part related to the North Atlantic Oscillation (NAO) (Witbaard et al., 2003). Schöne et al. (2003) showed a remarkable positive relationship between the winter NAO index and *A. islandica* shell growth series from the central North Sea and the Norwegian shelf, and suggested that the NAO was impacting shell growth via its influence on food supply.

Recently, with ultra-high-resolution sampling of an *A. islandica* shell, it was demonstrated that there is no ontogenetic trend in oxygen or carbon isotopes (Schöne et al., 2005). This result is important for reconstructing past ocean environments because it means that geochemical records from both young and old shell portions can be used to infer past conditions. In addition, it was found that *A. islandica* off Iceland continue to deposit shell material during the winter months, indicating that there is no substantial 'shut-down' period (Schöne et al., 2005). This result was later corroborated in the Gulf of Maine (Wanamaker et al., 2008b). The data illustrated in Figure 3 shows that *A. islandica* is comparable to corals, providing ultra-high-resolution proxy records for centuries at a time.

Recently, we counted 405 annual bands in a sectioned *A. islandica* shell from the north Icelandic shelf, and radiocarbon analysis from the first annual shell layer (ontogenetic age 0-1) confirmed its sclerochronological age (Wanamaker et al., 2008a). We then calculated the reservoir age of the waters north of Iceland for ca.

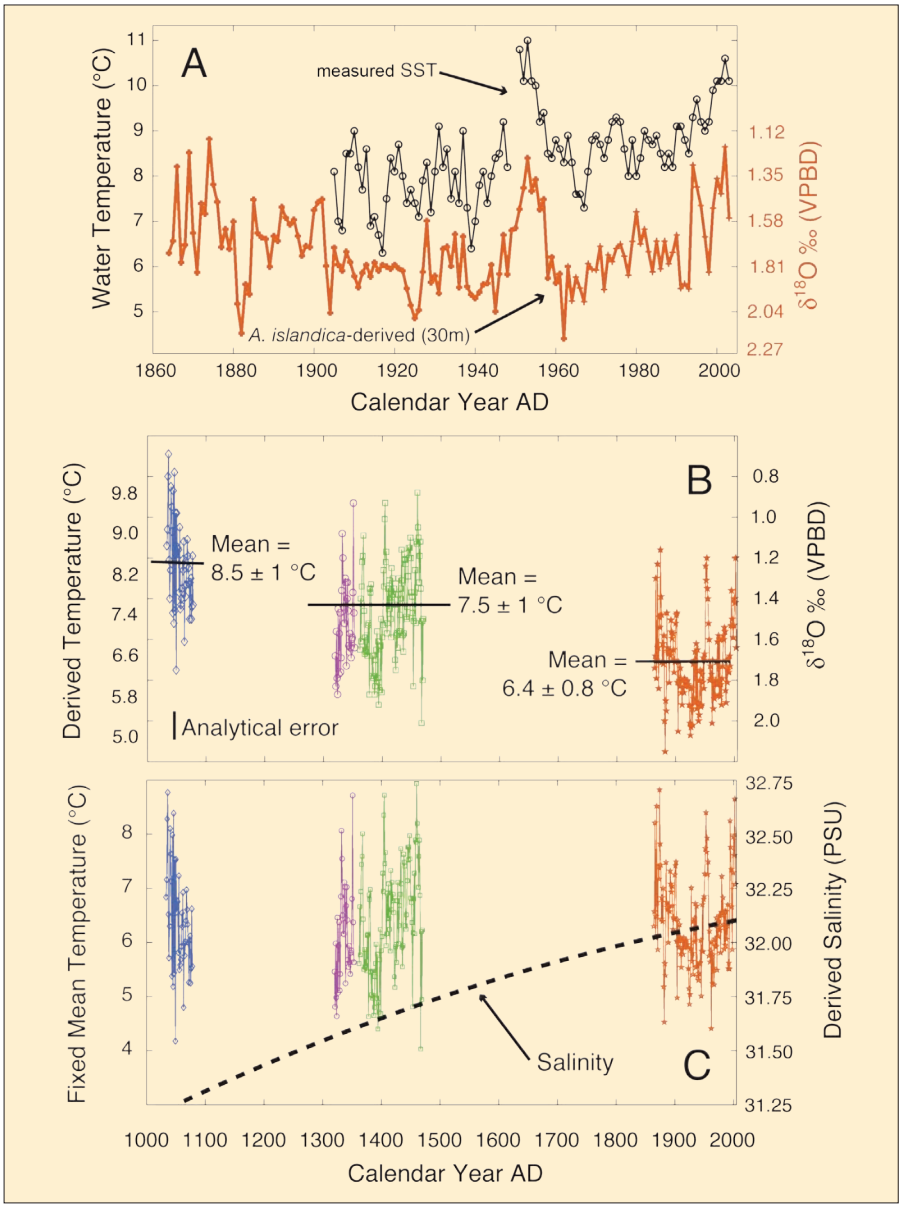

Figure 3: Oxygen isotope record ($\delta^{18}O$) from A. islandica *shells from the Gulf of Maine (modified from Wanamaker et al., 2008b);* ***A****) Modern relationship between a local sea surface temperature (SST) instrumental record and the shell-derived water temperature at 30 m water depth. Nearby salinity values from 1928-2003 were used to constrain $\delta^{18}O_{water}$ values, based on an established salinity/$\delta^{18}O_{water}$ mixing line;* ***B****) Late Holocene shell-derived water temperatures (based on mean 50 m salinity values prior to 1928) and $\delta^{18}O$ record (indicates a cooling trend over the last millennium);* ***C****) Late Holocene shell-derived water salinity. Dotted line represents possible salinity increase in the Gulf of Maine if long-term mean annual seawater temperatures remained constant. The data in both (B) and (C) estimate maximum end-member conditions for the Gulf of Maine (see Wanamaker et al., 2008b for details).*

AD 1600, which was found to be 637 ± 35 ^{14}C years. This reservoir age compares rather well with tephra-based ages from sediment cores from the same location, which are about 600 ^{14}C years at AD 1650 (e.g., Eiríksson et al., 2004). The >400 year *A. islandica* is considered to be the oldest mollusk yet discovered and perhaps the longest-lived non-colonial animal. We have begun to build a 1000-year master shell chronology from the north Icelandic shelf to reconstruct paleoceanographic changes in a region that is very sensitive to climate change. Preliminary results indicate that shell growth from the north Icelandic shelf is highly synchronous among samples from this region, and approx. 50% of the interannual growth variation is related to seawater temperatures during the summer (Wanamaker et al., 2008a).

Future applications

Shell data from *A. islandica* can be used to reconstruct meridional overturning circulation changes in key regions in the North Atlantic. Radiocarbon analyses from *A. islandica* shells can provide a continuous calibration of the marine reservoir age with an absolutely dated chronology (e.g., Wanamaker et al., 2008a). Furthermore, data from master shell chronologies can help characterize rates and/or the type of change (step-like, monotonic, complex) over key climate periods during the Holocene (e.g., 8.2 kyr event, Medieval Warm Period/Little Ice Age transition, and 20th century warming) in key oceanographic settings. Ultra-high-resolution geochemical data can provide seasonal information (amplitude and variability) on water temperature changes and seasonal stratifica-

tion dynamics for the shallow shelf seas in the North Atlantic. Recent advances in geochemical techniques, including the "carbonate clumped isotope" method (Ghosh et al., 2006; Came et al., 2007), provide an excellent opportunity to reconstruct past ocean temperatures independent of $\delta^{18}O_{water}$ or salinity using *A. islandica* and other suitable archives. Although *A. islandica* is restricted to the North Atlantic, other long-lived bivalves with annual banding, such as the geoduck clam (*Panopea abrupta*) from the Pacific, can also serve as reliable climate proxies (e.g., Strom et al., 2004), extending the geographical range in which ocean climate can be reconstructed using the methods described here.

Summary

A. islandica is a remarkable, yet underutilized, marine archive. Geochemical and master shell-growth records have the potential to greatly improve our understanding of key climate events/transitions, as well as ecosystem changes in the North Atlantic throughout much of the Holocene. Its great longevity, its fidelity as a proxy record, and its abundance and wide geographical distribution make *A. islandica* a key proxy archive for the North Atlantic region.

Acknowledgements

We thank the participants who attended the *A. islandica* workshop held at the Gregynog Conference Center, Wales in March 2008. This research is funded by European Climate of the Last Millennium (http://geography.swan.ac.uk/millennium/) (Project no. 017008).

Note

Isotope data for Figure 3 is available from the NOAA paleoclimate database www.ncdc.noaa.gov/paleo/data.html

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For full references please consult:

www.pages-igbp.org/products/newsletters/ref2008_3.html

On the abyssal circulation in the Atlantic basin at the Last Glacial Maximum

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Our understanding of oceanic variability on timescales longer than the time span of direct oceanographic measurements (about a century for most common measurements) relies on our capability to interpret the marine sediment record. Sediment observations have reached the point where hypotheses regarding oceanic conditions during specific time intervals of the geological past can be tested. An interval of preeminent interest is the Last Glacial Maximum (LGM, ca. 20 kyr BP), when large ice sheets occupied North America and northern Europe, and global sea level was reduced by more than 100 m. Much effort has been devoted to estimating oceanic conditions during the LGM, in particular in the Atlantic basin. Hypotheses regarding the ocean circulation during the LGM are particularly relevant, given the postulated role of ocean circulation in climate change. Here we report on a test of the null hypothesis that observations from glacial sediments in the Atlantic basin are consistent with the modern circulation.

A conventional view

Among the most common measurements performed on glacial sediments are two isotopic ratios of calcite shells of benthic foraminifera (bottom-dwelling organisms): the oxygen isotopic ratio $^{18}r = {}^{18}O/{}^{16}O$ and the carbon isotopic ratio $^{13}r = {}^{13}C/{}^{12}C$. Both ratios are usually expressed as a relative

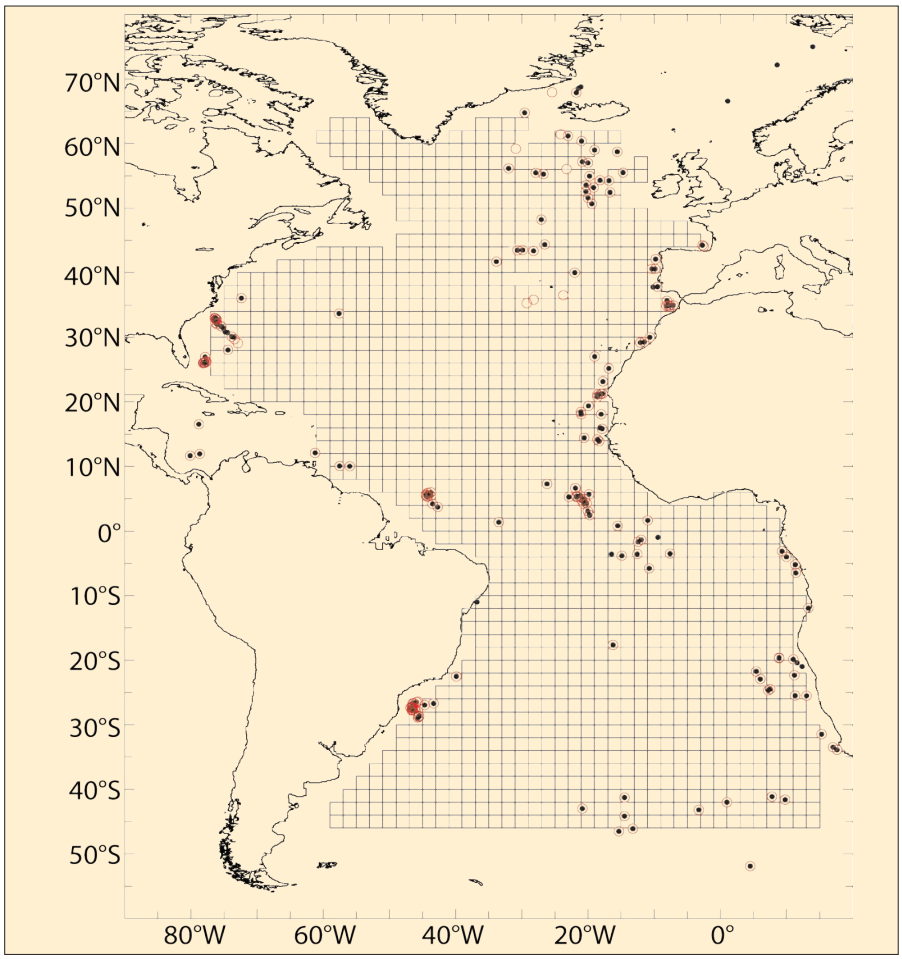

Figure 1: Location of the sediment cores considered in the compilation of benthic foraminiferal data for the Holocene (black dots; 198 measurements; defined as 0-3 kyr BP) and LGM (open red circles; ^{18}O measurements; 18-21 kyr BP). Compilation includes data from both earlier syntheses and other sources. The benthic $\delta^{18}O$ and $\delta^{13}C$ measurements were conducted exclusively on the benthic taxa Cibicidoides *and* Planulina. *Both post-glacial and glacial values are available at most core locations. Locations span the depth range 280–5105 m, with 50% (80%) of the cores from depths shallower than 2500 m (3750 m). Figure modified from Marchal and Curry, 2008.*

deviation from a standard ratio, i.e., $\delta^{18}O = (^{18}r/^{18}r_s - 1) \times 1000$ and $\delta^{13}C = (^{13}r/^{13}r_s - 1) \times 1000$, where $\delta^{18}O$ and $\delta^{13}C$ units are per mil. Comparison of each ratio in benthic foraminifera from surface sediment with properties of ambient bottom water show that, in general, the benthic foraminiferal $\delta^{18}O$ ($\delta^{18}O_b$) varies with the $\delta^{18}O$ and temperature of the water (e.g., Lynch-Stieglitz et al., 1999), whereas the benthic foraminiferal $\delta^{13}C$ ($\delta^{13}C_b$) varies with the $\delta^{13}C$ of dissolved inorganic carbon ($\delta^{13}C_{DIC}$) (e.g., Duplessy et al., 1984).

The observation that the benthic foraminiferal $\delta^{18}O$ and $\delta^{13}C$ generally reflect bottom water properties suggests that measurements of these ratios on fossil shells could constrain the same properties in the past. The dominant deepwater masses in the modern Atlantic—the North Atlantic Deep Water (NADW, with $\delta^{13}C_{DIC} \geq 1‰$) and the Antarctic Bottom Water (AABW, $\delta^{13}C_{DIC} \approx 0.4‰$)—are characterized by different $\delta^{13}C_{DIC}$ (Kroopnick, 1985). Thus, $\delta^{13}C_b$ measurements from Atlantic sediments would also constrain the distribution of water masses of northern and southern origin. The most recent data compilation has been taken to imply the existence in the glacial Atlantic of a southern source water near 1000 m water depth (with low $\delta^{13}C_{DIC}$), a northern source water near 1500 m (high $\delta^{13}C_{DIC}$), and a southern source water below ca. 2000 m (low $\delta^{13}C_{DIC}$; Curry and Oppo, 2005). These inferences resonate with a conventional view that the glacial analoge of NADW was shallower and the glacial analoge of AABW was more voluminous in the Atlantic basin.

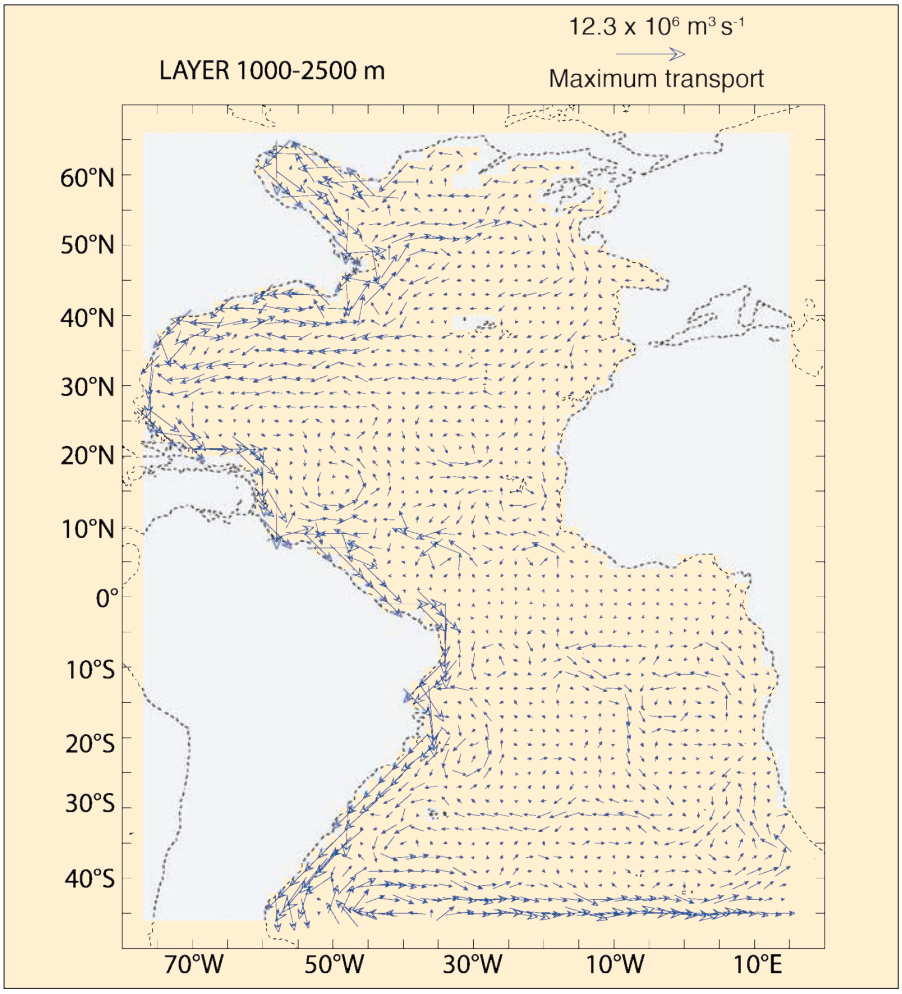

Figure 2: Distribution of the horizontal water transport (in units 10^6 m^3 s^{-1}) between 1000–2500 m in the modern Atlantic, estimated by combining hydrographic data and a circulation model. Maximum transport is 12.3 x 10^6 m^3 s^{-1}. Dashed line and white areas show the coastline and model topography, respectively. The hydrographic data used are (i) climatologies of temperature and salinity; (ii) observational estimates of volume transport of NADW, AABW, and Mediterranean Outflow Water at specific locations; and (iii) observational estimates of the zonally integrated meridional flow at different latitudes based on transatlantic sections. The circulation model is a non-linear, geostrophic, finite-difference model. Horizontal flow is dominated by southward current along the western boundary (NADW) and several coherent structures in the interior. Figure modified from Marchal and Curry (2008).

The challenge

The conventional notion that the distribution of NADW and AABW was different in the glacial Atlantic has not gone unchallenged. LeGrand and Wunsch (1995) argued that NADW and/or AABW, which are "formed" at high latitudes, could have entered the deep Atlantic with initial $\delta^{13}C$ values that are different from today, which would then be recorded in the sediment. They showed that different assumptions about these initial values lead to significantly different depictions of these water masses in the glacial Atlantic. Whereas some of the values they assume may not be consistent with recent observations, the effect of varying initial composition on inferences about water mass distribution can be significant (e.g., Rutberg and Peacock, 2006). LeGrand and Wunsch used an inverse method to conclude that benthic $\delta^{18}O$ and $\delta^{13}C$ data for the LGM are consistent with any flux of NADW in the North Atlantic. None of these data, they argued, can provide constraints on the rates of water motion.

New insights

Recently, we have used an updated compilation of benthic $\delta^{18}O$ and $\delta^{13}C$ data from Atlantic sediments (Marchal and Curry, 2008; Fig. 1) to provide a new test of the null hypothesis that these data are consistent with the modern circulation in the abyssal basin. First, an inverse method was applied to produce an estimate of the abyssal circulation in the modern Atlantic (Fig. 2). This circulation estimate served as a reference state for the null hypothesis that the sediment data are compatible with the modern flow. Second, the inverse method was used to combine, with the modern flow, estimates of two different water properties derived from the sediment data: The $\delta^{18}O$ of equilibrium calcite or $\delta^{18}O_c$ (derived from $\delta^{18}O_b$)—which is a linear combination of water $\delta^{18}O$ and temperature—and $\delta^{13}C_{DIC}$ (derived from $\delta^{13}C_b$).

We found that relatively small adjustments in the $\delta^{18}O_c$ distributions for the Holocene and LGM are necessary to bring these distributions into consistency with the modern circulation. On the other hand, the adjustments in the $\delta^{13}C_{DIC}$ distributions to comply with the modern flow are larger. Assuming that (i) the $\delta^{13}C_{DIC}$ estimates have an uncertainty of 0.1‰ and (ii) deep water $\delta^{13}C_{DIC}$ is governed primarily by a balance between the effects of water advection and organic matter remineralization (organic matter has a low $^{13}C/^{12}C$, so its oxidation tends to depress the $\delta^{13}C$ of ambient DIC), the number of $\delta^{13}C_{DIC}$ values closest to core locations that are adjusted by more than 2 standard deviations is 8 for the Holocene and 25 for the LGM (Fig. 3). These numbers correspond, respectively, to 7% and 21% of the total number of $\delta^{13}C_{DIC}$ values at grid points closest to core locations (note that the number of these points is less than the number of core locations, since more than one core location may have the same grid point as the closest point). Thus, under the assumptions

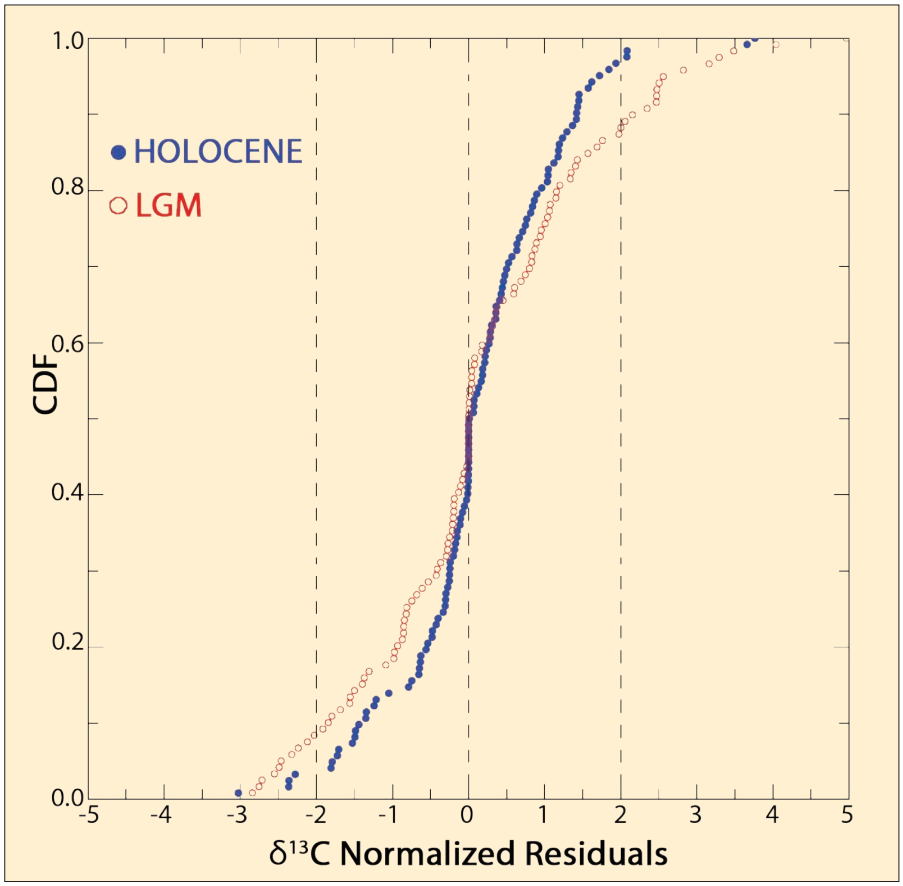

Figure 3: Cumulative distribution function (CDF) of the $\delta^{13}C_{DIC}$ normalized residuals at the grid points closest to core locations. The $\delta^{13}C_{DIC}$ normalized residual is the difference between the posterior value of $\delta^{13}C_{DIC}$ (i.e., the value after inversion or "demanded" by the modern circulation) and the prior value of $\delta^{13}C_{DIC}$ (i.e., the value before inversion or "demanded" by $\delta^{13}C_b$ data), divided by the standard deviation in the prior value. Thus, a normalized residual larger than 2 in absolute magnitude signifies that the prior value of $\delta^{13}C_{DIC}$ needs to be adjusted by more than 2 standard deviations in order to be consistent with the modern circulation. A large fraction of such residuals would suggest that the benthic $\delta^{13}C_b$ data are not consistent with the modern circulation. Figure modified from Marchal and Curry, 2008.

stated above, compatibility of the glacial $\delta^{13}C_b$ data with the modern circulation can be rejected with a certain confidence.

Note that the uncertainty in the glacial $\delta^{13}C_{DIC}$ estimates should account for the errors both in sediment core chronology and in how representative $\delta^{13}C_b$ measurements are of glacial $\delta^{13}C_{DIC}$. Importantly, it is the statistical dispersion of the difference between $\delta^{13}C_b$ and $\delta^{13}C_{DIC}$ that is relevant for the interpretation of $\delta^{13}C_b$ data in our analysis, not the difference itself. Whereas the errors arising from chronology are not readily quantifiable, the dispersion for $\delta^{13}C$ measurements on the genus *Cibicidoides* appears to be less than 0.1 ‰ on average (Duplessy et al., 1984). Measurements of $\delta^{13}C_b$ are clearly useful, but the current uncertainties in the distribution and budget of $\delta^{13}C_{DIC}$ in the glacial Atlantic must be better understood in order to increase the probability of correctly rejecting H0 (=power of the test).

Prospects

We think that our work significantly extends earlier applications of inverse methods to paleoceanography (LeGrand and Wunsch, 1995; Winguth et al., 2000; Gebbie and Huybers, 2006; Huybers et al., 2007). It also offers several prospects. The geographic locations where the largest adjustments in $\delta^{13}C_{DIC}$ estimates are needed to reach consistency with the modern circulation have been identified. These locations provide guidance as to where additional measurements (in sediment as well as in water) should be conducted to better constrain the uncertainties in the glacial $\delta^{13}C_{DIC}$ estimates and hence increase the power of the test. Furthermore, our work has considered only two types of paleoceanographic observation. Other types of observation have been presented as constraints on deep ocean circulation in the geological past (for a recent review see Lynch-Stieglitz et al., 2007). Although the number of such observations is much less than the number of $\delta^{18}O_b$ and $\delta^{13}C_b$ data, they should be considered if an exhaustive analysis of paleoceanographic observations for the glacial Atlantic is to be attempted.

Note

Data are available from NOAA National Geophysical Data Center www.ngdc.noaa.gov/

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Climate variability recorded in tropical and sub-tropical speleothems

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Speleothems provide records that fill an important gap in knowledge about past terrestrial environmental conditions. They complement paleoclimate records from marine/lacustrine sediments, corals and ice cores by supplying additional information about hydrologic variability on land from a wide array of karst settings, ranging from the deep tropics to the Alps. The combination of precipitation and temperature paleoclimate records help to describe the mechanisms of climate changes, and can be used as targets for global climate models run with different climatic forcings.

Speleothems have several advantages as paleo-proxies that make them well-suited to the study of past terrestrial climates. First, speleothems form in caves, which are ubiquitous on all continents, including the tropics, where ice cores are rare. Second, the uranium-decay series provides the means to measure absolute ages on speleothems as old as ~600 kyrs with relative errors of ~0.5-1% (Edwards et al., 1987; Cheng et al., 2000). Third, several different proxies (some geochemical and some morphological) can be used to probe patterns of past hydrological variability. Fourth, average growth rates of ~10-100 µm/yr yield decadally resolved proxy records, although in some cases faster growing speleothems allow for the generation of sub-annually resolved records (Treble et al., 2003; Johnson et al., 2006). Lastly, speleothems form over many millennia, yielding long records of hydrologic variability. These attributes mean that long high-resolution records can be obtained from areas, such as the tropics, which are poorly resolved by other paleoclimate archives.

There are many proxies that have been successfully applied in speleothems, including oxygen isotopic composition ($\delta^{18}O$), carbon isotopic composition ($\delta^{13}C$), trace metal ratio (i.e., Mg/Ca), and growth rate/annual band thickness. Arguably, the most robust proxy in speleothems is carbonate $\delta^{18}O$, which tracks rainfall $\delta^{18}O$ variability at the site assuming equilibrium carbonate precipitation (although see Mickler et al., 2004 for a discussion of disequilibrium effects). Rainfall $\delta^{18}O$ variability is driven by large-scale evaporation, precipitation, and circulation patterns. To a first approximation, rainfall $\delta^{18}O$ is correlated to temperature at high-latitude sites, and anti-correlated to precipitation amount at tropical sites. In the tropics and sub-tropics, the empirical, inverse relationship between rainfall amount and rainfall $\delta^{18}O$ is called the "amount effect" (Dansgaard, 1964; Rozanski et al., 1993). Efforts to calibrate speleothem proxies, including $\delta^{18}O$, to instrumental climate data have identified key environmental controls on speleothem proxy composition, such as rainfall amount, rainfall source, and temperature.

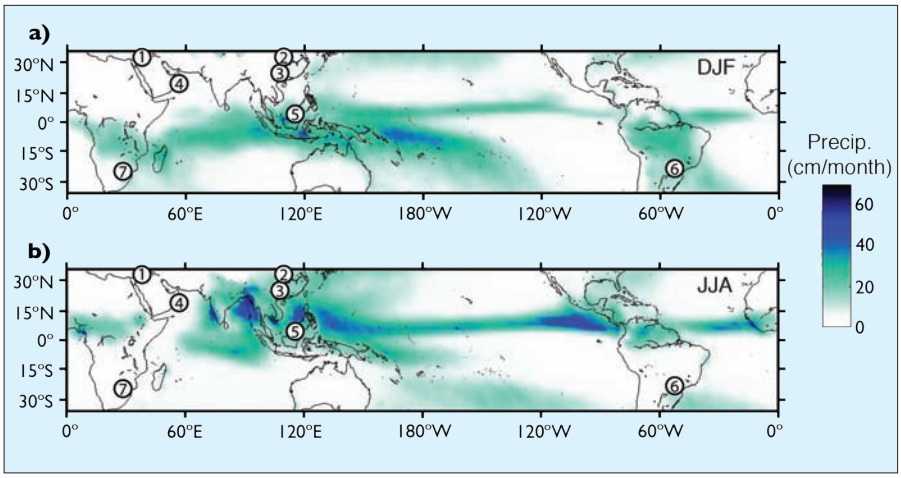

*Figure 1: Austral (**a**) and boreal (**b**) summer precipitation (cm/month), where the values represent an average over the months of December, January, and February (DJF) and June, July, and August (JJA) respectively. Numbers indicate the geographical location of Holocene speleothem records plotted in Figure 2. 1) Soreq Cave, Israel (Bar-Matthews et al., 2003); 2) Heshang Cave, China; (Hu et al., 2008); 3) Dongge Cave, China (Wang et al., 2005); 4) Qunf Cave, Oman (Fleitmann et al., 2003); 5) Snail Shell and Bukit Assam Caves, N. Borneo (Partin et al., 2007); 6) Botuverá Cave, Brazil (Wang et al., 2006); 7) Cold Air Cave, S. Africa (Holmgren et al., 2003).*

An excellent example of a speleothem-based map of the paleo-hydrologic cycle comes from the last 10 kyr, when precessional insolation forcing dominated. At the beginning of the Holocene (~10 kyr), boreal summer insolation was at a relative maximum and austral summer insolation was at a relative minimum. Over the course of the Holocene, the strengths of the summer seasons in each hemisphere reversed, such that boreal summer insolation is presently at a relative minimum and austral summer insolation is at a relative maximum.

Speleothem records prove that these changes in summer insolation impacted the strength and position of deep convection throughout the tropics (seasonal extremes of which are shown in Fig. 1). These speleothem records exhibit sensitivity to changes in the strength and/or position of the large-scale atmospheric circulations associated with monsoonal troughs and/or the Intertropical Convergence Zone (ITCZ). In all cases, the speleothem records suggest that summer insolation forcing is one of the most important drivers for tropical hydrological variability over the Holocene (Fig. 2). For example, locations in both Oman (Fleitmann et al., 2003) and China (Wang et al., 2005; Hu et al., 2008) lie near the northern limit of the Indian-Asian Monsoon system, and speleothem records from these sites display an early Holocene minimum in $\delta^{18}O$ (interpreted as a maximum in rainfall). As a result, these precipitation records display a maximum in precipitation during the early Holocene when boreal summer insolation was at a maximum (Fig. 2). An early Holocene precipitation maximum is even present in the Mediterranean climate of Israel (Bar-Matthews et al., 2003), suggesting that summers there may have been wetter 10 kyr ago (Fig. 2).

Alternatively, speleothem records from Brazil (Wang et al., 2006) and South Africa (Holmgren et al., 2003), which lie at the southern edge of monsoonal circulation regimes in the southern hemisphere, are consistent with early Holocene minima in rainfall (Fig. 2). The wettest part of the record occurs during the late Holocene, consistent with austral summer insolation increasing through the Holocene (Fig. 2).

The records from northern Borneo (Partin et al., 2007) display a mid-Holocene maximum that is consistent with its location at the fulcrum of the northern and southern hemisphere monsoonal circulations. The records from Borneo highlight the complex hydrological response of locations in the deep tropics to seasonal

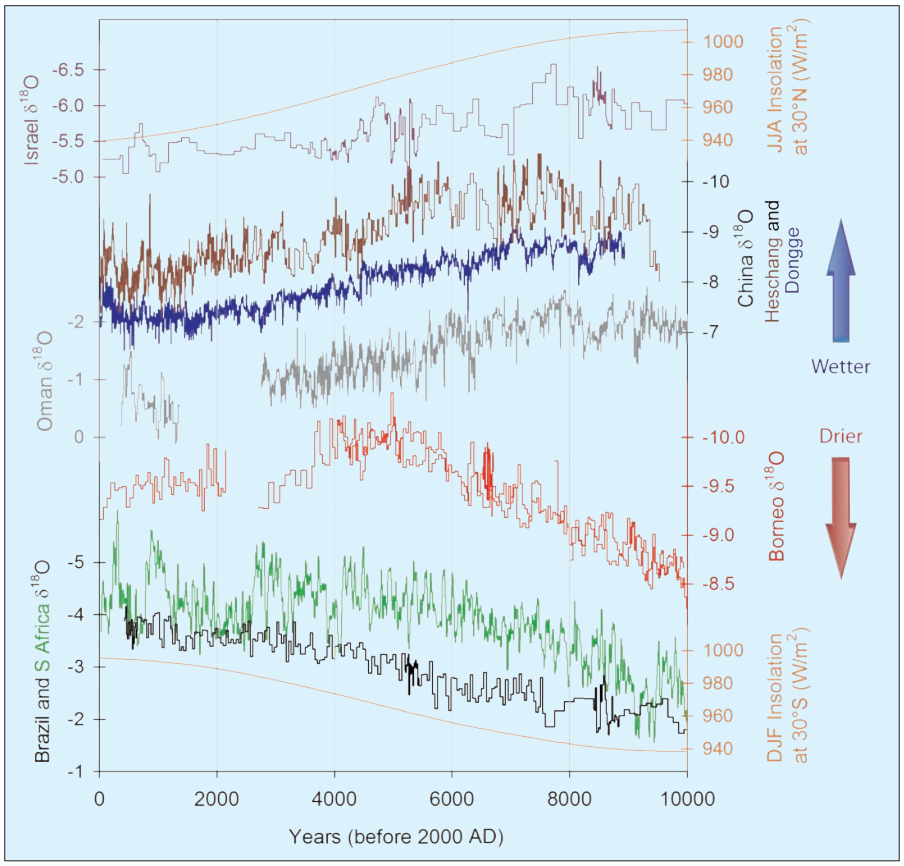

*Figure 2: Speleothem $\delta^{18}O$ records for the last 10 kyr arranged from N to S., Soreq Cave, Israel (**purple**); Heshang Cave, China (**brown**); Dongge Cave, China (**blue**); Qunf Cave, Oman (**gray**); Snail Shell and Bukit Assam Caves, N. Borneo (**red**); Cold Air Cave, S. Africa (**green**); Botuverá Cave, Brazil (**black**). Also plotted are summer insolation curves at 30°N (top) and 30°S (bottom) (**orange** curves). Wetter conditions correspond to smaller $\delta^{18}O$ values and drier conditions larger $\delta^{18}O$ values via the "amount effect" (note that axes are reversed).*

changes in insolation, and the need for additional records from the deep tropics.

Speleothems also offer the opportunity to determine the magnitude and absolute timing of terrestrial climate changes with records that contain sub-annual- to decadal-resolution. This characteristic makes them invaluable for the study of abrupt climate change. For example, a landmark speleothem record from China links abrupt changes in the Asian Monsoon to abrupt climate changes recorded in the Greenland ice cores (Wang et al., 2001). The speleothem records from China have recently been extended to 224 kyr at a temporal resolution of ~40-70 years (Wang et al., 2008). These extraordinary records highlight the ability of speleothems to provide climate information past MIS 5, beyond the limit of the Greenland ice cores. Speleothem records will continue to provide key constraints on the geographical extent to which abrupt climate changes affect precipitation over land.

While this article does not represent an exhaustive review of speleothem records, the small compilation of Holocene speleothem records shown in Figure 2 highlights the potential of constructing a global database of speleothem records that could be used to address a variety of paleoclimate challenges. Tree ring databases for the United States are a standard that speleothem records should try to attain; high spatial coverage at high temporal resolution over a long time period. Many authors have developed speleothem records across the globe from 100 kyr until present, however many gaps still exist. Also needed are companion calibration studies that enhance the climatic interpretation of the stalagmite records. Speleothem records show how the hydrologic cycle is affected by external forcing, as well as how natural climate variability impacts large-scale precipitation patterns. These past changes in the hydrologic cycle have important consequences for water resource planning today as they define the range that planners should consider.

Note

The websites for data from Soreq Cave (Israel), Heshang and Dongge Caves (China), Qunf Cave (Oman), Snail Shell and Bukit Assam Caves (N. Borneo), Botuverá Cave (Brazil), and Cold Air Cave (S. Africa), can be found at www.pages-igbp.org/products/newsletters/ref2008_3.html

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Paleotemperature reconstruction using noble gas concentrations in speleothem fluid inclusions

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Stalagmites are gaining importance in paleoclimate research as they provide high-resolution stable isotope signals over glacial-interglacial timescales. The major difficulty in the interpretation of stalagmite oxygen and carbon isotope data is to disentangle the various effects and processes that control the signals. One important parameter is the mean air temperature in the cave. Cave temperatures remain relatively constant throughout the year at approx. the local mean air temperatures. Accordingly, paleotemperature reconstructions from cave deposits provide good estimates of the past annual mean outside the cave (McDermott, 2004; Genty et al., 2002). However, there is a lack of tools for direct determination of cave paleotemperatures. Noble gases dissolved in water inclusions can provide this informa-

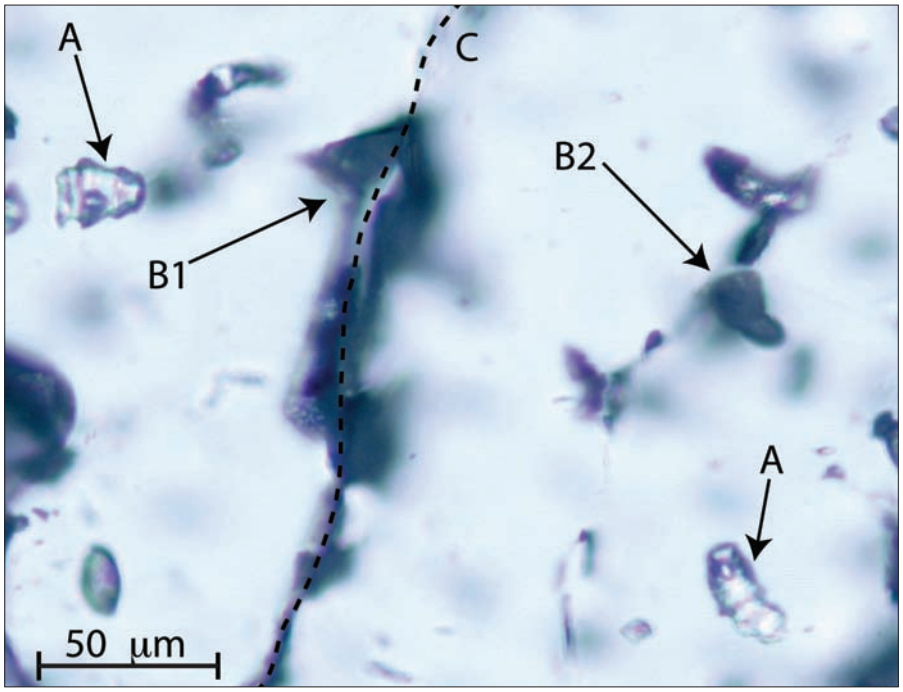

*Figure 1: Photomicrograph of a thin section of a stalagmite (H12 from Oman; see Fleitmann et al., 2007, for further details). **A**: Intra-crystalline water inclusions (with an air bubble), **B1**: Inter-crystalline air inclusion, **B2**: Intra-crystalline air inclusion, **C**: Grain boundary.*

tion, as they are a direct proxy of the temperature during air–water partitioning.

Noble gas thermometer

The concentrations of dissolved noble gases (He, Ne, Ar, Kr, Xe) in water depend on the atmospheric pressure (i.e., the altitude), as well as the temperature and salinity of the water at the time of its last contact with air (i.e., during the entrapment of fluid inclusions in stalagmites). Using the well-established noble-gas concepts employed in lakes and groundwater (e.g., Aeschbach-Hertig et al., 2000; Kipfer et al., 2002), the concentrations of dissolved atmospheric noble gases in water inclusions provide a noble-gas archive that allows the direct and quantitative reconstruction of past cave temperatures.

During stalagmite growth, minute quantities of drip water and cave air are trapped in the calcite in the form of one and two phase (i.e., just liquid, just gas, or liquid and gas) fluid inclusions (Fig. 1). Microscopical investigations showed that air and water inclusions clearly differ in their size and spatial arrangement within the calcite. The abundance of inclusions and the volume ratio of enclosed air to water (V_{air}/V_{water}) are highly variable between different stalagmites and even differ within a single stalagmite (Scheidegger et al., 2007). The water content of a stalagmite lies typically between 0.01 and 0.1 wt.% (Schwarcz and Harmon, 1976).

Noble gas analysis of water inclusions in stalagmites is challenging. First, due to the low solubility of noble gases in water, air inclusions contain atmospheric noble gases in much higher abundance than those filled with water. Noble gases from air inclusions, however, do not contain any information about past climate conditions. Thus, for a successful determination of noble gas temperatures (NGTs), the gases originating from water inclusions have to be separated from those liberated from air inclusions, to reduce the amount of "excess air" ("excess" noble gases from one phase air inclusions and/or air bubbles in water inclusions).

Secondly, noble gas concentrations are required for the calculation of NGTs, therefore the amount of water extracted from the sample has to be known with high precision. These amounts are typically about 1 µl, thus for an average noble gas analysis, relatively large stalagmite samples of ca. 1 cm^3 are needed, leading to an average temporal resolution of ca. 100 years (average growth rate of 50 µm).

Extraction of noble gases

For stalagmites with a high water content and low abundance of air inclusions, noble gases are extracted in a single crushing step. The stalagmite sample is crushed under vacuum in a crusher consisting of a stainless steel cylinder, with a polished steel ball inside that can be moved manually by a permanent magnet placed around the cylinder.

For stalagmites with a higher abundance of air inclusions (V_{air}/V_{water} ~1), a single crushing step leads to a high amount of excess air. This excess air masks the noble-gas signature of the water, thus making it very difficult to calculate NGTs. In this case, stepwise extraction techniques are more suitable. Figure 2a and b show the results of stepwise crushing and stepwise heating (with an initial crushing step) experiments, respectively. Several subsequent extraction steps, by either heating or crushing, lead to noble gas signatures that are closer to air-saturated water and lead to a significant reduction of excess air (from V_{air}/V_{water}

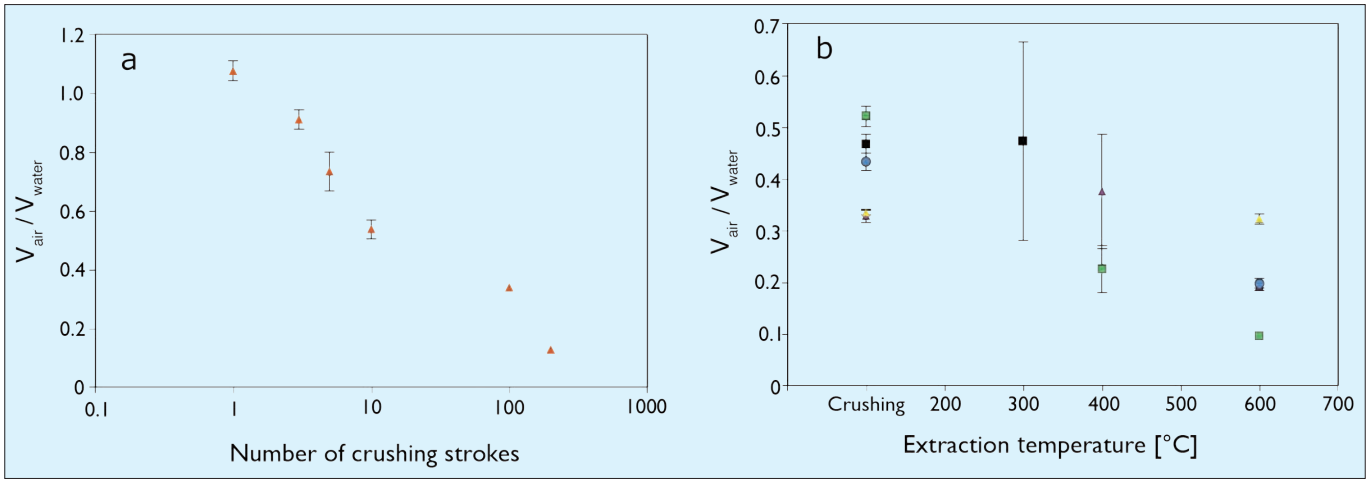

*Figure 2: Results of (**a**) stepwise crushing and (**b**) stepwise heating experiments used to reduce the effect of "excess air". **a**) Air-water volume ratio (V_{air}/V_{water}) of a piece from the H12 stalagmite (Oman) displayed against the number of crushing strokes. V_{air}/V_{water} was reduced by about one order of magnitude using the stepwise crushing extraction. **b**) V_{air}/V_{water} of several stalagmite samples (indicated by different symbols) after an initial crushing step followed by one or several heating steps. Using this technique the value of V_{air}/V_{water} can be substantially reduced (data from Scheidegger, 2005).*

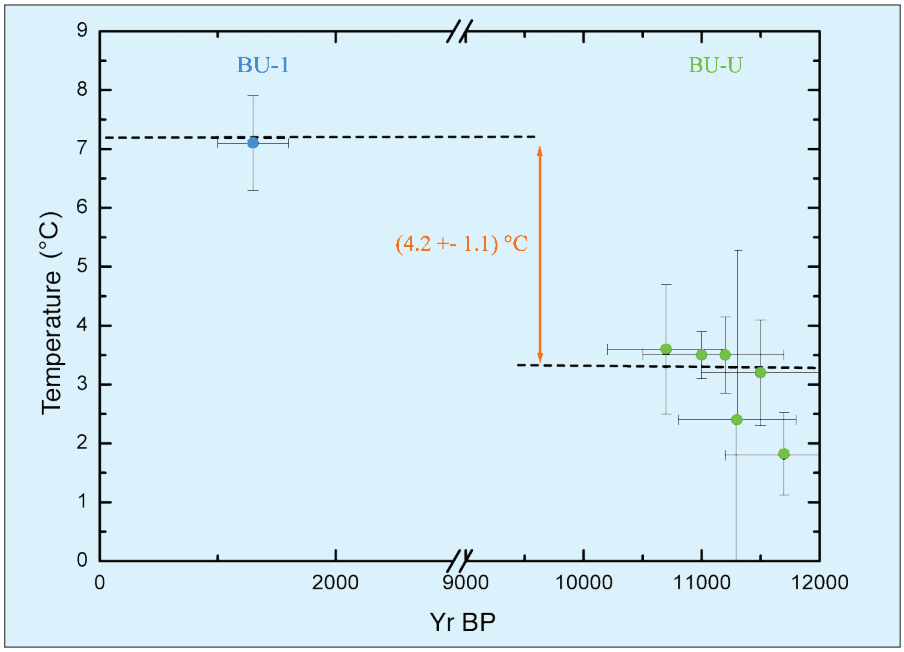

Figure 3: Noble gas temperatures calculated for a sample from a 1300-yr-old stalagmite BU-1 and six sub-samples from an early Holocene stalagmite BU-U.

>1 to V_{air}/V_{water} <0.3). For both extraction methods, the amount of the liberated water is determined manometrically. At ETH-Zurich, the pressure of the water vapor is measured in an accurately known volume and at a well-defined temperature provided by a water bath. This system allows calculation of water amounts in the range of 1 mg with a precision of 1.5%. In Heidelberg, known amounts of water prepared in capillaries are used to calibrate the system. With this procedure, a precision of about 2% is achieved.

Paleotemperatures

In order to derive temperatures from the determined noble gas concentrations, inversion techniques, as developed for groundwater (Aeschbach-Hertig et al., 1999), can be used. It is assumed that stalagmites contain noble gases originating from air-saturated water (water inclusions) and atmospheric air (air inclusions and/or air bubbles in water inclusions). Samples from Bunker Cave (NW Germany) confirm this assumption. In this case, the simple extraction with only a single crushing step is adequate to determine NGTs with a typical uncertainty of about 1°C (Kluge et al., 2008). Six sub-samples have been extracted from the uppermost cm of the stalagmite BU-U from the Bunker Cave, which belongs to one growth layer with an U-Th age of 10.8 to 11.7 kyr BP. They yielded reproducible values with a mean of 2.9 ± 0.7 °C. Although different extraction procedures and measurement methods have been used, e.g., squeezing in copper tubes and crushing in a steel cylinder, the calculated temperatures all fall within the range of uncertainties (Fig. 3). The considerably younger sample from another Bunker Cave stalagmite (BU1), extracted at 5 cm distance from the top, with an U-Th age of 1300 ±300 yr BP, resulted in a higher temperature of 7.1 ± 0.8°C. Thus, the noble-gas-derived temperature difference between samples from the early Holocene (BU-U) and the early Middle Ages (BU-1) was found to be about 4°C (Fig. 3). This agrees well with results from other studies in this region (Davis et al., 2003).

Noble gas concentrations of several samples from other caves, however, show noble gas signatures that are distinct from those expected by a simple mixture of noble gases from air and air-saturated water. Modern stalagmites from caves in Yemen and Oman (Fleitmann et al., 2007) show strong fractionations due to an enrichment in the light noble gases (He, Ne) and partly also in xenon. These results show that the noble gas signatures cannot be explained, in all cases, by a simple mixture of noble gases from air and air-saturated water alone, indicating that the entrapment of water inclusions in the growing stalagmite may be more complicated than initially assumed. For several samples from Yemen, calculated NGTs lie in the range of the modern cave temperature (27 °C), but errors are large (3-6°C) due to the fractionated noble gas signature.

Our results indicate that the determination of NGTs in stalagmites is, in principle, possible. For samples with a very low value of V_{air}/V_{water} (V_{air}/V_{water} <0.1), noble gases are clearly a mixture of air and air-saturated water and therefore NGTs can be calculated with a precision of about 1°C. For other samples, more sophisticated, stepwise extraction techniques that are currently being tested may strongly reduce the value of V_{air}/V_{water} by separating air and water inclusions more quantitatively. These techniques are thus expected to increase the number of samples suitable for NGT determination in the future.

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Paleotemperatures from fluid inclusion liquid-vapor homogenization in speleothems

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To date, paleoclimate information from stalagmites has mainly been obtained from stable isotope compositions ($\delta^{18}O$ and $\delta^{13}C$) and trace element contents of the calcite host mineral (e.g., Wang et al., 2001; Fleitmann et al., 2003). However, stable isotopes and trace elements are influenced by several climatic and environmental factors, making it difficult to employ these climate proxies for quantitative temperature reconstructions. Therefore, we propose a novel approach to determining paleotemperatures, based on direct microthermometric measurements of the liquid-vapor homogenization temperatures (T_h) of fluid inclusions in stalagmite sections of approx. 300-µm thickness.

Analytical Approach

The liquid-vapor homogenization temperature (T_h) is a measure of the fluid density that, for a given composition, depends on the pressure-temperature conditions under which the fluid has been trapped. In the particular case of speleothems, which grow from calcite (over)-saturated water at ambient atmospheric pressure, T_h is theoretically equal to the formation temperature of the fluid inclusions, i.e., the cave temperature, which often corresponds closely to the mean annual surface temperature outside the cave.

Because of the low formation temperatures of stalagmites, all of the fluid inclusions are one-phase liquid at room temperature, except for those in which an air bubble was additionally trapped. In the one-phase inclusions, spontaneous nucleation of the vapor phase generally fails to occur on cooling below the homogenization (calcite formation) temperature. This means that below T_h, the fluid is in a metastable liquid state at negative pressures, i.e., under tension. In order to overcome this metastability, we cool the inclusions to 4.0°C (the density maximum of pure water) using a microscope heating-freezing stage and apply a novel technique that uses single, amplified pulses of a tightly focused femtosecond (10^{-15} seconds) laser to induce nucleation of the vapor phase (Krüger et al., 2007). Once the vapor bubble has formed the fluid is in a thermodynamically stable phase state (on the liquid-vapor curve) and T_h can subsequently be determined by microthermometry in the same experimental setup. Figure 1 illustrates the procedure on the basis of the phase diagram of water. Although nucleation of the vapor bubble requires high laser pulse intensities in the order of 10^{12} W/cm^2 (Tera-Watt), the energy deposited on the sample is relatively low due to the ultra-short pulse duration. Therefore, the laser pulses usually do not induce changes of the inclusion volumes, provided that the pulse intensity is set close to the nucleation threshold.

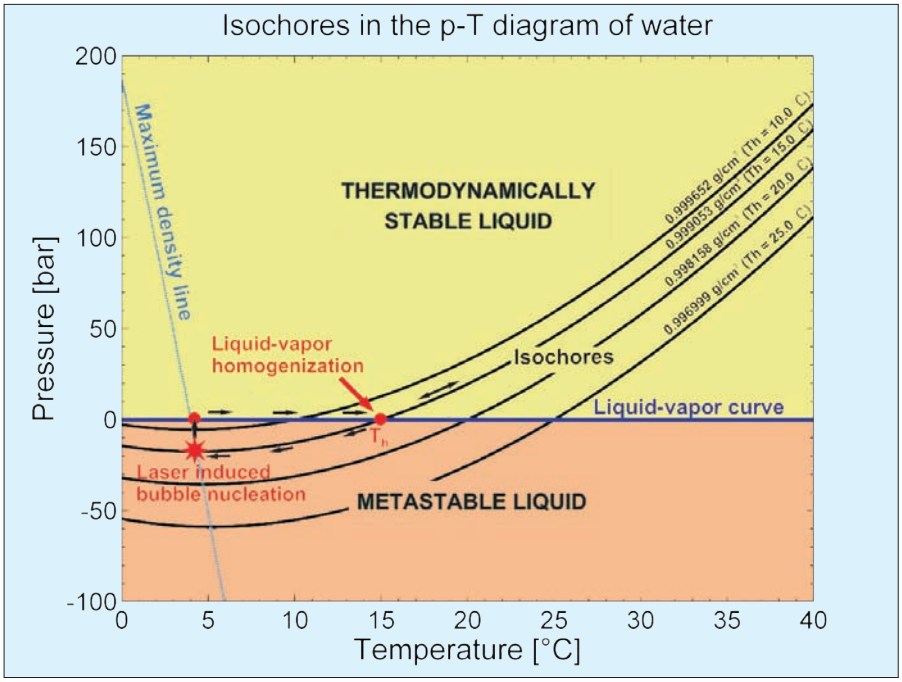

Figure 1: Part of the phase diagram of water as a projection in the Pressure-Temperature plane and calculated from the International Association for the Properties of Water and Steam (IAPWS) 1995 formulation (Wagner and Pruß, 2002). At pressures above the liquid-vapor curve (purple line), liquid water is in the thermodynamically stable phase state, whereas below the liquid-vapor curve at negative pressures (tension forces) liquid water is, although mechanically stable, thermodynamically metastable. In high-density fluid inclusions, where spontaneous nucleation of the vapor bubble generally fails to occur on cooling below T_h, the fluid pressure follows along the metastable extension of the liquid-isochore (line of constant fluid density) to negative values, i.e., the fluid becomes stretched. On further cooling, the liquid-isochores reach a pressure minimum at the intersection with the maximum density line of water (red star). To induce bubble nucleation using ultra-short amplified laser pulses, the inclusions are cooled down to the pressure minimum of the isochores. Due to the vapor bubble formation, the fluid pressure increases to the liquid-vapor curve. On heating the sample, the fluid pressure follows the liquid-vapor curve whereby the liquid phase expands at the cost of the bubble volume. Note, the density of the total fluid (liquid + vapor) does not change because the inclusion volumes are constant (isochoric). At T_h, the vapor bubble disappears and on further heating the pressure leaves the liquid-vapor curve and increases along the isochore in the liquid stability field.

Results

In a study that was conducted to test the feasibility of this novel approach of paleotemperature determination, we measured liquid-vapor homogenization temperatures of 130 previously one-phase fluid inclusions hosted within the most recently formed calcite bands of an actively growing stalagmite from Sofular Cave, Northern Turkey. Although the T_h values showed a wide and apparently unsystematic scatter, ranging from 6 to >30°C, they displayed a clear maximum at ca. 10°C, which closely matches the present day cave temperature of 10.5 to 11.7°C (Fig. 2). The observed variability in T_h does not result from the high intensity laser pulses used for vapor bubble nucleation as the reproducibility of T_h of individual inclusions is, with few exceptions, within ± 0.1°C. Therefore the scatter of the T_h values is attributable to a number of potential mechanisms that can directly affect inclusion volumes, fluid densities and/or liquid-vapor homogenization temperatures:

1. From the equation of the state of water, International Association for the Properties of Water and Steam (IAPWS) 1995 (Wagner and Pruß, 2002), we can deduce that irreversible volume expansion (stretching) of the inclusions of only a few per mil, leads to an increase of T_h by several °C. Stretching of the inclusions may, for example, result from mechanical stress on the calcite host, induced by sample preparation. In order to minimize this effect, we used a low speed rock saw (Bühler Isomet) to cut the 300-µm sections. To avoid additional stress, the samples were not ground

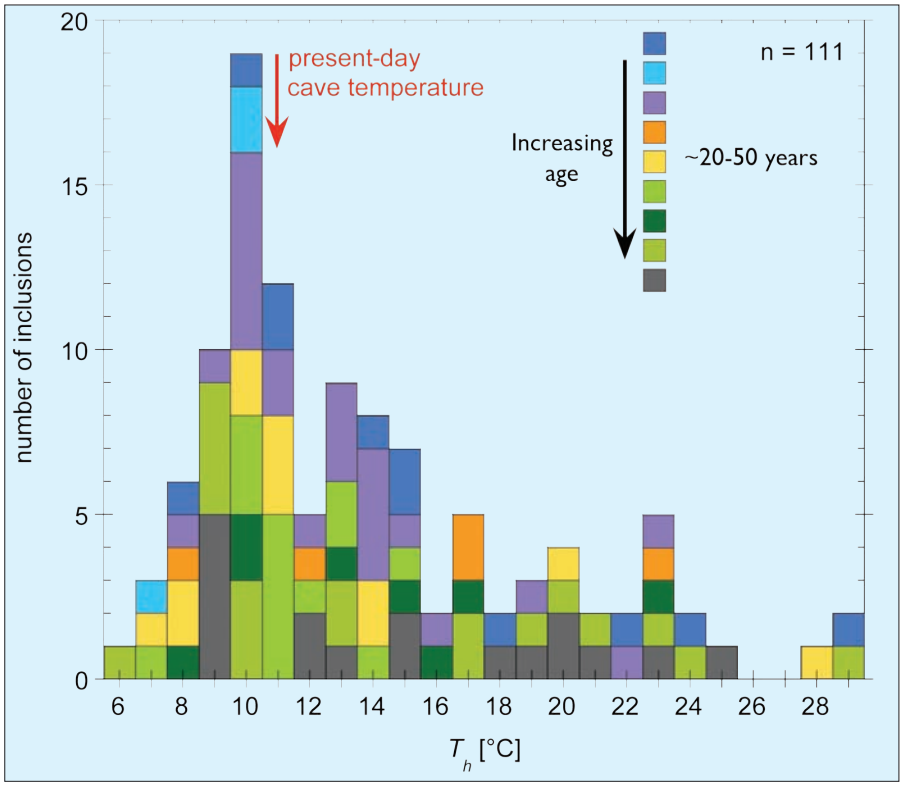

Figure 2: The distribution of 111 (of a total 130) fluid inclusion liquid-vapor homogenization temperatures (T_h) obtained from an actively growing stalagmite from Sofular Cave, Northern Turkey (sample So-2). The inclusions are located within the most recent calcite layers and were formed during the last ~20–50 years. Color scale indicates the relative ages of the inclusions. Temperatures derived from the inclusions can be directly compared to the actual cave temperature. Although T_h values scatter widely, from 6 to >30°C (T_h values ≥30°C are not shown), they display a clear maximum at ca. 10°C, which is close to the present day cave temperature (measured in 2007). Note, there is no correlation between T_h and the age of the inclusions.

and polished, but were instead made transparent using immersion oil.

Besides sample preparation, stretching may also result from high internal fluid pressures, which occur when the inclusions are exposed to temperatures above T_h during transport and storage of the stalagmite samples or in the laboratory. From the phase diagram of water (Fig. 1), it can be seen that fluid inclusions that formed, for example, at 10°C, reach internal pressures of approx. 60 bar at 25°C. To avoid such large internal fluid pressures in future studies, the stalagmite samples have to be kept at a constant temperature (i.e., the cave temperature) during the transport from the cave to the laboratory and during sample preparation.

2. Some of the inclusions have strongly elongated shapes orientated parallel to the growth direction of the stalagmites, as shown in Figure 3. We propose that capillary forces could lead to stretching of the fluid during formation of the inclusions. Thus, the original density of the trapped fluid is too low and does not correspond to the actual formation temperature, and consequently the resulting T_h values are too high. Since this is a primary effect, it cannot be avoided or minimized. It is not yet known to what extent capillary forces affect the fluid density but it is likely that their influence would be larger during dry periods, when the evaporation rate at the surface of the stalagmites increases.

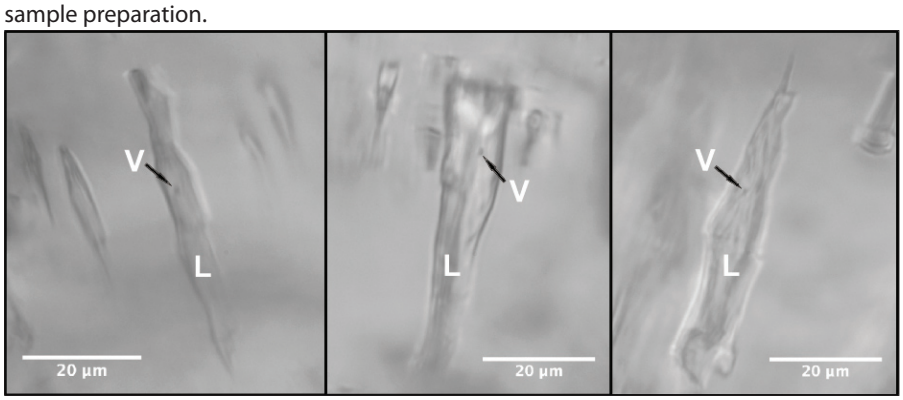

*Figure 3: Fluid inclusions with strongly elongated shapes containing liquid water (**L**) and a tiny bubble of water vapor (**V**) that has previously been induced by single ultra-short laser pulses. Maximum volume of these vapor bubbles only accounts for ~0.04% of total inclusion volume (at 4.0°C). Therefore, the vapor bubble can only be optically resolved in relatively large inclusions (typically >20 – 30 µm in length). The visibility of the vapor phase is additionally hindered by the fact that the vapor bubble generally sticks to the inclusion walls and does not move around within the inclusion.*

3. As the vapor bubble cannot become infinitely small upon heating, T_h can be regarded as the temperature of bubble collapse i.e., the temperature where the bubble is no longer mechanically stable. We therefore assume that T_h does not solely depend on the fluid density but also on the factors that control surface-tension-mediated bubble collapse. Thus collapse of the vapor bubble provides a potential explanation for T_h values that are too low but presumably also for values that are too high, due to mechanisms that can stabilize the vapor bubble. A better understanding of the fundamental physical parameters that control bubble collapse in the fluid inclusion micro-systems could potentially help us to apply a correction to the measured T_h values.

Conclusions and outlook

The results of this study confirm that liquid-vapor homogenization temperatures of fluid inclusions can be used to determine stalagmite formation temperatures and thus represent a promising analytical approach to reconstructing paleotemperature variations. However, considerable effort is still necessary to develop a reliable methodology that provides a reduced scatter of the T_h values and useful criteria for acquisition, processing and refinement of the data. The goal is to achieve an accuracy in temperature determination within ± 1.0°C or better, at a high temporal resolution. In our study, we measured T_h values from fluid inclusions that are hosted within several growth bands that cover approx. the last 20-50 years. Future work will focus on inclusions that formed during distinct climatic events (e.g., the Younger Dryas) to reconstruct paleotemperature evolution. These studies may also provide an insight into potential re-equilibration of the volume properties of the inclusions under changed temperature conditions and recrystallization of the calcite host.

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Cave monitoring and calibration of a $\delta^{18}O$–climate transfer function for a Gibraltar speleothem

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Speleothems represent a fast and direct record of local climatic events and provide rich archives of past climate change. Interpretation of speleothem isotope proxies in terms of regional climate change requires a good understanding of what features of local climate they record, and the relationships between local and regional climate systems (McDermott, 2004; Fairchild et al., 2005). Cave monitoring plays a vital role in understanding the links between seasonal and synoptic-scale weather events and the capture of the $\delta^{18}O$ signal in speleothem calcite, which can be traced back to incoming rainfall via a set of processes that may operate in a multitude of different ways, even within the same cave system.

Cave monitoring in Gibraltar

Cave systems in Gibraltar are in close proximity to the location of the longest instrumental climate (back to 1792) and precipitation isotope (from 1962) records in the Mediterranean region, and are therefore an ideal location to test and calibrate modern capture processes under different cave environments (Mattey et al., 2008). A comprehensive cave monitoring program began in June 2004 in the New St. Michaels Cave system, using new data logging systems designed specially for the Gibraltar project. These included a drip counter that records water discharge at high resolution and a multi-channel monitoring system that simultaneously records CO_2 concentrations at two hourly intervals from seven different sites within the cave system (Fig. 1).

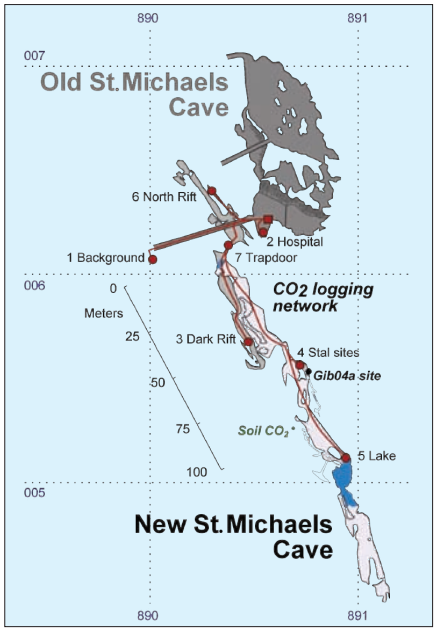

Figure 1: Plan view of the St. Michaels cave system. The show cave (dark gray) and the New St. Michaels system (light gray) are located near the summit at an altitude of 300 m asl. Red circles mark position of monitoring sites at cave entrance (1) and within the cave (2-7); 'Soil CO_2' marks the position of the soil monitoring site above the cave. The tube network leading back to the CO_2 analysis and logging system (red square) is shown as red line. Isotope data for a stalagmite from the Gib04a site is shown in Figure 3. Grid lines are 100 m UTM coordinates. Figure adapted from Mattey et al., 2008.

The seasonality in rainfall, drip rates, cave-air CO_2 concentrations and drip water chemistry are shown in Figure 2. Drip monitoring focused on three sites where speleothem deposition is taking place and reveals strikingly different hydrological regimes, even where drip sites are only meters apart. The site showing the highest discharge rates in Figure 2 is forming new flowstone from highly supersaturated water that dye tracing has shown to descend down bedding planes from a recharge area near the summit of the rock. The discharge pattern at the flowstone site does not immediately respond to the onset of winter rain and suggests the operation of a siphon, where water is accumulating in a reservoir before siphoning over to create the main discharge, which then gradually declines for the rest of the year.

Drip responses for the two other sites (Gib04 and Dark Rift; Fig. 2) indicate that these sites are fed by water percolating downwards across bedding planes from a recharge zone vertically above the cave. The deeper of the two sites, Gib04a, is lo-

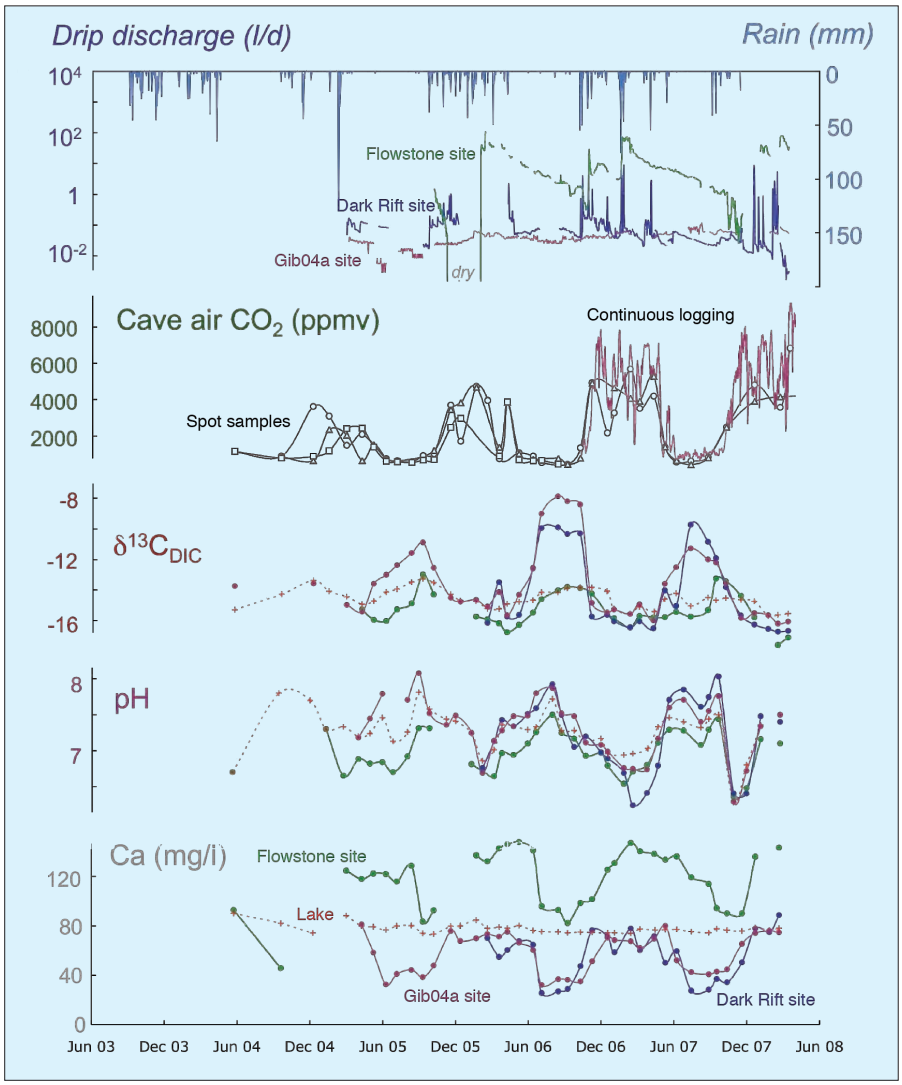

*Figure 2: Some results of the cave monitoring in New St. Michaels Cave from June 2004 to April 2008. From top to bottom: Daily rainfall (**light blue**; Crown Copyright, the Met Office); drip discharge at sites Flowstone (**green**), Dark Rift (**blue**) and Gib04a (**dark pink**), measured at 30-min. resolution by acoustic drip counting; concentration of CO_2 in cave air by spot sampling at three sites (**dark gray** curves) and by continuous logging at 2-hr resolution at the Gib04a site; $\delta^{13}C$ of dissolved bicarbonate, pH and Ca ion concentration in drip waters (same color notation as discharge curves) and in the lake (**red** crosses).*

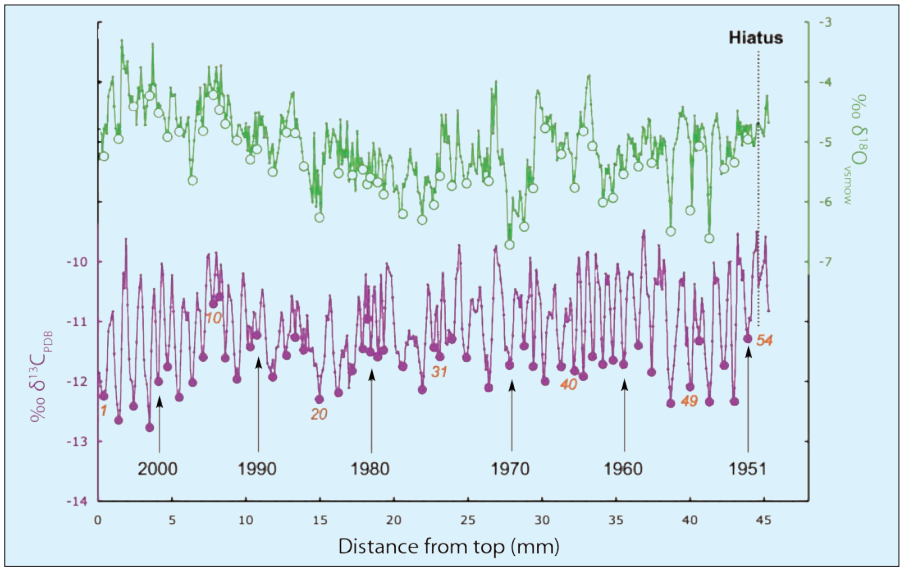

Figure 3: The $\delta^{13}C$ and $\delta^{18}O$ time series obtained by contiguous micromilling in 100 µm steps plotted as a function of distance from top of Gib04a. $\delta^{13}C$ minima, representing calcite deposition in April, are marked with closed circles and the corresponding $\delta^{18}O$ values are plotted as open circles. Annual cycle numbers (orange) and age model relative to year 2004 are also shown. Figure adapted from Mattey et al., 2008.

cated only a few meters from the siphon site described above but shows a very different drip response to rainfall. Here, discharge rates are very low throughout the year, in the order of 40 to 80 ml/day, with only a weak response to incoming winter rain. In contrast, the Dark Rift site, situated among bedding planes at a higher level where the roof thickness is thinner, shows a fracture flow response where drip rate is highly correlated with rain events.

Cave air CO_2 concentrations have been measured from spot samples taken at monthly intervals since 2004, and continuously from October 2006 by the multichannel logging system. CO_2 concentrations show strong seasonality caused by a reversing ventilation pattern driven by the difference in temperature between the cave interior and exterior. Ventilation is not just taking place through local artificial or natural entrances, as these can be temporarily closed off with no effect on the overall seasonal pattern. The results suggest that the ventilation is a diffuse process exploiting fissures through the Gibraltar limestone, which allows large-scale winter convection of CO_2-rich ground-air through the high-level cave systems.

The cave-air CO_2 levels change by almost an order of magnitude from summer to winter and pCO_2 is the main forcing mechanism controlling drip water degassing and calcite precipitation. The seasonal changes in cave air CO_2 strongly influence drip water pH, $\delta^{13}C$ of dissolved bicarbonate and Ca concentrations, which all co-vary with CO_2 (Fig. 2). The enhanced degassing when the cave is ventilated with outside air during the summer drives $\delta^{13}C$ to higher values (Baker et al., 1997; Bar-Matthews et al., 1996; Spötl et al., 2005), resulting in rapid deposition of calcite, which develops a distinctive compact fine-grained microporous texture. High CO_2 levels in the winter result in slower calcite precipitation to form more massive elongate crystals, and year-by-year the speleothem fabric develops as pairs of laminae representing alternate summer and winter growth (Mattey et al., 2008).

An actively growing speleothem at the Gib04 site with well-developed paired laminae also preserves spectacular seasonal oscillations in $\delta^{13}C$ in the calcite (Fig. 3) (Mattey et al., 2008). Monitoring of drip water bicarbonate $\delta^{13}C$ demonstrates that the minimum $\delta^{13}C$ values in the calcite must develop towards the end of the winter season (April-May), and this places a crucial time marker on the more complex $\delta^{18}O$ record of the annual hydrological cycle. The isotopic composition of Gibraltar rainfall varies by up to 8‰, with winter rainfall dominated by the lowest $\delta^{18}O$ values but variability in the $\delta^{18}O$ of drip water, and hence the calcite depositing from it, can become attenuated by mixing and storage in the feeding aquifer (Cruz et al., 2005).

The attenuated oxygen isotope record in the active Gib04a speleothem still preserves a component of annual cyclicity that is superimposed on longer-term trends. The weak annual $\delta^{18}O$ cycles in the Gib04a speleothem record (Fig. 3) may be a result of evaporation of groundwater in the soil (Bar-Matthews et al., 1996) and operation of kinetic fractionation (Mickler et al., 2004), which may increase under 'summer' cave conditions when calcite supersaturation and precipitation rates increase as a response to low levels of cave CO_2. The annual $\delta^{13}C$ cycles provide both a seasonal time marker and a precise chronology, which enables the $\delta^{18}O$ of late winter-deposited calcite to be identified for each calendar year. The calcite $\delta^{18}O$ values recalculated as drip water then provide an upper limit on the annual composition rainfall from 1951 to 2004, and can be compared to the local GNIP rainfall record, providing one of the first opportunities to test a speleothem "isotope recorder" against long instrumental records.

The correspondence between the $\delta^{18}O$ of reconstructed winter drip water and $\delta^{18}O$ of winter rainfall (as weighted

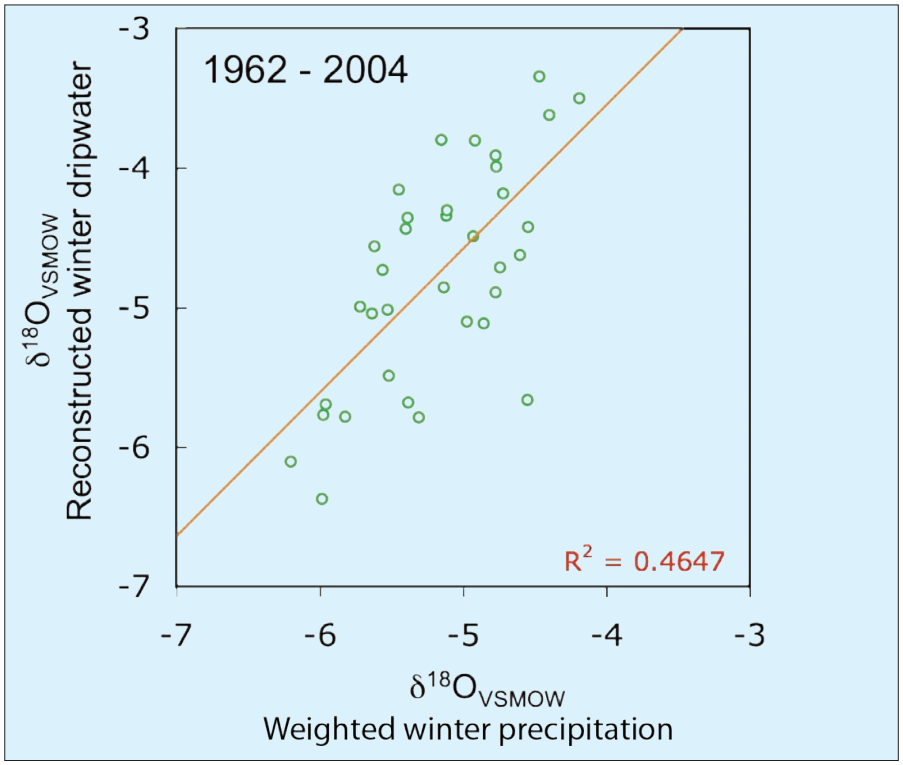

Figure 4: Correspondence of the $\delta^{18}O$ of reconstructed winter dripwater with $\delta^{18}O$ of weighted October-March precipitation for 1961 to 2004 (IAEA/WMO, 2004). Figure adapted from Mattey et al., 2008.

GNIP ONDJFM monthly values) is plotted against the $\delta^{18}O$ of reconstructed winter drip water and shown in Figure 4, which shows a significant correlation with r^2 = 0.47 and a slope of ≈1. Such good correspondence between the instrumental precipitation isotope record and the oxygen isotope signal encoded in speleothem calcite shows that precipitation isotope time series can successfully be reconstructed from speleothem calcite and is an important milestone in producing a calibrated $\delta^{18}O$–climate transfer function, which can then be applied to longer speleothem records.

Conclusions

Monitoring of cave processes over multi-annual time periods provides essential insight into the way that speleothems record climate, and is an important step towards defining the response times of the cave and speleothem "recording systems" to changes in local climate and water balance. Close monitoring of just three sites in Gibraltar reveal significant differences in drip hydrology and speleothem forming processes, and to properly understand the drip water isotope evolution it could be necessary to monitor over 5–10 hydrological cycles to reveal the long term patterns. Monitoring for shorter periods still provides vital insight into the local processes that may bias the isotopic signal, and provides a framework for developing the calibrated $\delta^{18}O$–climate transfer functions that are necessary to unlock the quantitative records that speleothems are capable of providing.

Acknowledgements

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Understanding climate proxies in southwest-Australian speleothems

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There are two avenues for comparing speleothem-derived climate proxy data against instrumental climate measurements: Via speleothems that have grown through the 20th century, and by carrying out dripwater monitoring studies. Both provide opportunities to investigate how speleothem geochemistry responds to changes in intra- and inter-annual variations in climatic parameters. Much work remains to understand these processes in general, and it is becoming increasingly apparent that such exercises are necessary to understand processes in individual caves, owing to the broad variation in climatic, geological and geomorphic environments that host speleothems. Here, we provide an overview of recent research findings from several cave sites in southwest Australia. Multiple geochemical proxies ($\delta^{18}O$, $\delta^{13}C$, P, U, Mg, Sr, Ba and Na) from calcite speleothems from this region have provided the longest duration (80 complete years from 1911-1992) calibration dataset between speleothem geochemistry and the instrument climate record. We use this case study of modern speleothem $\delta^{18}O$ and trace elements to provide examples of techniques for calibration and to highlight the importance of carrying out such investigations.

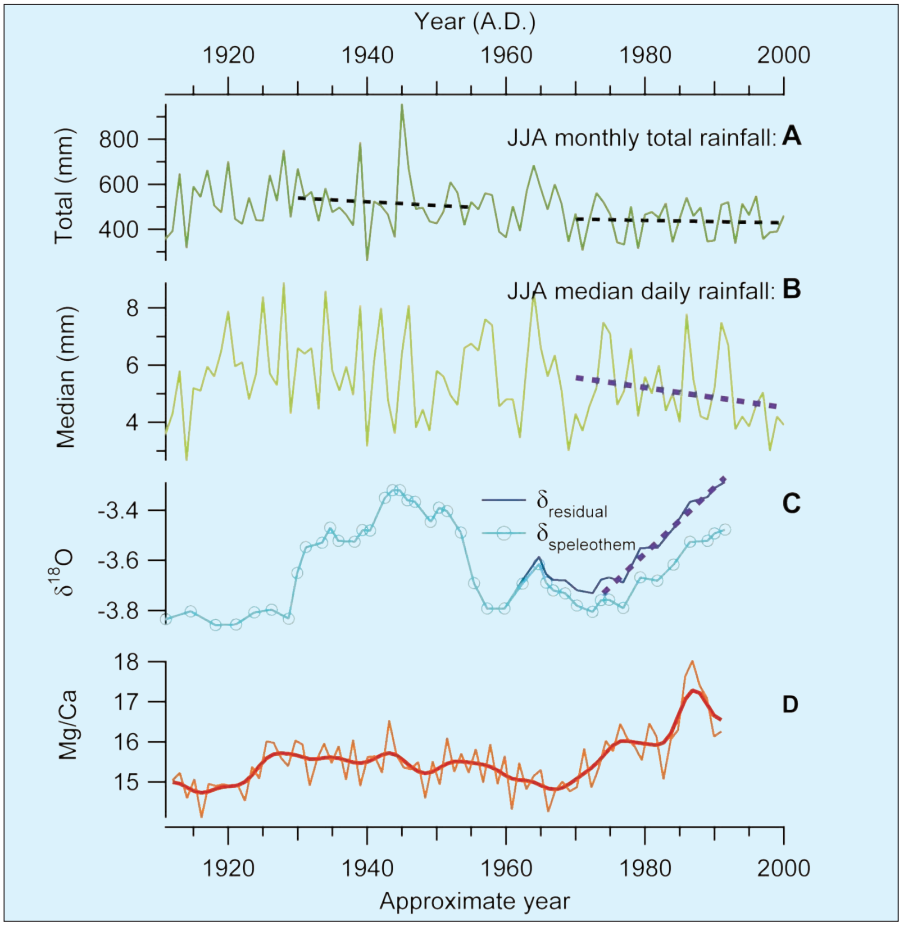

*Figure 1: **A**) Perth total June-August rainfall (instrumental), **B**) median daily rainfall (instrumental), **C**) Moondyne Cave speleothem $\delta^{18}O$ and **D**) Mg/Ca ratios (µmol/mol). Speleothem $\delta^{18}O$ ($\delta_{speleothem}$) is corrected ($\delta_{residual}$) for the rise in mean temperature. Speleothem $\delta^{18}O$ increases during 1930-55, owing to a change in moisture source region (Fischer and Treble, in press).*

Southwest Australia

Southwest Australia provides an ideal location for comparing speleothem records with instrumental data. Caves in this region typically underlie native forests that have not been significantly altered by European settlement. Caves are hosted in porous Pleistocene calcarenite, resulting in a relatively smooth transfer of drip water and its chemical signal by predominantly seepage-type flow. Southwest Australia has a Mediterranean-type climate with distinctly wet winters and dry summers, which give rise to clear seasonal responses in drip water chemistry. On the longer term, rainfall has markedly decreased across the southwestern corner of Western Australia from about 1970 (Allan and Haylock, 1993; Fig. 1A,B), providing a test for the geochemical record. This rainfall decrease was recorded within the growth interval of a stalagmite (MND-S1) that grew in Moondyne Cave between 1911 and 1992 (Treble et al., 2003; 2005). Here, we provide a review of these data, including new findings and drip water data from a nearby cave.

Oxygen isotopes

The instrumental record shows that the mean annual temperature in southwest Australia rose by approx. 0.6°C from the mid-1950s (Indian Ocean Climate Initiative, 2002), until the stalagmite MND-S1 was removed in 1992. The effect of this temperature rise on the $\delta^{18}O$ of the stalagmite (calculated using the temperature-dependent fractionation of $^{18}O/^{16}O$ at calcification (-0.23‰/°C; O'Neil et al., 1969)) was subtracted from the $\delta^{18}O$ data to produce a record reflecting variation in rainfall $\delta^{18}O$ trends between 1911 and 1992 (Fig. 1C). Speleothem $\delta^{18}O$ rose after 1970, which is consistent with the recorded decrease in rainfall (Treble et al., 2005). This result is expected based on the inverse relationship between rainfall $\delta^{18}O$ and amount determined from one year of daily rainfall sampling in 2000-01 (12 km from Moondyne Cave; Treble et al., 2005). A common assumption with climate proxies is that one can apply short-interval calibrations to interpret trends in proxy data back in time. However, MND-S1 showed that this simple inverse relationship between rainfall amount and $\delta^{18}O$ has not been consistent through time, evidenced by the higher $\delta^{18}O$ values between 1930-1955 when there was no measured decrease in rainfall.

This key finding led to a recent investigation where the simple rainfall amount-$\delta^{18}O$ regression model was improved to include interannual climate variance (Fischer and Treble, in press). The new model was modulated by dominant modes of interannual climate variability or climate indices. The dominant interannual modes are calculated as the principal components of sea level pressure over the period 1850-2004 (for which sea level pressure data are available; Allan and Ansell, 2006). In the winter in the southern hemisphere extratropics, these principal components (i.e., dominant modes) of climate variability describe major zonal and meridional pressure anomalies between, and in, the mid- to high-latitudes. The new model produced a positive shift in $\delta^{18}O$ between 1930-55, similar to that in the speleothem record. This suggests that the dominant modes of interannual variability can cause shifts in vapor-source regions (or other effects) that can affect $\delta^{18}O$ independent of amount-type effects.

In southwest Australia, it appears that the interannual mode most responsible for isotopic changes related to vapor source is Zonal Wave 1 (ZW1). ZW1 is a persistent zonal pressure anomaly between the South Pacific Ocean and the South Indian/Atlantic Oceans. An empirical orthogonal function (EOF)-based reconstruction of ZW1 over the last century suggests that, for southwest Australia, a negative ZW1 state from 1930-55 favored the advection of ^{18}O-enriched moisture from low latitudes, while a positive ZW1 state post-1970 resulted in more ^{18}O-depleted moisture advected from the sub-polar region (Fischer and Treble, in press).

As a result, we now have an improved regression model for $\delta^{18}O$ rainfall in southwest Australia that replicates key patterns at daily to inter-decadal timescales. This is a statistical forward model and thus it can be used to compare paleoclimate simulations to proxy data. The inversion of the model will require a multi-proxy approach (e.g., isotopes and trace elements), because the new model relies on two predictors ($\delta^{18}O$ depends on both precipitation amount and vapor source).

Speleothem trace elements

A suite of trace elements was also investigated in speleothem MND-S1 (Treble et al., 2003). Of these, annual speleothem Mg and P concentrations were found to clearly record the rainfall decrease from 1970: Mg increased and P decreased (Fig. 1D; Mg only) but were unaffected by the change in vapor source in the 1930-55 period. Empirically, therefore, the trace elements are able to provide a distinction between the two episodes, which could be applied further back in time. However, the use of empirical relationships without an understanding of mechanisms is unlikely to be a sufficiently robust approach. Annual trace element cycles in the Moondyne speleothem provide evidence for mechanisms but there is a need for drip water studies too. Since Moondyne Cave has become largely dry since the late 1990s and only small areas contain perennially active dripping stalactites, this component of the study was conducted at Golgotha Cave, 20 km away.

Cave drip waters in Golgotha Cave were measured at 4 sites within the cave from July 2005. Drip water Mg/Ca ratios display clear seasonal cycles at all sites (Fig. 2; site 1B shown) similar in form to the annual Mg cycles measured in speleothem MND-S1. In principle, such cycles can be caused by varying amounts of prior calcite precipitation (PCP; see Fairchild et al., 2000): The more calcite precipitated from solution before reaching the stalagmite (in the aquifer or on the stalactite tip), the lower the Ca solution concentration and the higher the Mg/Ca in the solution and the resulting stalagmite. Initially, this was considered to be a simple response to the seasonal drying of the aquifer, leading to high Mg/Ca in austral summer (Treble et

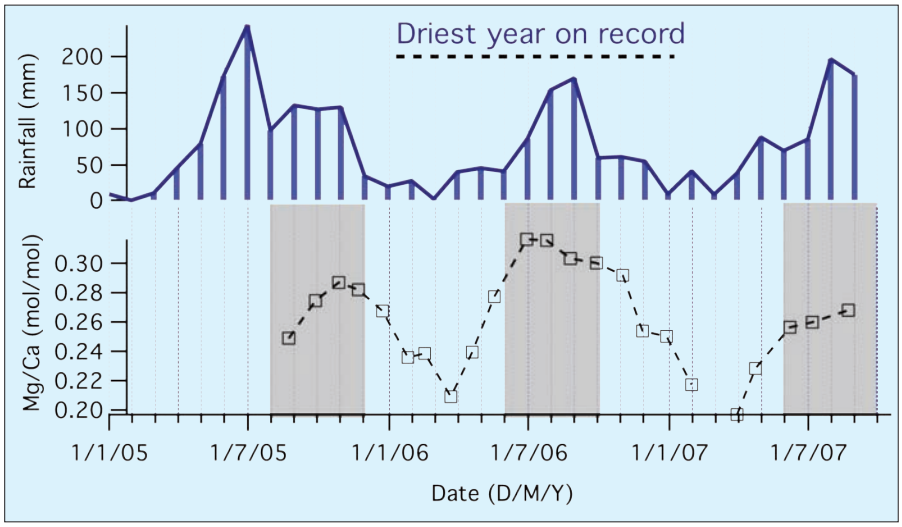

Figure 2: Monthly rainfall and drip water Mg/Ca ratios (mol/mol) for site 1B in Golgotha Cave. Gray shading indicates months when drip water saturation indices ($CaCO_3$) are ≥0.2.

al., 2003) but the drip water study suggests otherwise. Drip water Mg/Ca ratios are in fact higher in the wet winter months than summer, and tracing with O isotopes does not suggest that this is due to a seasonal lag. Rather, we interpret from seasonal variations in pH and alkalinity data that cave PCO_2 is modulating seasonal $CaCO_3$ saturation and hence inducing more PCP in the winter; the PCO_2 variations also have the consequence that speleothem deposition in Golgotha Cave predominantly occurs during the winter/spring months. In more recent work, we have also established that PCO_2 (via solution pH) is the strongest influence on sulfate incorporation and, hence, we propose that sulfate profiles can be used to identify PCO_2 controls. The PCO_2 variations may also have a specific interpretation in terms of seasonality (cf., the Austrian Obir cave where low PCO_2 reflects strong incoming air circulation at low winter temperatures; Spötl et al., 2005).

However, at Golgotha Cave we also observe that the highest Mg/Ca ratios were reached during 2006 (Fig. 2), which was the driest year on record. This implies that the effect of residence time on this process is also important on interannual timescales (cf., McDonald et al., 2004) and can possibly be distinguished from the seasonal PCO_2 effect. We are currently working on identifying the specific processes in relation to Mg sources that lead to these residence-time effects, in order to gain more confidence in the expected range of values that can be attributed to it under present-day conditions.

Note

Moondyne Cave stable isotope data is available for download from www.ncdc.noaa.gov/paleo/speleothem.html

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Temperature and precipitation records from stalagmites grown under disequilibrium conditions: A first approach

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A variety of climate archives are studied to reconstruct past variations in Earth's climate. Recently, stalagmites have come into focus, due to their long, continuous growth periods and improved dating techniques. These archives provide high-resolution stable carbon ($\delta^{13}C$) and oxygen ($\delta^{18}O$) isotope profiles, which record information about climate-related parameters at the time of growth. While the $\delta^{18}O$ of stalagmites grown under conditions of isotopic equilibrium depend on the $\delta^{18}O$ value of the drip water and the cave temperature, the interpretation of stable oxygen isotopes from stalagmites grown under disequilibrium conditions is more difficult, due to the additional influence of the drip interval (i.e., the time between two subsequent drops feeding the stalagmite) on the isotope signal. Stalagmites that demonstrably grew under disequilibrium conditions, as indicated by a positive correlation between $\delta^{13}C$ and $\delta^{18}O$ along individual growth layers (Hendy tests), have traditionally been rejected from intensive research. However, a quantitative description of disequilibrium fractionation effects might enable the reconstruction of both temperature and drip interval from isotope profiles of these stalagmites.

Here, we present a numerical model (Combined Stalagmite Model; CSM) that calculates variations in temperature and precipitation from the isotope profiles from along the growth axis and individual growth layers, as well as the age-depth relation of stalagmites grown under conditions of isotopic disequilibrium.

Model parameters

The CSM is based on the inversion and combination of previously developed models. These are growth models (Dreybrodt, 1999; Kaufmann and Dreybrodt, 2004; Mühlinghaus et al., 2007) and isotope models that calculate the isotopic enrichment of carbon and oxygen, both along the growth axis and individual growth layers, assuming an irreversible Rayleigh fractionation process (i.e., the irreversible removal of CO_2 through degassing and $CaCO_3$ through precipitation, from the solution layer on top of the stalagmite) (Mühlinghaus et al., 2007; submitted). The models include the most important parameters describing the cave system and the overlying soil, such as temperature, water supply, the isotopic composition of the drip water, the partial CO_2 pressure of the soil and the cave atmosphere, and the mixing coefficient, which describes mixing between the impinging drop and the existing solution layer.

The idea of the CSM is based on the consideration that the proxies (i.e., growth rate, $\delta^{13}C$ and $\delta^{18}O$ profiles), which are common stalagmite data sets, show different dependencies on the model parameters. Due to their high temporal resolution, the most important data sets are the carbon and oxygen profile along the growth axis. While $\delta^{13}C$ reveals a strong dependence on drip interval but only a weak one on temperature, $\delta^{18}O$ is influenced by both parameters (Mühlinghaus et al., submitted). Thus ,a drip interval signal can be extracted from the carbon isotope profile, which is in turn used to correct the oxygen isotope profile for the influence of the drip interval, in order to obtain the temperature information.

Model description

To determine the characteristics of temperature and precipitation using the CSM, some simplification of the model parameters need to be made. Firstly, all parameters (except temperature and drip interval) are assumed to stay constant over the whole growth period of the stalagmite. This is a major simplification for parameters such as the isotopic composition of the drip water, which indeed shows temporal variations due to its correlation to temperature, for instance. For other parameters, like the mixing coefficient, this approximation may only disguise small temporal variations, which can be neglected in this first approach.

This simplification enables the CSM to run with only two input variables: The mean $\delta^{13}C$ value of the drip water and the temperature at any point of time during the growth period of the stalagmite (e.g., the recent cave temperature). This yields a temperature and drip interval record of high temporal resolution. For the other parameters, such as the CO_2 partial pres-

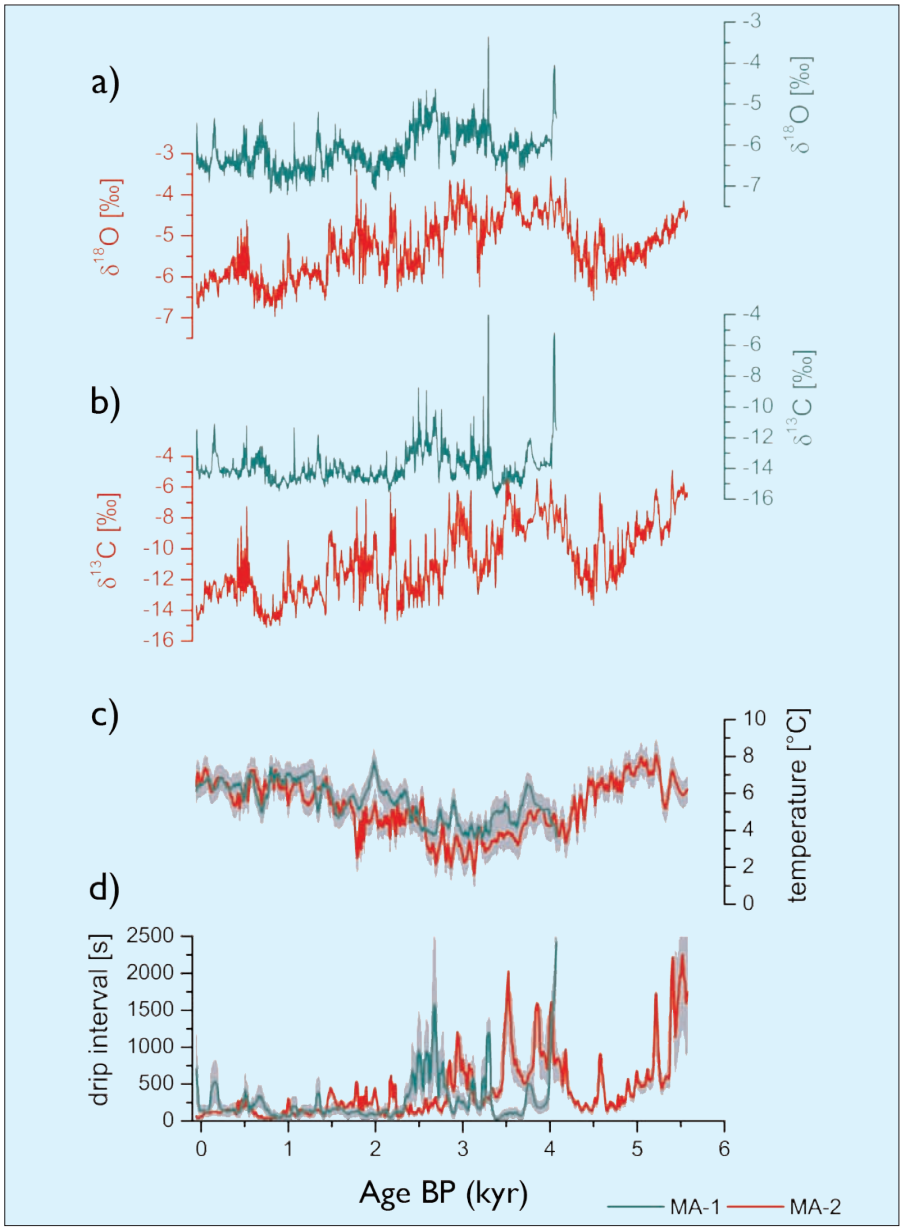

*Figure 1: (**a**) Oxygen and (**b**) Carbon profiles of stalagmites MA-1 (cyan) and MA-2 (red) from southern Chile. (**c**) Temperature and (**d**) drip interval (in seconds), reconstructed from the stalagmites, were calculated by the Combined Stalagmite Model and are smoothed by a 20-point running mean. Errors have been determined using a Monte Carlo simulation (at least 6000 iterations) and are indicated by the gray shaded area.*

sure of the soil and cave atmosphere, the mixing coefficient and the isotopic oxygen composition of the drip water mean values are determined. These represent the most likely values of the corresponding parameters during the growth period of the stalagmite. For these parameters no temporal variations can be obtained with the current model version.

Application and results

The model was applied to two stalagmites from southern Chile that were precisely dated using the Th/U method (MA-1 and MA-2, Schimpf, 2005; Kilian et al., 2006). These stalagmites grew under disequilibrium conditions in a small cave next to each other during the last 4.5 kyr BP. However, the $\delta^{13}C$ and $\delta^{18}O$ profiles along the growth axis showed a different influence of isotope fractionation under disequilibrium (Fig. 1a, b).

The drip intervals calculated from the two stalagmites show no correlation, most likely resulting from different degrees of kinetic fractionation, which may be explained by different drip sources (Fig. 1d). Hence, the amount of drip water feeding stalagmite MA-2 seems to have been limited, perhaps due to a smaller water reservoir. In the case of these stalagmites, drip intervals are directly related to precipitation, due to the sparse soil zone above the cave. Thus, large drip intervals indicate drier periods, while short drip intervals reflect wetter conditions. Although there is no significant correlation between the two reconstructed drip intervals, they follow a general trend of high water supply during the last 2.5 kyr BP, and lower water supply between 2.5 kyr and 6 kyr BP.

In contrast to the differences in the calculated drip intervals, the temperature records show a rather high correlation coefficient of $R^2 = 0.76$. This is remarkable since the correlation coefficient for the $\delta^{18}O$ profiles, which are used to extract temperature, is only $R^2 = 0.51$. This suggests that the CSM is able to extract reasonable temperature information from the given data sets. Although the two profiles reveal an offset of approx. 1°C between 1.6 kyr and 4 kyr BP (where MA-2 reconstructs lower values than MA-1), the trend and the main peaks of the two records show good agreement, particularly during the last 1.6 kyr, and between 2.5 kyr and 3.5 kyr (Fig. 1c).

The absolute temperature values, however, need to be handled with care. Stalagmite MA-1 shows a temperature variability of approx. 3-4°C and stalagmite MA-2 a variability of almost 4-5°C. These large variations may reflect regional effects on meteorology, however, one reason for this variability probably lies in the assumption of a fixed $\delta^{18}O$ value for the drip water. This value will change with temperature and variations in wind/rain trajectories. If, for instance, the $\delta^{18}O$ value of the drip water increased between 2 kyr and 4 kyr BP, the decreasing temperature trend would be attenuated and the variability damped. Thus, this temperature record might also include information about the $\delta^{18}O$ value of precipitation.

Summary

A combined stalagmite model was developed to extract drip interval and temperature variations at high temporal resolution from stalagmites grown under disequilibrium conditions. The obtained temperature signals are highly correlated over the whole growth period. The calculated drip intervals show no correlation, due to the different kinetic influence, and reflect a variable water supply. In addition, the results are insensitive to small variations of the input variables, which give confidence in the algorithm of the model and for further temperature reconstructions from stalagmites grown under such conditions.

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The origin of lamination in stalagmites from Katerloch Cave, Austria: Towards a seasonality proxy

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Macro- to microscopic layering, comparable to that in polar ice, corals and some lake and marine sediments, is common in many speleothems. Laminated stalagmites and flowstones occur from the high- and mid-latitudes down to low-latitude cave sites. In many cases, the observed lamination is demonstrably of annual origin and is an expression of seasonality (Broecker et al., 1960; Frisia et al., 2003). Locally, however, event laminae on sub-annual to multi-annual timescales are present, being stochastic or periodical in nature (e.g., Baker et al., 2002). Various material properties give rise to macro- and microscopic lamination patterns in speleothems, most notably changes in mineralogy (Railsback et al., 1994) and crystal fabric (Kendall and Broughton, 1978; Frisia et al., 2000), as well as the abundance and distribution of pores, fluid and particle inclusions or organic components. The latter, when exposed to a source of blue or ultraviolet light, cause fluorescence, which is a useful tool to study lamination patterns (e.g., Meyer et al., 2006).

Laminated speleothems have received increasing attention in recent years, as they provide very highly resolved paleoenvironmental proxy records for the terrestrial realm when combined with high-precision U-Th dating. Exploitation of their full potential for paleoscientific reconstructions requires a thorough calibration and validation of speleothem lamina proxy data, which ideally should be performed for each cave site using a combination of multi-annual cave monitoring and comparison with nearby meteorological data (e.g., Mattey et al., 2008). Constant-diameter (or candle-stick; see Fig. 1A) stalagmites are best suited for lamina-based paleoclimate studies because their morphology indicates rather constant growth, and the lamina thickness directly reflects the amount of annually precipitated carbonate, which is controlled by the prevailing environmental conditions.

In order to better understand the mechanism of lamina development and to assess the paleoenvironmental potential of annually layered stalagmites, Katerloch Cave in Austria was monitored over a period of three years. A holistic approach was used, which comprised analyses of soil air, cave air, drip water, modern calcite precipitates on artificial substrates and stalagmite samples.

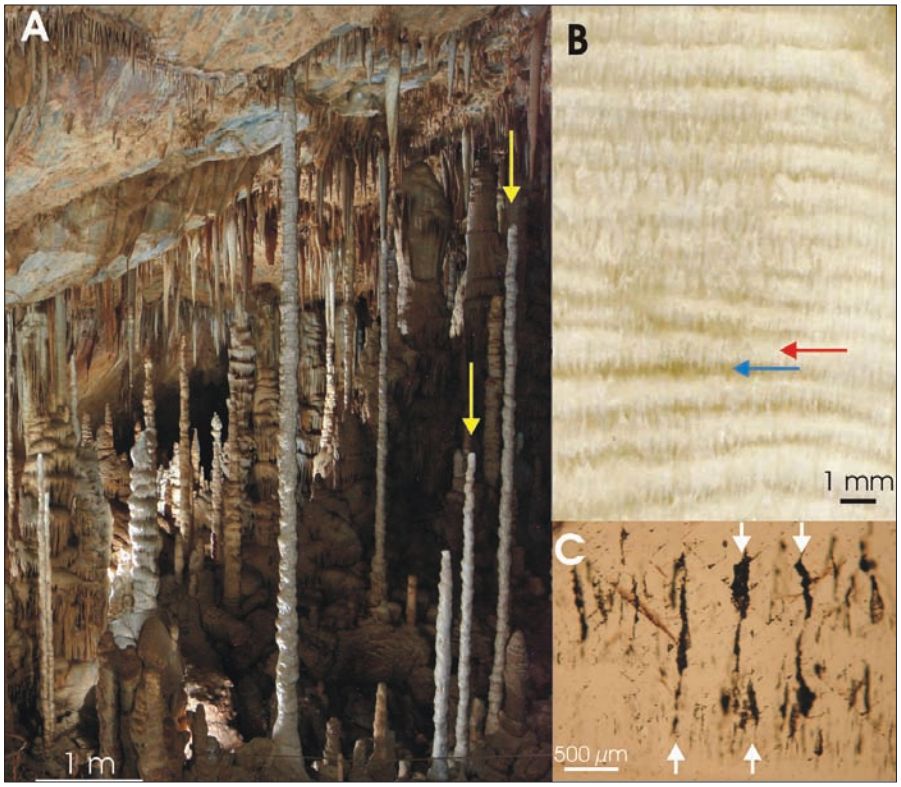

*Figure 1: (**A**) Actively dripping, fast-growing candle-stick stalagmites in Katerloch Cave (yellow arrows). (**B**) Their internal structure reveals a regular lamination consisting of white, porous laminae (red arrow) and typically thinner, translucent, dense laminae (blue arrow; slabbed section). This pattern is the expression of a seasonally changing calcite fabric, characterized by the presence and absence of elongated pore spaces (white arrows in (**C**); transmitted-light photomicrograph showing two porous laminae with relatively large pores).*

Seasonally controlled lamination in Katerloch stalagmites

Located at the south-eastern fringe of the European Alps (900 m asl), Katerloch Cave hosts abundant stalagmites of the candle-stick morphology, several of which are actively growing (Fig. 1A). Eight recovered samples of Holocene, Last Glacial period and Last Interglacial ages reveal a rather uniform macroscopic lamination pattern consisting of white, porous calcite layers and typically thinner, translucent and dense calcite layers (Fig. 1B). This regularly alternating pattern is caused by changes in the calcite fabric, in particular by the abundance of elongated pore spaces commonly containing fluid inclusions (Fig. 1C). The white, porous (inclusion-rich) laminae are further characterized by enhanced vertical growth and thinning towards the edges, whereas the translucent, dense laminae show a more constant thickness along individual growth layers. A similar type of lamination was also reported from speleothems elsewhere (Genty and Quinif, 1996; Yadava et al., 2004; Mattey et al., 2008). U-Th dating confirms the annual origin of the lamination in several Katerloch stalagmites. Stable C isotope values obtained from the stalagmites follow the lamination pattern, whereby high values typically coincide with the translucent, dense laminae and low values correlate with the white, porous laminae (Fig. 2).

The origin of this pattern is linked to the seasonally changing airflow in this cave (Boch, 2008): during the warm season the pCO_2 builds up in the cave, whereas the large temperature gradient between the outside atmosphere and the interior cave air results in increased ventilation during the cold season. Low cave air CO_2 concentrations during the latter season, as a result of enhanced exchange with the outside atmosphere, give rise to enhanced CO_2-degassing of the drip waters and high calcite supersaturation. This results in a change of the crystal fabric (from white, porous to translucent, dense) and an increase in the C isotope values in drip waters and dense calcite. In contrast, stable H and O isotope analyses reveal no seasonal variations in the drip waters, indicating discharge from a well-mixed

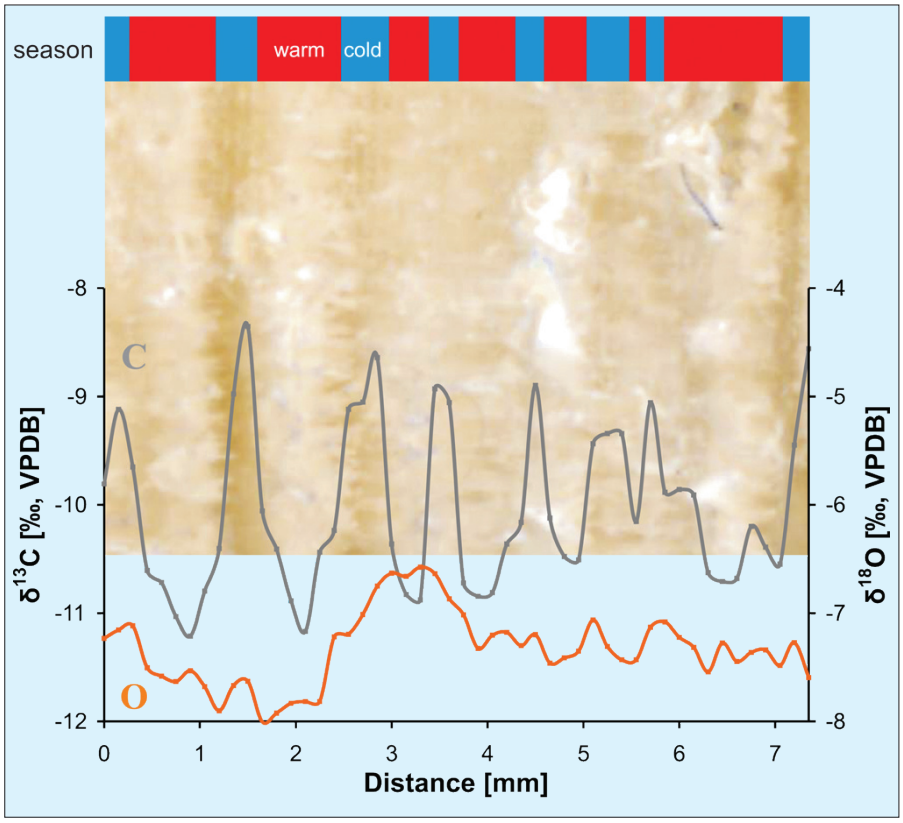

Figure 2: Example of the stable isotopic composition of a laminated Last Interglacial stalagmite from Katerloch Cave: White, porous laminae typically coincide with low calcite $\delta^{13}C$ values and cave monitoring indicates that these laminae form during the warm season when cave air and drip water C isotope values are low (sluggish air exchange, relatively high pCO_2 level). Translucent, dense laminae show high C isotope values and form during the cold season (enhanced air exchange, low pCO_2 level and kinetically controlled C isotope fractionation). The O isotopes, in contrast, are unaffected by these seasonal oscillations and record multi-annual changes.

aquifer, i.e., the mean residence time of the water in the vadose zone (the zone between the land surface and the water table) is well above one year. Likewise, O isotope data from the stalagmites do not follow the high-frequency (seasonal) variability of the C isotope data but show rather gradual, multi-annual changes (Fig. 2). In addition to the seasonal air-exchange dynamics (driven by temperature contrasts), the lamina thickness data are also a record of the amount of meteoric precipitation (or, more precisely speaking, of the net infiltration) in the catchment area. This is consistent with other studies including experimental and theoretical work, which demonstrate the strong dependence of stalagmite vertical extension rates on drip water discharge (e.g., Baker et al., 1998; Kaufmann, 2003).

In summary, the results from both multi-annual cave monitoring and studies of speleothems show that the observed lamination pattern is primarily the expression of a seasonally variable cave air composition that affects the drip water chemistry (e.g., supersaturation) and, further, the fabric of the precipitating calcite within the annual laminae. As air exchange is high and also more variable during the cold season, the lamination is primarily a proxy of the intensity of winter air circulation in the cave and, hence, of winter air temperature. For example, long and cold winters give rise to relatively thick, translucent and dense laminae showing high C isotope values. In addition, the lamina thickness also reflects the amount of precipitation that recharges the karst aquifer. Wet years promote thick laminae due to increased drip rates (enhanced ionic supply). The relative thickness, however, of summer and winter laminae will eventually also depend on the specific flow conditions at a particular drip site. This study therefore highlights the value of cave monitoring in order to understand the speleothem growth dynamics at a specific cave site, which is an indispensible requirement for exploiting lamination patterns as a quantitative seasonal climate proxy. Given the scarcity of reliable winter proxy data in most currently available terrestrial paleoclimate archives, dynamically ventilated caves such as Katerloch offer a potentially unique opportunity to fill this gap.

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Precipitation records of the last century reconstructed from annual growth-rate parameters of two Ethiopian stalagmites

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The Ethiopian highlands, located in the central section of the horn of Africa, are one of Africa's rain sensitive regions where monsoon rainfall variability plays a key role in triggering frequent droughts. The region is subject to the seasonal migration of the Inter-Tropical Convergence Zone (ITCZ) and is very sensitive to monsoon variability (Seleshi and Zanke, 2004). The main rainy season (July–September) occurs when the northward movement of the ITCZ dominates the airflow, while the spring rainy season (March–May) coincides with the southward migration of the ITCZ.

Ethiopia, being a largely agricultural country, is severely affected by even slight anomalies or irregularities of monsoon rainfall. However, inadequate understanding of historical patterns and their wider associations and causes, hamper our understanding of the climate system and the reliability of climate projections for the region. Moreover, instrumental weather records in the region are short (rarely longer than 30 years) and of poor quality, making calibration and, subsequently, forecasting of drought and flood events difficult. Long, high-resolution climate records are therefore required to investigate the nature of rainfall variability, the frequency of failure of rain periods, and the presence of decadal to centennial periodicity in climate that cannot be detected through short instrumental series.

To address this urgent problem, we have made use of recent advances in the understanding of the climatic meaning of parameters derived from well-dated speleothems (Fairchild et al., 2006). Since 2004, we have been conducting multi-proxy analysis of speleothems from the Mechara karst system located in the southeastern Ethiopian highlands (Fig. 1a), and have derived high-resolution paleohydrological (Asrat et al., 2007) and paleoclimate parameters, such as mean annual and seasonal precipitation over the last ~100 years (Baker et al., 2007). We are currently extending this research to include speleothems from the Holocene and late Quaternary. In this contribution, we present morphological and growth rate data of two Ethiopian stalagmites that have grown over the last 100 years, showing how a transfer function approach can be used to reconstruct rainfall records from these parameters.

Asfa-3 and Merc-1 Stalagmites

In a recent study (Baker et al., 2007), we combined surface climate data (from meteorological stations in the region, tested for homogeneity with the longest existing record in the country from Addis Ababa station), cave climate monitoring data (for 2004-2006), and geochemical data (cations and hydrogen, carbon and oxygen isotopes) of various surface and cave waters, with multi-parameter analysis (variations in calcite texture and stalagmite morphology, lamina width, O and C isotopes, ^{14}C activity) of two modern stalagmites (Asfa-3 and Merc-1) to reconstruct rainfall series. Here, we present only the morphological and growth rate data and their relation to precipitation data.

Asfa-3 and Merc-1 (Fig. 1b and c, respectively) were being actively dripped upon when collected from depths of about 30 m and 25 m, respectively in Rukiessa cave (average temperature: 19°C; relative humidity: 97±2%). They are both between 50 and 100 mm high, exhibiting continuous visible laminae. Merc-1 has a broader width-to-height ratio than Asfa-3, while observed drip rates onto Asfa-3 were much slower than onto Merc-1, suggesting that Asfa-3 was predominantly fed by stored groundwater, whereas Merc-1 had a greater proportion of event water.

Lamina counting conducted on these stalagmites (following the protocol of Tan et al., 2006) revealed a total of 94 laminae in Asfa-3 and 111 laminae in Merc-1. Lamina widths typically varied between 0.2 and 0.4 mm (Fig. 1d), with some thicker laminae in periods of colorless calcite deposition in Asfa-3 (WC1 and WC2 in Figure 1). Laminae deposition was continuous, and the lamina width matches that predicted for annual accumulation based on the temperature of the cave, atmospheric PCO_2 and calcium and magnesium concentrations of the drip waters. Assuming annual deposition, Asfa-3 and Merc-1 were deposited from 1910 AD and 1894 AD, respectively, until sampled in 2004.

Growth rate as a rainfall proxy

The two stalagmites have similar mean growth rates but differing growth rate variability. Asfa-3 has long periods of very stable growth rates, interspersed with jumps to short periods of fast growth. Mer-1 has a more constant growth rate but with greater interannual variability. We correlated the growth rate series (monthly, seasonal and annual means) of both stalagmites against the instrumental rainfall series, as well as appropriate linear 'climate transfer functions' that smoothed previous years' monthly, seasonal and annual rainfall in a manner that potentially reflects the mixing of waters of different ages within the limestone aquifer. Correlations with the amount of rainfall in the year of calcite deposition were weak and statistically insignificant (Fig. 2a and b). Seasonal and smoothed correlations were stronger, especially for Asfa-3, for which decadal averaged growth rate correlated with June–August rainfall (r = 0.50). Applying the climate transfer function demonstrated that, for this sample, a simple decadal average had a correlation that was statistically as strong as a more complex climate transfer function. The reconstructed summer

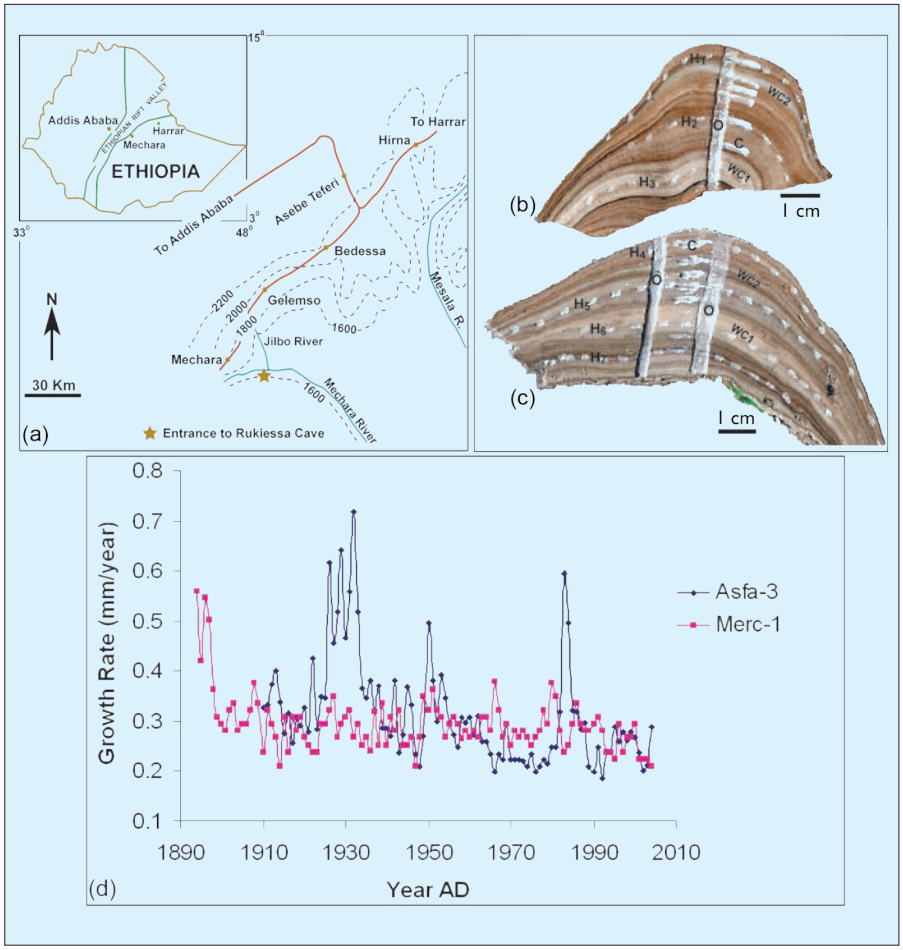

*Figure 1: **a**) Location of sample site: Meteorological stations are located at Mechara, Gelemso, and Bedessa; **b**) and **c**) Stalagmites Asfa-3 and Merc-1, respectively. Also shown are isotope profiles (vertical suites of samples labeled O; duplicate analyses on Merc-1); and horizontal suites of samples labeled H_{1-7}; and samples for ^{14}C analyses labelled C; see Baker et al., 2007). WC and WC2 refer to white (colorless) calcite layers that correlate between the two stalagmites; **d**) Time series of growth rate for Asfa-3 (blue diamonds) and Merc-1 (pink squares). Figure modified from Baker et al., 2007.*

rainfall, based on a transfer function of 10% 'event' water (May-July rainfall of that year) and 90% 'storage' water (an average of May-July rainfall of the previous nine years), is presented in Figure 2c.

In contrast, only weak correlations between growth rate and precipitation were observed for Merc-1. As this sample was fed by a more variable drip rate, as inferred from its morphology and cave observations, this weak correlation suggests that greater drip water flow variability confounds any strong correlations between lamina width and surface climate. Applying the same transfer functions as for Asfa-3, it can be seen that the function leading to the strongest correlations has a greater proportion of relatively short residence-time water (20% event water, 80% stored water of <5 years). The strongest correlation was with July rainfall, again reflecting increased water availability, as well as probable increased soil CO_2 but the correlation coefficient was only 0.30 and therefore growth rate in this stalagmite is of little use for rainfall reconstruction.

Conclusions

Our recent studies (Baker et al., 2007; Asrat et al., 2007) show that multiple parameters (e.g., oxygen isotopes, growth rate) from individual stalagmites have different climate sensitivities and would yield contrasting proxy-climate forcing functions. Multi-parameter, multi-proxy approaches are essential when using stalagmites to reconstruct climate, due to the individual nature of the link between surface climate and cave stalagmite, resulting from the heterogeneity and complexity of karst groundwater flow (Fairchild et al., 2006). From their morphology, it would be expected that Merc-1 responded more to high frequency ('event') precipitation, while Asfa-3 to low frequency ('storage') climate. Despite this, it is Asfa-3 that has a greater range of lamina width due to short periods of fast growth associated with changes in calcite texture. Different parameters (e.g., O and C isotopes, growth rate) also respond to different seasons, and in some cases the same parameter responds to different months or seasons via different forcing mechanisms (e.g., rainfall seasonality or amount, or non-equilibrium factors). For our stalagmites, growth rate is among the various parameters that show the highest correlations with monthly or seasonal mean rainfall. Therefore, we propose that growth rate, as part of multi-parameter, multi-proxy analyses of long time series of speleothems, offers a most promising approach to characterizing the variability of rainfall in this region, where the population largely depends on rain-fed agriculture for survival.

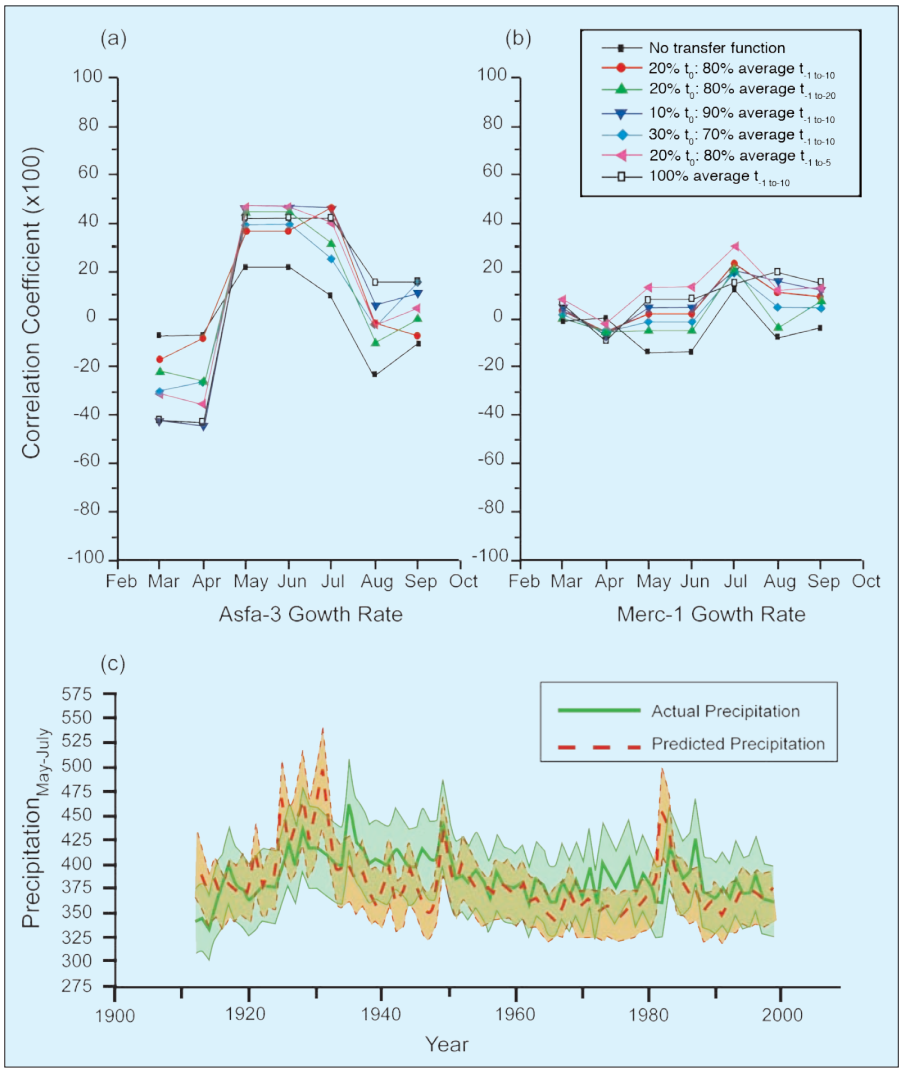

*Figure 2: Correlation coefficients between monthly rainfall and stalagmite parameters: **a**) Asfa-3 growth rate and **b**) Merc-1 growth rate, with various transfer functions of the preceding years' rainfall: e.g., the simplest 'no transfer function' option, which is a correlation with that years' rainfall in the given month against growth rate (black filled square); correlation of the average of last 10 years rainfall for a given month against growth rate (black open square); a transfer function that is weighted 10% on the given month's rainfall that year ('event' water) and 90% on the rainfall of the preceding nine years ('storage' water) for the given month, correlated against growth rate (blue inverted triangle). Correlation coefficients are shown x100 for clarity; **c**) May–July rainfall reconstructed from Asfa-3 growth rate, applying a transfer function of 10% 'event' water (May-July rainfall of that year) and 90% 'storage' water (an average of May-July rainfall of the previous nine years); errors on reconstructed precipitation are based on regression errors, errors on instrumental precipitation reflect differences between local and Addis Ababa series. Figure modified from Baker et al. (2007).*

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The use of stalagmite geochemistry to detect past volcanic eruptions and their environmental impacts

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Our knowledge of past volcanic eruptions and their climatic impact is still far from complete. Establishing a clear link between volcanism and anomalous tree ring growth is difficult (Pearson et al., 2005), and ice core chemical signals do not match the historical record of eruptions or do not allow recognition of distal eruptions (Zielinski, 2000; Oppenheimer, 2003). Evidence for a causal connection between past volcanism and climate anomalies relies on the robust correlation between climate proxies and chemical fingerprinting of past eruptions within the same well-dated archive. Stalagmite geochemistry has the potential for providing precisely dated and spatially well-distributed records of past volcanic eruptions and of their environmental impacts (Frisia et al., 2005).

The chemical signal that is the most likely proxy of past volcanic eruptions is Sulfur (S) concentration variability in the carbonate. The S emitted from volcanic eruptions to the atmosphere oxidizes to sulfate and reaches the soil as wet and dry deposition, or as ions adsorbed onto ash particles. The propagation of the atmospheric sulfate signal from the surface to the cave depends on the transmissivity of the karst aquifer (i.e., the hydraulic conductivity multiplied by the thickness of the aquifer). Caves cut in fissured limestone overlain by relatively thin soil with large interconnected pores ensure fast transmission, and in a few days or a few months after the eruption, the growing calcite will form from waters with a higher-than-background concentration of sulfate. Sulfur in the form of sulfate (S-sulfate) will be incorporated either in the calcite crystal lattice, or as micro-inclusions in the stalagmite. A volcanically induced increase in local acidity levels may also mobilize S adsorbed onto ash or associated with wet deposition and make it available to the ecosystem. This may result in a lagged response in the speleothem compared to the atmosphere, associated with the mineralization of sulfate in organic form followed by re-oxidation to sulfate. Volcanic eruptions are, therefore, likely to be recorded in stalagmites by both a narrow S-sulfate peak coinciding with the year of a large eruption, or by a broader S-sulfate peak in the years immediately following the eruption (Frisia et al., 2005; Wynn et al., 2008).

S concentration in speleothems: Analytical methods

In order to resolve S peaks related to volcanic eruptions, a very high spatial resolution, low limit of detection (ppm to ppb) and precise correlation between growth laminae (microstratigraphy) and geochemical signals must be achieved. Synchrotron radiation based micro X-ray fluorescence (SR-μXRF) microscopy coupled with X-ray absorption near-edge spectrometry allow for high spatial resolution, low limits of detection, assessment of the molecular environment of S and precise location of the analyzed regions (Borsato et al., 2007). This technique appears to be best suited for annually resolved sulfate variability in stalagmites. The use of SR-μXRF to detect S-sulfate in stalagmites from different geographic locations highlights both direct and delayed responses of the ecosystem to the emission of S in the atmosphere after volcanic eruptions.

Tambora and Krakatau recorded in a stalagmite from N. Italy

Annually laminated stalagmites from Grotta di Ernesto, located at ca. 1160 m asl in NE Italy, show distinct S peaks in the years 1815–1816, 1884–1888, and 1947 (Fig. 1). The age model is based on annual lamina counting and radiometric dating. Quantification of the S content from synchrotron data (counts per second; cps) was carried out by calibration with five dissolved specimens from the same samples, analyzed using high-mass resolution inductively coupled mass spectrometry (ICP-MS; Borsato et al., 2007).

The 1815 to 1816 AD increase in S-sulfate from ca. 15 to ca. 30 ppm coincides with the eruption of Mount Tambora in Indonesia (April 1815). However, a connection between volcanism, stalagmite growth anomaly and climate cannot be attested as the Tambora eruption falls within a Little Ice Age portion of the stalagmite that is characterized by very thin annual laminae (2–10 μm) indicative of low mean annual temperatures (Frisia et al., 2003). In 1884–1888, sulfate peaked to values between ca. 10 and 40 ppm, then returned rapidly (within ca. 2 years) to values below 10 ppm. This peak coincides with the eruption of Mount Krakatau (Indonesia) in August 1883, and falls within a portion of the stalagmite where lamina thickness averages 100 μm. The thickness decreases abruptly throughout the 1884-1888 time span to less than 50 μm, which supports the causal connection between a volcanic eruption, sulfate aerosol and short-lived cooling. The sulfate peak in the 1947 lamina may correspond to the eruption

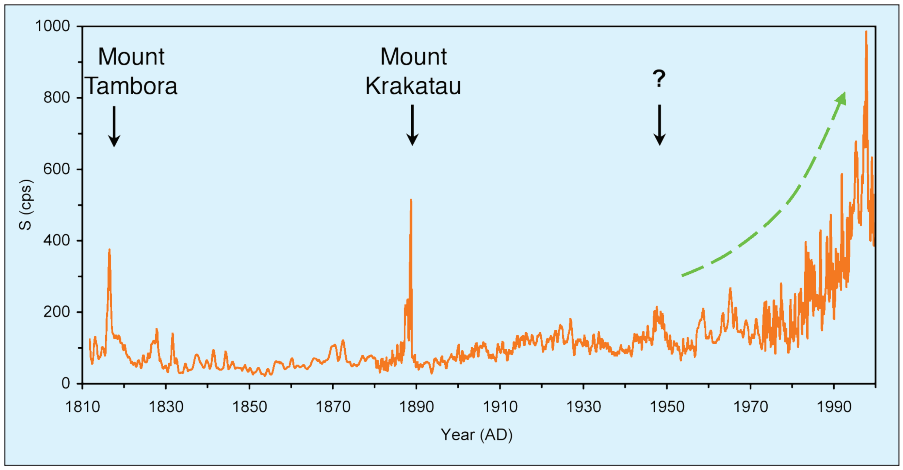

Figure 1: Results of the SR-μXRF analyses of S-sulfate in the 1810–2000 AD portion of a stalagmite from Grotta di Ernesto (N. Italy). Arrows indicate that sulfate concentration peaked in the years 1815–1816, 1884–1888, and 1947. These peaks can be ascribed to atmospheric S load increases following the Tambora, Krakatau and (possibly) Hekla eruptions, respectively. The pronounced increasing trend in S-sulfate concentration (counts per second; cps) after the year 1960 is due to anthropogenic sulfate emissions (Frisia et al., 2005) and masks any more recent volcanic-related S-sulfate peaks.

of Hekla (March 1947) in Iceland and is followed by a short-lived diminution in lamina thickness from the average of 100 µm to ca. 80 µm. Since 1960, the anthropogenic S-sulfate signal is overwhelming and masks possible peaks related to more recent volcanism.

The Santorini eruption recorded in a N. Turkey stalagmite

The stalagmite of Grotta di Ernesto shows, through a decrease in lamina thickness and variability in calcite sulfate concentration, a rapid climatic response (cooling) to the rapid, short-lived stratospheric sulfate aerosol emissions generated by the aforementioned volcanic eruptions. In contrast, a stalagmite from Sofular Cave in western Anatolia, Turkey, shows a delayed response (Fig. 2). The S-sulfate investigation of this sample focused on a well-dated portion of the stalagmite encompassing the Santorini eruption, from ca. 3350 –3800 years before 2008. The Santorini eruption, which is believed to have spread a huge layer of ash over the Eastern Mediterranean region, has been recently dated at 1627-1600 BC (3633-3608 BP) on the basis of a wiggle-matched sequence of radiocarbon-dated olive tree ring groups (Friedrich et al., 2006).

A distinct S-sulfate peak, here reported in cps as the calibration has yet to be performed, occurs between 3550 and 3600 BP. This indicates that increased S levels had been available for mobilization from the surface to the subsurface for several decades. The S peak detected in the Sofular stalagmite could thus be related to the environmental acidity increase following the eruption. The question remains about timing of the Santorini eruption. The broad S-sulfate peak immediately follows a distinctive, short-lived peak in $\delta^{13}C$, which shifts to more positive values at ca. 3610 BP and reaches its maximum (of ca. 0.8‰) at ca. 3600 BP. A preliminary interpretation is that the $\delta^{13}C$ peak may be related to vegetation stress following the eruption. Based on the interpretation of millennial- to secular-scale $\delta^{13}C$ changes recorded in the stalagmite from Sofular Cave, this stress is most likely related to a dry spell (Badertscher et al., in prep.). The timing of the Santorini eruption in the Sofular record is thus marked by a rapid, short-lived (few years) $\delta^{13}C$ shift to more positive values (ca. 3010 BP). Given the uncertainties related to different dating methods, the dating of the Santorini eruption from the Sofular record is similar to that obtained by Friedrich et al. (2006), and supports an eruption on Santorini in the late 17th or early 16th century BC.

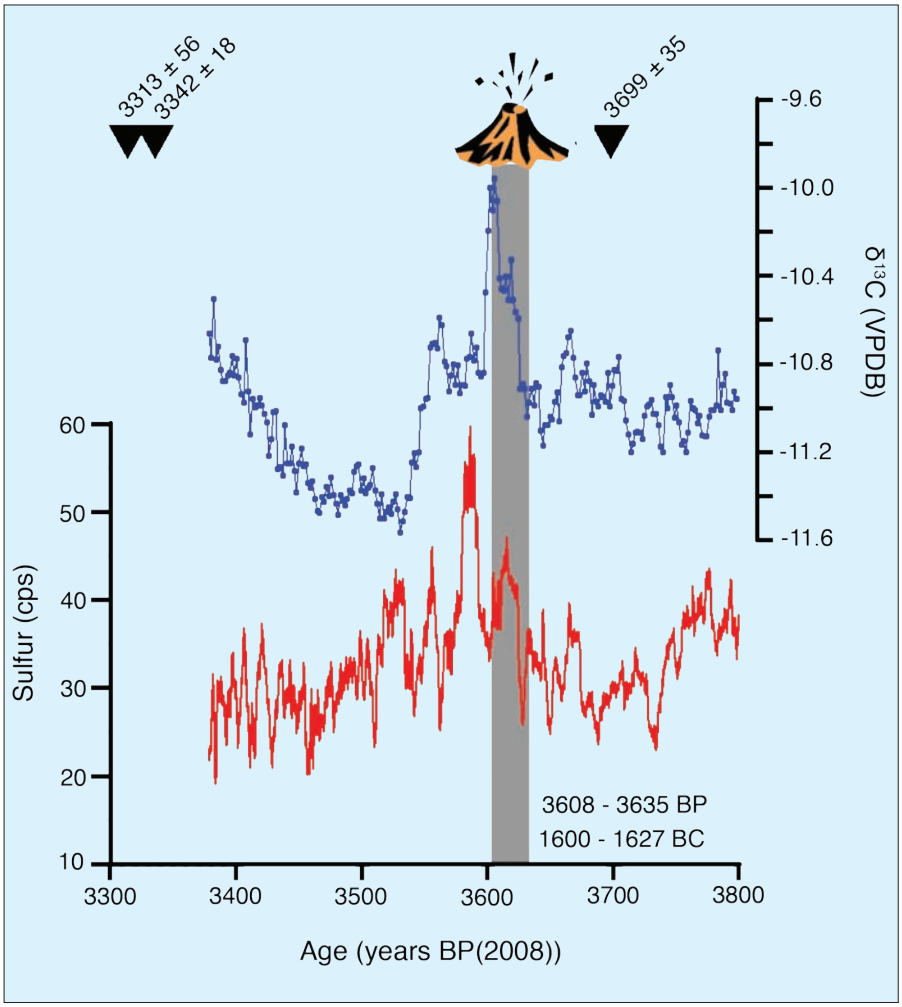

Figure 2: The effects of the Santorini eruption recorded by SR-µXRF analyses of S-sulfate in the ca. 3350–3800 BP portion of the Sofular stalagmite from Anatolia (Turkey). Note that the peak in S concentration follows a rapid shift of the inverted $\delta^{13}C$ curve to more positive values, which can be related to vegetation stress. The gray shaded area marks the timing of the Santorini eruption as given by Friedrich et al., 2006.

Future work

High-resolution pilot studies of S-sulfate concentration of precisely dated stalagmites indicate that there is a high potential for studying the timing and climatic/environmental impact of massive volcanic eruptions.

The ability to extract a causal effect between volcanic eruptions and atmospheric cooling relies on the unequivocal interpretation of the climate proxy data variability, which coincides with S-sulfate peaks in well-dated stalagmites. In the case of the stalagmite from Grotta di Ernesto, for example, the effects of the Tambora eruption cannot be distinguished from a general cooling trend that characterized the late Little Ice Age in the Alps. The Sofular record highlights that the timing of an eruption may not exactly coincide with the onset of a S-sulfate peak, probably because of the delayed rise in acidity levels following the eruption. The use of stalagmites for the evaluation of global and regional effects of major volcanic eruptions thus has some issues to overcome. However, we believe that a multi-proxy, multi-disciplinary approach will solve some of the problems concerning the timing and effects of past volcanism.

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Monitoring environmental pollution using a stalagmite from Hungary

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Speleothems are well known paleoclimate archives but their potential for monitoring environmental pollution has not been fully explored (e.g., Frisia et al., 2005; Fairchild et al., 2006; Borsato et al., 2007; Perrette et al., 2008). This study deals with an actively growing stalagmite whose trace-element concentration suggests anthropogenic contamination, rather then natural forcing. Our objective was to determine the possible effect of the four-decade-long uranium (U) ore mining activity on the environment, as recorded by a cave deposit.

The Trio Cave (46.7°N, 18.9°E) is located in the western part of the Mecsek Mountains (S. Hungary) at the base of the Szuado Valley, approx. 1.5 km east of the nearest entrance and air shaft of the Mecsek uranium mine (mine-pit no. IV). The karst system developed in the Triassic Lapis Limestone and the cave is ca. 200 m long. There is one artificially enlarged entrance (opened once in 1969 and finally in 1997), which intermittently acts as a sinkhole for the Orfű creek, with a catchment area of 3.5 km^2. A stalagmite located about 150 m into the cave was drilled and the 42 cm-long core was investigated for stable isotope and trace element composition using continuous-flow mass spectrometry and laser-ablation inductively coupled plasma mass spectrometry (ICP-MS), respectively (Siklósy et al., 2007).

The uppermost ca. 3 cm of the core were selected for this study (Fig. 1a), which represents the last few hundred years (based on an estimated growth rate of 0.05 mm/year by ^{230}Th age dating of older parts of the core).

Geochemical results

The older part of the 3-cm section is characterized by systematic co-variations between U and Phosphorus (P) concentrations, and P concentrations and δ^{13}C values, which can be related to soil activity (Figs. 1b, 1c). Silica (Si), aluminium (Al), thorium (Th) are positively correlated, and their concentrations increased strongly in the uppermost ca. 3 mm, due to the elevated detrital content of the stalagmite (Fig. 1d). With increased detrital input, the U concentration also increased radically (especially in the topmost 1.3 mm), starting from a background value of 0.2-0.3 ppm, increasing gradually to about 2 ppm, followed by constant values for about 0.5 mm, and then declining to about 1.5 ppm (Figs. 2a, and 2d).

The increase in U concentration coincided with a significant decrease in $\delta^{234}U_{initial}$ values suggesting contribution from a U source different from the natural weathering input (Fig. 2a). This is also supported by a change in the P/U ratio and much weaker correlation of the U concentrations with P (Fig. 2b) in the U-enriched section of the stalagmite ("mining-period"). According to the average growth rate of the stalagmite, this period represents the last 30-50 years.

Origin of pollution

Possible U sources are fertilizers used in agriculture, industrial pollution (e.g., ash from coal-heated power plants), and U ore mining in the vicinity of the cave site. Fertilizers may contain considerable amounts of U (10-360 ppm, Hamato et al., 1995), however, the lack of agriculture in the valley and the region around the cave, consistent with the absence of high P concentrations (Fig. 1b) in the topmost section of the speleothem, strongly argues that fertilizers are not the source of the high U values. Coal-derived pollutants can likewise be exluded as a source for the high U concentrations in the stalagmite, as the only major industrial city and power plant in the southern part of Hungary (Pécs) is approx. 12 km from the cave. In addition, the dominating wind currents flow in the opposite direction and coal ash is rich in metals (Co, V, Zn, etc.) that were not enriched in the stalagmite.

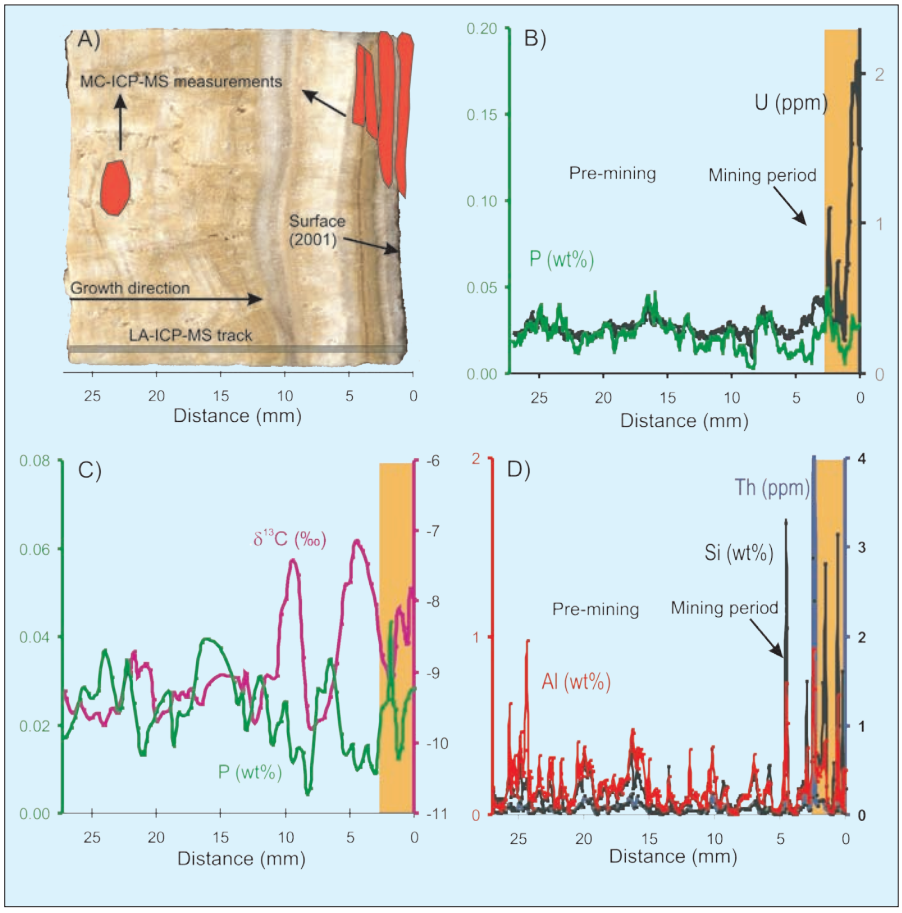

*Figure 1: **a**) Image of the sub-recent part of Trio stalagmite core (southern Hungary). Gray bar shows the position of the trace-element profile. Position of the MC-ICP-MS data reported in Fig. 2b is indicated by the red regions. **b**) Phosphorus (P) vs. Uranium (U) contents; **c**) Stable C isotope and P concentrations of the stalagmite; **d**) Th, Al and SiO_2 content. All data are plotted against the distance from the active surface of the stalagmite (time of collection: 2001). Orange shaded area represents approx. period of mining activity.*

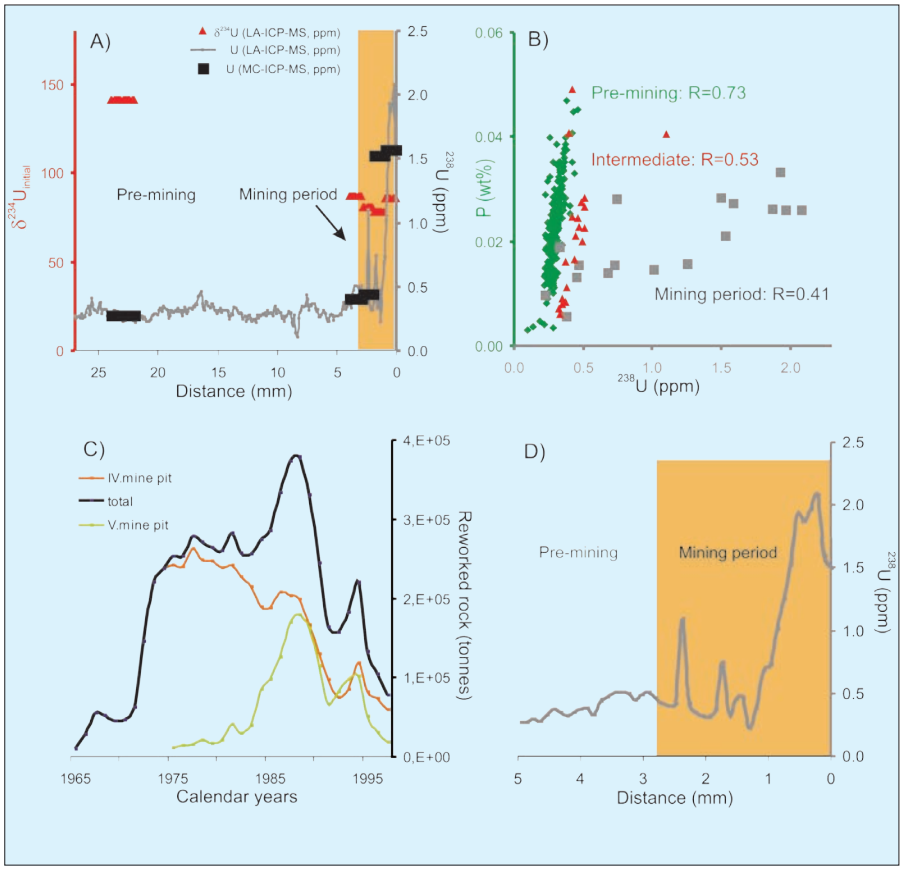

*Figure 2: **a**) Uranium (^{238}U) concentration (measured independently by Laser Ablation- and MC-ICP-MS) and $\delta^{234}U$ values of the Trio stalagmite. Data are plotted against the distance from the active surface of the stalagmite (time of collection: 2001). **b**) Correlation between P and U for assumed pre-mining and mining period of the studied sample. Intermediate period is also indicated. Note the reduction of the R-values and the change in the P/U ratio. **c**) History of U ore production, as shown by the mass of reworked U-bearing sandstone. **d**) ^{238}U concentration for the topmost 5 mm of the stalagmite representing approx. the last 100 years of growth. Orange shaded area represents approx. period of mining activity.*

The third possible source of U is the 40-year-old Mecsek uranium mine, which produces ca. 500 tons of U concentrate per year and has reworked millions of tons of solid material (Bánik et al., 2002). U production in S. Hungary started in 1957 and was expanded closer to the cave site in 1969 (Fig. 2c), covering a mining plot area of ca. 65 km^2. The deep-level ore production ended in 1997 and remediation of the mine site has since been completed.

The possible relationship between high U concentration in the most recent stalagmite section and U ore mining was investigated by the comparison of the measured U concentration (Fig. 2d) and the mining intensity (Fig. 2c). The deep level of the mine was ventilated by air shafts, therefore the total mass of the exploited U-bearing sandstone and the dust released by the mine's shafts are directly correlated. The water catchment area of the Trio Cave is situated at a higher elevation than the closest mine pit (no. IV), which excludes the possibility that U was fluvially transported from the mining area to the cave. Eolian transport of dust released from air shafts and subsequent dust deposition, however, have been identified by previous studies of soil, plants and leaves (Mecsekérc Ltd., unpublished reports). Although the higher proportion of washed-in particles can be clearly followed by the elevated detrital content of the stalagmite (Fig. 1a), Fig. 1d clearly illustrates that the exceptionally high Si, Al and Th peaks coincide with both dark and U-enriched layers (Fig. 1b). The gradual increase in U concentration to the highest values (0.2 to 2.1 ppm) may be due to the weathering of the mine-derived dust that settled in the catchment area. Plotting of P vs. U can be used to separate the pre-mining and the mining section (at ca. 1.7 mm from the top of the sample), as they are characterized by different elemental ratios (Fig. 2b).

Mobility and transportation of U

Considering the mobility of the U(VI) uranyl ion in oxidized environments, the major part of polluting U is not expected to accumulate in the soil zone but would rather be transported into the vadose zone (zone between the ground surface and the water table). Natural aquifer systems always contain organic macromolecules (e.g., wood degradation products, humic acids) originating mostly from the soil-zone solutions. In the case of U(VI), humic acid exhibits a clear mobilizing effect (Sachs et al., 2005), therefore acting as an aid for the transport of U into the karst system. In solutions of near-neutral pH, the presence of carbonate ions induces the formation of highly soluble uranyl-carbonate complexes, $UO_2(CO_3)_3$ (Finch and Murakami, 1999), minimizing adsorption to soil particles and enhancing U mobility (Elless and Lee, 1998). Finally, U is removed from seepage water by precipitation of carbonates.

To summarize, the relationship between changes in the U content and $\delta^{234}U$ values of the speleothem, and the history of U ore production and pollution strongly suggest a causal link. Our study therefore demonstrates that speleothems can reliably record environmental pollution signals, a hitherto largely unexplored source of information.

Note

All data are available for download from www.geokemia.hu/~siklosy/index.html

Acknowledgements

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Timing of the 8.2-kyr event in a stalagmite from Northern Oman

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The 8.2-kyr event (Alley et al., 1997) is the most outstanding Holocene cooling episode in Greenland ice cores. This distinct event was triggered by a large outburst of meltwater from glacial Lake Agassiz through the Hudson strait into the North Atlantic at around 8.470 ± 0.300 kyr BP (Barber et al., 1999). The resultant freshening of the North Atlantic led to a weakening of the thermohaline circulation and widespread cooling in the North Atlantic regions (Alley and Ágústsdóttir, 2005). Furthermore, low levels of atmospheric methane indicate a drying of the tropics (Alley et al., 1997).

However, in many paleoclimate time series, unambiguous identification of the 8.2-kyr cold event is hindered by considerable age uncertainties and low temporal resolution. Furthermore, this event seems to be superimposed on a longer-term cold and dry period that lasted from around 8.6 to 8 kyr BP (Rohling et al., 2005). As a result, climatic anomalies occurring at approx. 8.2 kyr BP have often been wrongly ascribed to the 8.2-kyr event (Rohling et al., 2005).

Overall, the timing and geographical extent of this event are still a matter of debate, as precisely dated paleoclimate records are rare, particularly throughout the tropics (Alley and Ágústsdóttir, 2005). Whether anomalies around 8.2 kyr BP observed in paleoclimate archives are really representative of one synchronous event, different events or a one-time transgressive event remains largely unanswered. In order to resolve this puzzle surrounding the 8.2-kyr event, precise dating becomes critical. Stalagmites have the potential to contribute high-quality time series due to their precise and absolute chronologies. Here, we present a precisely-dated stalagmite from N. Oman to document the response of the Indian summer monsoon to the 8.2-kyr event in great detail.

Stalagmite H14

Stalagmite H14 (Fig. 1a) was collected from Hoti Cave in N. Oman (23°05'N, 57°21'E). Present-day climate in this area is arid and most of the total annual rainfall occurs during winter and spring, and limited rainfall occurs during summer by local thunderstorms (Fleitmann et al., 2007). During the early- to mid-Holocene, however, N. Oman was under the direct influence of the Indian summer monsoon and most of the annual precipitation occurred during summer (Burns et al., 1998; Fleitmann et al., 2003a). High monsoon precipitation is indicated by abundant early- to mid-Holocene stalagmites, whereas today, only a few and comparably small stalagmites are actively growing.

Stalagmite H14 shows well-developed annual bands over its entire length. Their thickness varies between 0.1 and 0.6 mm around a mean of 0.3 mm. The chronology of H14 is based on new ^{230}Th dating techniques, which allowed us to obtain 15 ^{230}Th dates with unprecedented small age errors of 15 to 45 years (2σ) for the interval between 7.65 and 8.6 kyr BP (Fig. 1b). Precise ^{230}Th dating is also facilitated by high uranium contents of ~1.5 ppm and very low detrital contamination (^{230}Th/^{232}Th atomic ratios are well above 1000). All ^{230}Th ages are in stratigraphic order, within the age uncertainties, and in very good agreement with annual band counts (Fig. 1b). This indicates that the observed sub-mm bands are indeed annual. The combination of accurate ^{230}Th dates and annual band counts allow us to construct a precise age model that is far more precise than linear interpolation between individual ^{230}Th dates.

A total of 388 oxygen isotope (δ^{18}O) measurements were made at an average temporal resolution of 2 years. Previous studies have shown that variations in δ^{18}O in early- to mid-Holocene stalagmites from Oman are primarily driven by changes in the amount of monsoon precipitation (Fleitmann et al., 2003; 2004), with more negative δ^{18}O values being associated with higher precipitation and vice versa (so-called "amount effect"). This assumption is supported by the good agreement between δ^{18}O and annual band thickness (ABT). Thicker (thinner) annual bands, indicative of greater drip-water supply at times of enhanced precipitation, coincide with more negative (positive) δ^{18}O values (Fig. 2).

Timing of the 8.2-kyr event

The depth versus age plot (Fig. 1b) indicates an interruption in stalagmite deposition at 8.193 ± 0.015 kyr BP (age error was taken from the closest ^{230}Th date at 8.205 ± 0.015 kyr BP) as a result of a cessation in drip water supply, due to a significant drop in monsoon precipitation. The most likely cause for the reduction in monsoon precipitation is that strong winter cooling and enhanced Eurasian snow cover weak-

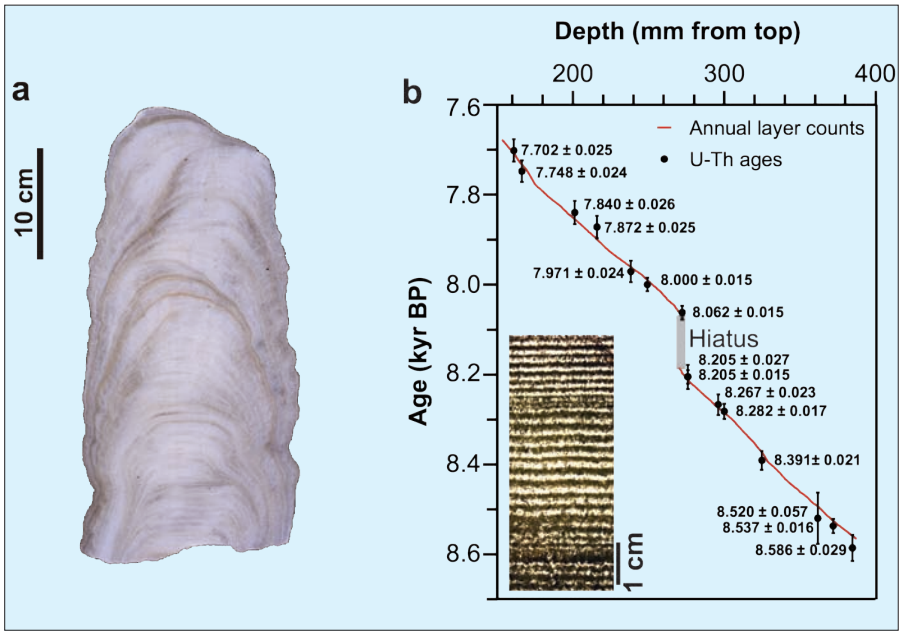

*Figure 1: **a**) Image of stalagmite H14 from Northern Oman; **b**) Depth versus age plot for Stalagmite H14. Black dots with error bars denote ^{230}Th dates, red line shows annual band counts. Small inserted image shows well-developed annual bands, which are couplets of a dense (white) and more porous (black) calcite layers.*

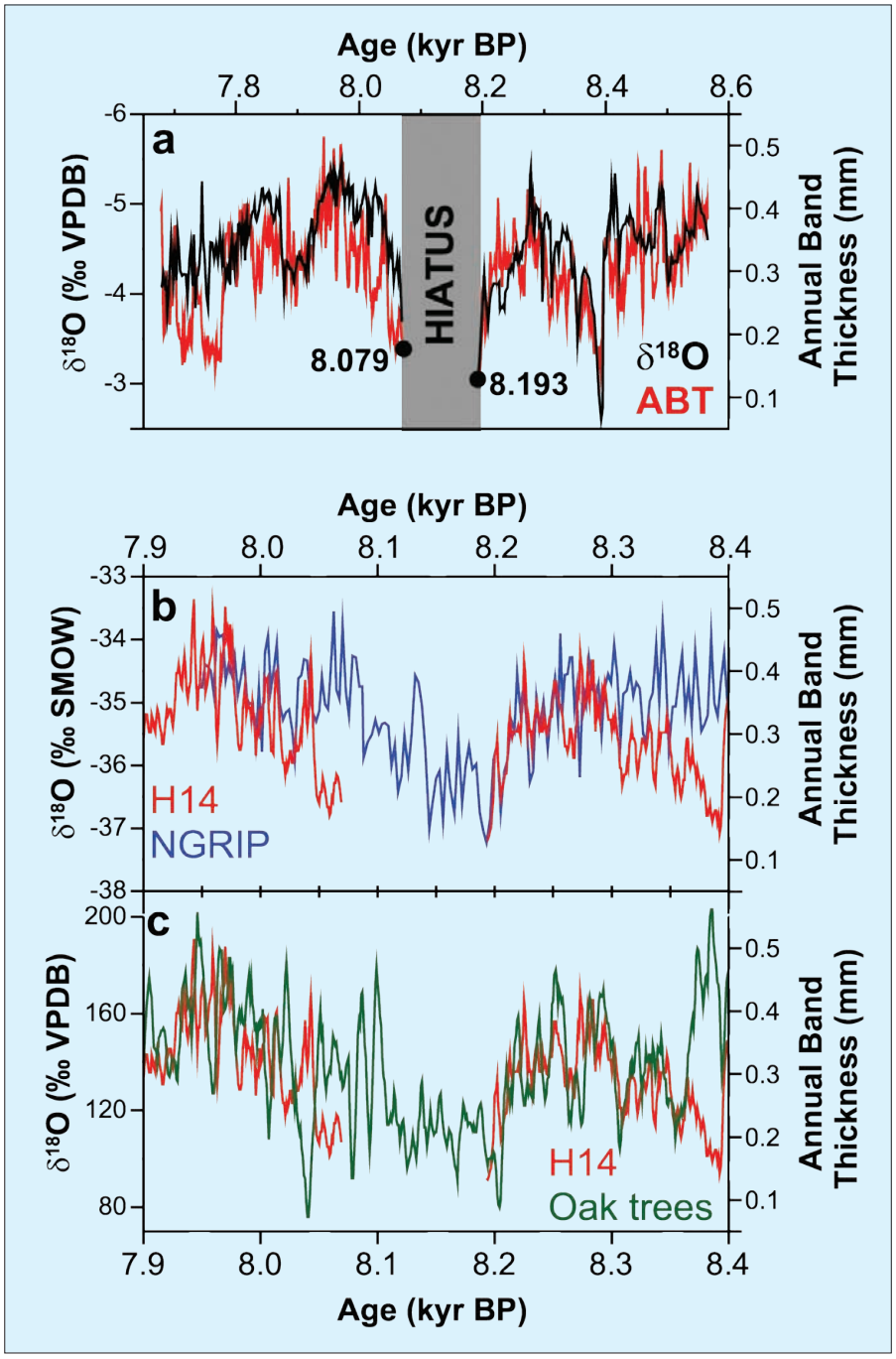

*Figure 2: **a**) Comparison between $\delta^{18}O$ and annual band thickness (ABT) in stalagmite H14; **b**) Comparison between stalagmite H14 ABT profile and NGRIP $\delta^{18}O$ profile (Rasmussen et al., 2006); **c**) Comparison between stalagmite H14 ABT profile and German oak tree-ring chronology (Spurk et al., 2002).*

ened monsoon circulation by reducing the land-sea thermal contrast that drives the Indian summer monsoon (e.g., Fleitmann et al., 2003b, 2007). The timing of the decline in monsoon precipitation in N. Oman is in excellent agreement with the rapid temperature drop of approximately 5-8°C at 8.210 ± 0.047 kyr BP (based on the most recent GICC05 chronology, Rasmussen et al., 2006) in ice cores from Greenland. Additionally, the annually-precise German oak tree-ring record shows that a shift towards cooler and drier climatic conditions occurs at 8.204 kyr BP (Spurk et al., 2002). This date is in good agreement with the abrupt positive shift in $\delta^{18}O$ and decrease in ABT in stalagmite H14 from Oman at around 8.202 ± 0.016 kyr BP (Fig. 2c). Further support derives from other precisely dated stalagmites from Dongge Cave (China) and Padre Cave (Brasil), which place the onset of the 8.2 kyr event at 8.230 ± 0.025 and 8.203 ± 0.015 kyr BP, respectively (Cheng et al., in prep.). The independently ^{230}Th dated stalagmite H14 thus confirms the most recent ice core (GICC05 timescale) and oak tree-ring chronologies, and suggests that the 8.2-kyr event is indeed synchronous across the northern hemisphere. We note that the mean calibrated ^{14}C age of 8.470 ± 0.300 kyr BP for the catastrophic meltwater outburst from Lakes Agassiz and Ojibway (Barber et al., 1999) is somewhat older but this offset can be explained by sediment mixing and considerable uncertainties in local marine reservoir age estimates in the Hudson Strait (Teller and Leverington, 2004). Providing a precise estimate for the duration of the 8.2-kyr cold event is somewhat difficult as its termination seems to be rather gradual. Nevertheless, the hiatus in stalagmite H14 lasted from 8.193 ± 0.015 to 8.079 ± 0.015 kyr BP, similar to the duration recorded in ice cores from Greenland (Fig. 2b).

Conclusion

Our results suggest a commencing date for the 8.2-kyr event of around 8.202 ± 0.015 kyr BP in N. Oman, which is in good agreement with dates provided by Greenland ice cores and German oak tree-ring record. The 8.2-kyr event thus occurred synchronously within the dating errors, revealing the hemispheric extent and atmospheric transmission of the event from the North Atlantic to the tropics. This example from Oman demonstrates that an important strength of stalagmite-based paleoclimate reconstructions is their superior chronology, which enables identification of abrupt climatic events with very high precision.

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Millennial-scale climate variability recorded in Brazilian speleothems

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There is steadily increasing interest in obtaining records of millennial-scale climate events in speleothems (McDermott et al., 2005 and references therein), as they have the potential for precise and accurate age control. Brazil comprises extensively developed karst landscapes, which are mainly located in the eastern part of the country (Auler et al., 2001). Over the last ten years, we have successfully collected speleothem samples from Brazil, with particular interest in samples from northeastern and southern Brazil (Fig. 1). The intent was to investigate (1) whether abrupt climate events are recorded in the southern hemisphere, particularly within the southern low latitudes, (2) if recorded, how they relate to their northern counterparts in both low- and high-latitudes, and (3) what kind of mechanisms could explain the overall geographic pattern of any recorded events.

Millennial-scale climate events

Speleothems and fossil travertine in the presently semi-arid northeastern Brazil suggest that this region experienced enhanced precipitation and groundwater recharge in the past (Auler and Smart, 2001) (Fig. 1). Constrained with a large quantity of U-Th dates, speleothem growth in northeastern Brazil was found to be highly episodic. The growth phases represent millennial-scale short pluvial periods during the last glacial period, whereas sample growth was not observed in dry conditions like today (Wang et al., 2004) (Fig. 2). When compared with the contemporaneous records from the northern hemisphere, these wet periods are synchronous with periods of weak East Asian summer monsoons (Wang et al., 2001), cold events in Greenland (Grootes and Stuiver, 1997), and periods of decreased river runoff to the Cariaco basin (Peterson et al., 2000).

Two continuous speleothem $\delta^{18}O$ records were reported from Botuverá Cave, southern Brazil (see site 1 in Fig. 1), covering the last 116 kyr and 90 kyr BP, respectively (Cruz et al., 2005; Wang et al., 2007). Both records successfully capture millennial-scale events that are superimposed on the orbital-scale variations during the last glacial period. The abrupt drop in $\delta^{18}O$ values associated with these millennial-scale events is large, with amplitudes of up to 2‰. Botuverá stalagmites were deposited in isotopic equilibrium, therefore, the calcite $\delta^{18}O$ mainly represents a regional change in precipitation regime (Cruz et al., 2005). Moreover, a trace element study on one of the samples shows that variations of Mg/Ca and Sr/Ca ratios are, in general, positively correlated with the calcite $\delta^{18}O$ change along the growth axis (Cruz et al., 2007), which confirms that Botuverá stalagmite $\delta^{18}O$ is dominated by the monsoonal rainfall amount.

Using their individual chronologies, we compared the 90-kyr-long Botuverá $\delta^{18}O$ record with the eastern China $\delta^{18}O$ profile, which is a combination of records from Hulu (Wang et al., 2001), Dongge (Yuan et al., 2004; Wang et al., 2005), and Sanbao caves (Wang et al., 2008) (Fig. 2). All records are precisely established with a typical relative 2σ age error of about 0.5–1%. Within dating errors, the comparison shows a remarkable anti-correlation between records on both orbital and millennial timescales. Throughout the whole

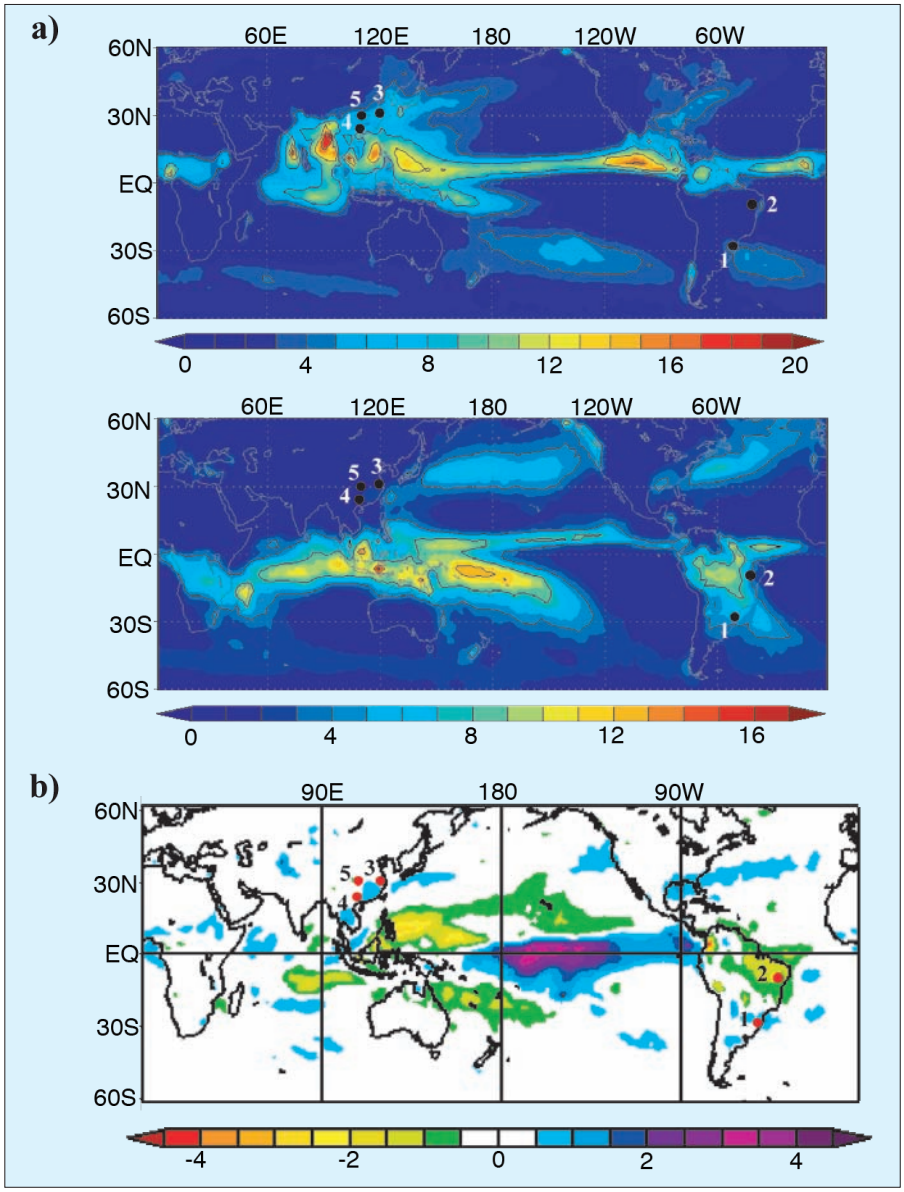

*Figure 1: **a**) Global long-term mean precipitation (mm/day) from 1979 to 2000. Upper: Averaged from June to August (JJA). Lower: Averaged from December to February (DJF). Numbers indicate cave locations in text: 1. Caverna Botuverá; 2. NE. Brazil caves (10°10'S, 40°50'W); 3. Hulu Cave (32°30'N, 119°10'E); 4. Dongge Cave (25°17'N, 108°5'E); and 5. Shanbao Cave (31°40'N, 110°26'E). (Images modified from www.cdc.noaa.gov/). **b**) Composite anomaly of precipitation rate (mm/day) during winter (Nov-Mar) associated with 8 El Niño events (1957-1958, 1965-1966, 1968-1969, 1972-1973, 1982-1983, 1986-1987, 1991-1992 and 1997-1998) from 1948 to the present. Anomalies are defined as the difference from the 1968 to1996 means.*

profile, the lower $\delta^{18}O$ in the Botuverá record coincides precisely with higher $\delta^{18}O$ in the eastern China speleothems, and vice versa, which indicates a rainfall seesaw between the two low-latitude regions. We also compared our Botuverá $\delta^{18}O$ record to the record of speleothem growth periods from northeastern Brazil (Wang et al., 2004), which is an indicator of pluvial phases in this semiarid region. The northeastern Brazil speleothem typically resumes growth when $\delta^{18}O$ values are low in the southern Brazil sample (Wang et al., 2007). Simply speaking, on millennial timescales, rainfall changes in southern Brazil and northeastern Brazil are in phase, and both anti-correlate with precipitation variation in eastern China.

ITCZ migration and climatic consequences

While the eastern China speleothem $\delta^{18}O$ profile shares similarities with the Greenland ice core record on millennial-scale climate events (e.g., Wang et al., 2001), southern Brazil rainfall variation does not resemble Antarctic temperature change (Fig. 2). Instead, the millennial-scale precipitation events in the Brazilian record generally anti-correlate with temperature changes over the Greenland. The asynchrony between Greenland and Antarctic warming suggests a possible teleconnection through an oceanic bipolar seesaw process and thermal inertia in the Southern Ocean (e.g., Stocker and Johnsen, 2003; Jouzel et al., 2007). The striking antiphase relationship between the Chinese and Brazilian records, however, suggests that the signal needs to be transmitted in a relatively rapid manner between the two low latitudes. Therefore, atmospheric interactions must be involved, likely through movement in the mean position of the intertropical convergence zone (ITCZ) and associated change in Hadley circulation.

During the last glacial period, an abrupt reduction in the Atlantic meridional overturning circulation (AMOC) induced sea ice expansion in the North Atlantic and a subsequent southward displacement of the ITCZ (e.g., Chiang et al., 2003; Zhang and Delworth, 2005). This may have caused an abrupt shift in the tropical hydrologic cycle, as seen in the Cariaco Basin (Peterson et al., 2000) and northeastern Brazil (Wang et al., 2004). Modeling efforts also indicate that weak ocean circulation may result in a positive sea surface temperature (SST) anomaly in the South Atlantic and a weaker pole-to-equator temperature gradient in the south (e.g., Crowley, 1992). As observed today (Liebmann et al., 2004), a warm SST anomaly in the western subtropical South Atlantic may stimulate a persistent intense South American Summer Monsoon and strong low-level jet, which consequently supplies isotopically depleted precipitation into southern Brazil (Vuille and Werner, 2005).

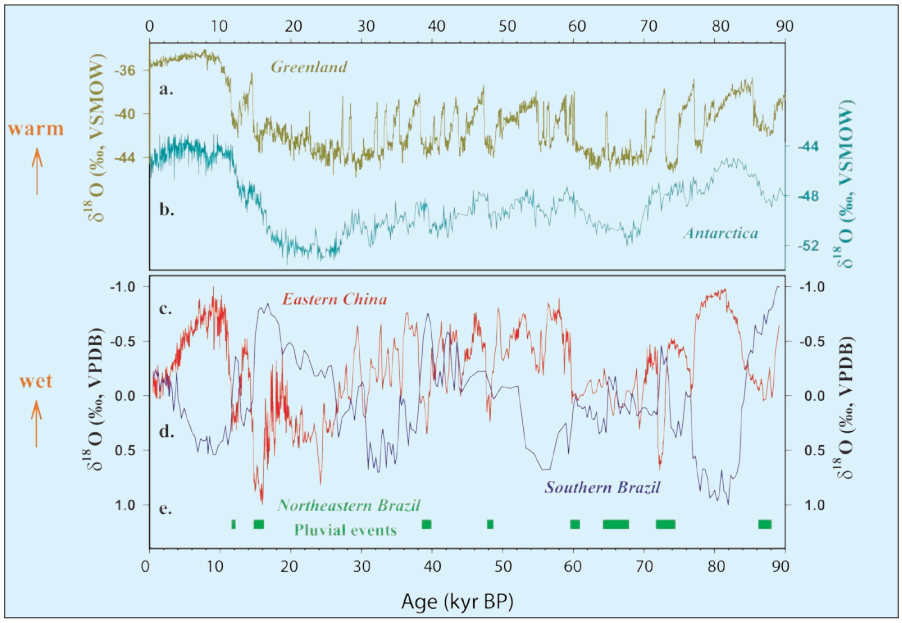

*Figure 2: Comparisons of speleothem records with polar ice core records. **a**) NGRIP ice core record from Greenland (light green, NGRIP members, 2004); **b**) EDML ice core record from Antarctica (dark green, EPICA members, 2006); **c**) Combined profile (red) of the eastern China speleothem $\delta^{18}O$ records, from Dongge Cave (Yuan et al., 2004; Wang et al., 2005), Hulu Cave (Wang et al., 2001) and Sanbao Cave (Wang et al., 2008); **d**) Southern Brazil speleothem $\delta^{18}O$ record (blue, Caverna Botuverá, Wang et al., 2007); **e**) Northeastern Brazil speleothem growth periods (green, Wang et al., 2004). Note the speleothem $\delta^{18}O$ records are normalized for easy comparison and scales are reversed, increasing downwards.*

Moreover, analogous to modern seasonal observations in boreal winters (Lindzen and Hou, 1988), southward ITCZ migration during millennial-scale stadial events may have caused meridional asymmetry in the Hadley circulation. A southward shift of the zonal-mean Hadley cell would change meridional moisture transport through intense ascending air masses in the southern low latitudes, and increased subsidence in the northern tropics and subtropics. Broadly, the northern low latitudes would be drier and the southern low latitudes wetter, which has been confirmed by recent model results (e.g., Chiang and Bitz, 2005; Timmermann et al., 2007). The opposite scenario would have been true during glacial interstadial periods.

AMOC vs. Super-ENSO mechanisms

It is still debated whether AMOC changes or tropical air-sea interactions, such as persistent El Niño-Southern Oscillation events (Super-ENSO), triggered the millennial-scale climate events (Broecker, 2003). Phase relationships of these events in Brazilian speleothem records may have implications on their mechanisms. The modern climate in both northeastern and southern Brazil is sensitive to the ENSO phenomenon. For example, modern El Niño events induce drought in northeastern Brazil and high precipitation in southern Brazil (Ropelewski and Halpert, 1987) (Fig. 1). If the modern ENSO behavior does not change substantially with time, the Super-ENSO scenarios may result in opposite rainfall patterns between the two regions. On the other hand, changes in the AMOC would cause a latitudinal ITCZ migration and associated changes in the Hadley circulation (e.g., Chiang and Bitz, 2005). The latter may cause in-phase precipitation changes in northeastern and southern Brazil on millennial timescales, which is confirmed by the speleothem record comparison (Fig. 2). This relationship is therefore consistent with shifts in the mean ITCZ position linked to AMOC changes but not with the Super-ENSO mechanism.

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Climate Change: The Karst Record (KR5)

Chongqing, China, 2-5 June 2008

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A total of 115 delegates from 18 countries attended the 5th international conference of Climate Change: The Karst Record (KR5). The conference, co-sponsored by PAGES, consisted of 76 oral presentations and 29 posters.

Participants presented studies from all over the world and on various climatic parameters, such as temperature, precipitation, monsoon strength and the occurrence of dust storms. Karst (speleothem) records presented at the conference obtained up to seasonal- to decadal-resolution, covered timescales back to Glacial Termination IV (about 340 kyr ago) and were compared to solar forcing and marine and ice core records. In addition to signals of $\delta^{18}O$, $\delta^{13}C$, lamination and trace elements in speleothems, more novel approaches, such as clumped isotopes, biomarkers, rare earth elements and pollen, were presented at the conference and their paleoclimatic interpretation was discussed.

The delegates also discussed problems and suggestions for future work:

(1) Interpretation of speleothem records: Delegates agreed that research should now focus on integrating stalagmite records on a regional climate basis, and on resolving differences between records. Also discussed was the need to untangle (A) the climate signal in stalagmite ^{18}O records—how much is attributed to changes in regional atmospheric patterns (i.e., relationship of monsoon 'strength' with rainfall amount) and how much to global temperature trends, and (B) discrepancies among different stalagmite records from the same region.

(2) Chronology: Although much progress has been made on ICP-MS $^{230}Th/U$ dating, we need to continue considering dating uncertainties, especially those relating to "contamination" by initial ^{230}Th. Initial ^{230}Th is introduced into karst deposits with detrital material but may also be carried by organics dissolved in the drip water. The resulting stalagmite $^{230}Th/^{232}Th$ ratio can therefore be substantially different from the mean crustal value that is often used to correct the dates. It was therefore agreed that more measurements of Th in drip water and modern samples of cave deposits are required to obtain the initial Th ratio. Furthermore, soil from above a cave should be analyzed to get an idea of the soil Th and U isotope ratio. Delegates also felt that greater use should be made of 'isochron' dating. The need to address how to combine analytical error and sampling error when performing U-Th dates was also recognised as an important focus for future research. It was agreed that raw ages should be quoted in publications and errors should also be cited when samples are detritally corrected.

115 delegates from 18 countries attended the conference and provided 76 oral presentations and 29 posters.

Delegates also discussed the use of U-Pb methods to extend the dating of karst records back as far as 10-20 Myrs for comparison with loess and marine records.

(3) Karst Hydrology: The hydrology of the vadose zone is important, as each drip site can be affected by different climatic and environmental conditions, and therefore each stalagmite may record different components of the climate. The need for more work on understanding hydrological processes in the karst system was recognized, for example, using dye tracing from soil to the cave to measure transfer time/climate signal transfer from the surface to the cave drips.

(4) Other applications: In addition to paleoclimate research, it was agreed that karst records should make a greater contribution to the understanding of natural hazards, environmental issues and ecosystem problems (e.g., karst-desertification).

(5) Data archiving: The importance of adding all data to the World Data Center for Paleoclimatology was recognised by the delegates. This must include archiving of ^{13}C and ^{18}O with the raw chronological data.

(6) Sustainability of sampling stalagmites: Stalagmites are non-renewable resources. It was felt that the community should archive samples for younger generations and help them to continue the karst record series. The delegates recommended that, where possible, coring techniques and/or broken stalagmites should be used.

The next karst record meeting, KR6, will be held at the University of Birmingham, UK in 2011.

Establishing a Northern Eurasian paleoecological database: The pollen data

University of Southampton, UK, 14-19 April 2008

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As part of the UK's QUEST program (http://quest.bris.ac.uk/), QUEST Deglaciation (Q-D) aims to evaluate the performance of a new, fast Earth System model in simulating the major changes of the late Quaternary (21 - 0 kyr). This model will include continuous simulations of vegetation patterns for this period, which can be evaluated by comparison with the reconstructed vegetation based upon pollen and macrofossil evidence. Additionally, Q-D focuses on the northern extra-tropics (NET), a region sensitive to climate change and thus particularly useful in evaluating the performance of climate models.

Previous data-model comparisons using the time-slice approach (0, 6, 21 kyr BP) (Bigelow et al. 2003; Kaplan et al. 2003) demonstrated the existence of a potentially large dataset of pollen sites in Northern Eurasia. However, continuous pollen records and related radiocarbon dates have not been brought together in a single database, and thus remained to be collected in full. The Global and European Pollen Databases (GPD & EPD) indicate that these databases are a good source of material for North America and Europe but that a considerable proportion of sites in the Former Soviet Union (FSU) are not archived.

Furthermore, during the macrofossil workshop held in 2007 that was partly funded by PAGES (see *PAGES News*, 15(1): 27-28), it became apparent that the participants either had collected or knew of additional pollen data from these data-sparse regions. Therefore, it was decided to hold a second data-gathering workshop, this time to obtain well-dated pollen records from sites in the FSU during the critical climate interval 21 kyr to present. The macrofossil and pollen data will be housed in the same database, providing the possibility to analyze the two datasets simultaneously. Crucial to such a database is the dating and the age-models, which are discussed further below.

The workshop took place at the University of Southampton, UK, and included nine academics who work entirely or primarily in the FSU.

Outcomes of the workshop

Pollen data from 87 sites were added to the database during the four-day workshop (Fig. 1). All sites are well dated and a small number of the sites extend as far back as 48 kyr BP. These earlier sites were included because they were deemed useful for other projects, such as Quaternary QUEST (http://quest.bris.ac.uk/research/themes/Quaternary.html). Metadata such as basin and lake size, sample type and landscape descriptions were included because these are important for understanding pollen source area and vegetation reconstructions. Taxonomy is also important, especially since the palynologists from the FSU often use a high level of taxonomic resolution in their investigations. All original identifications will be stored in the database alongside a standardized taxonomic list that will allow comparison with other regions in the NET. The pollen data will be stored as original counts with a facility for calculating percentages depending on the pollen sum required.

Figure 1: Location of the 87 sites with pollen data added during the workshop.

To allow data-model comparisons with optimal precision, all radiocarbon dates were calibrated to calendar years using the INTCAL04 calibration dataset (Reimer et al., 2004). Age-depth models were then constructed for each site for which a series of radiocarbon dates was available. This was undertaken by Ian Matthews (Royal Holloway, University of London, UK) using Bayesian-based OXCAL4.0 sequence algorithms (http://c14.arch.ox.ac.uk/ see Bronk Ramsey, 2008). This approach is statistically rigorous and generates the best-constrained age models that can be determined using currently available calibration information. During the workshop, 35 individual age-depth models were constructed and the results for the remaining sites will be completed shortly.

Use of the database

Following data checking, the pollen data will be placed in the Eurasian Pollen Database, which will eventually be combined with the Eurasian Macrofossil Database (both Microsoft Access). The first objective will be to produce a synthesis of vegetation dynamics in Eurasia for the interval 21 kyr to present, of a temporal and spatial resolution suitable for comparison with the QUEST-Deglaciation GCMs. The combined Eurasian Pollen-Macrofossil database will also enable a better understanding of the position of key ecotones, such as the northern treeline, and the distribution of palynologically poorly represented species, such as Larix.

Note

The pollen element of the database will be available in 2009 via a public portal, following the production of a synthesis paper by workshop participants. If you wish to be kept informed of the progress of the database, please contact Heather Binney or visit http://gg-svr7.geog.soton.ac.uk/staff/mee/nepd/default.aspx

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International Partnerships in Ice Core Sciences (IPICS) Steering Committee meeting

Vienna, Austria, 19-20 April 2008

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Ice cores have become a cornerstone of research into climate and biogeochemistry, and have provided some of our iconic paleoclimate datasets. IPICS is a group of scientists, engineers and logistics experts from the leading laboratories and national operators carrying out ice core science. It was formed as a result of meetings in 2004 and 2005, and now has membership from 21 nations. Its mission includes defining priorities for ice core science and facilitating the steps to make the priority projects happen.

29 scientists and engineers, representing 16 of the IPICS nations, attended a meeting that followed the European Geosciences Union General Assembly. A fundamental aspect of IPICS is exchange of information about future plans, and this was achieved before the meeting by the preparation of a document with plans from each nation. This freed up time at the meeting for the main business, which was to approve science plans and outline implementation or coordination plans for the four IPICS priority projects.

One of the four priority projects is already underway: 14 nations have banded together to drill a new ice core (NEEM), with the intention of obtaining a complete record of the last interglacial from northwest Greenland. During that period, both the Arctic and Antarctic appear to have been warmer than present. Knowing precisely the climate signal and the ice sheet response in this period would place serious constraints on the impact of warming on ice sheet stability, highlighted by IPCC as one of the big unknowns for the next century and beyond. For this reason, the meeting discussed the option of building on the NEEM project to make a bipolar contribution to understanding the last interglacial, and a small group will consider this. Meanwhile, as of mid-May, 9 people are setting up the camp ready for drilling to start this summer.

The most ambitious IPICS project aims to extend the Antarctic ice core record beyond the 800-kyr period of EPICA Dome C. We know from marine data that 40 kyr climate cycles prevailed (c.f., 100 kyr) before about a million years ago, and an ice core extending that far would allow us to assess, for example, the role of CO_2 in this change of pace. A major survey and modeling program has to be completed before the drilling sites can be identified, and this will start with large airborne surveys in austral summer 2008/09. The meeting approved a plan that outlines the steps to be completed before a site is considered suitable, and agreed general principles for how international drilling and science consortia might work.

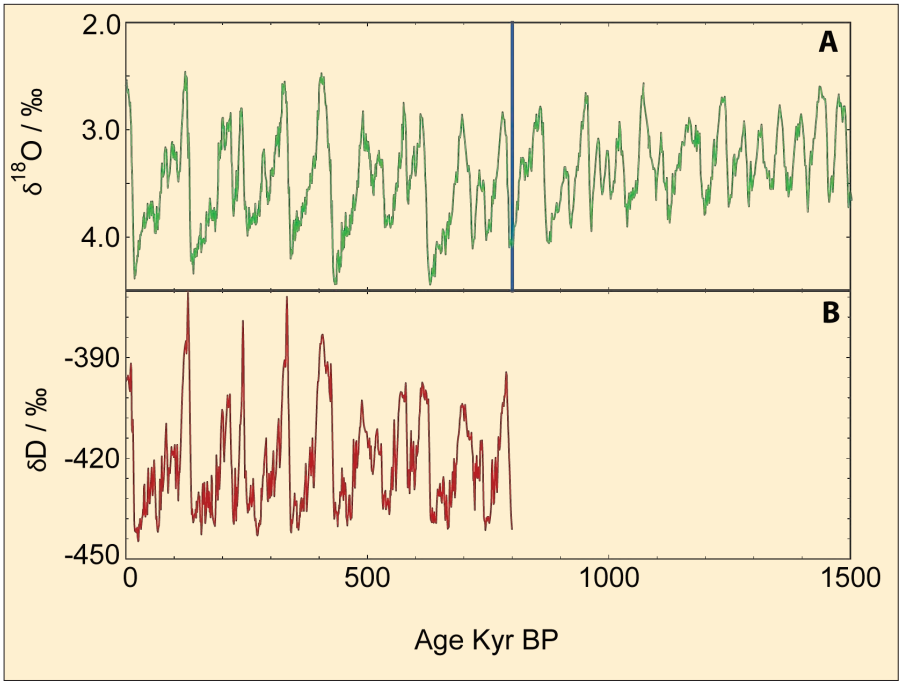

*Figure 1: The ice core record (**B**; Jouzel et al., 2007) so far extends back 800 kyr, spanning only the period of 100 kyr cycles. Marine records (**A**; Lisiecki and Raymo, 2005) show that 40-kyr cycles prevailed before about 1 Myr.*

The third project aims to obtain a bipolar network of cores covering around 40 kyr, with the aim of understanding the spatial pattern of change during Dansgaard-Oeschger events and their Antarctic counterparts, and the last glacial termination. Many of the cores have already been drilled, and the members agreed where further cores should be encouraged, what measurements must be made, and the need for specific funded synthesis activities.

The final project will obtain a global set of ice cores extending back around 2000 years, suitable for inclusion in IPCC-style reconstructions. The meeting agreed on the requirements for cores to be acceptable for such work, and again on the essential need for synthesis, involving the kind of stringent statistical study now done in some other communities with annually resolved data. Part of the discussion also focused on non-polar ice cores, and the urgent need to collect an archive of material from glaciers that are expected to melt in the near future.

Of course these projects require not just science but significant technical achievement, both in drilling, logging holes, proper curation of ice, and analysis. A technical sub-group is tasked with, for example, finding suitable drilling fluid for the oldest ice project. Sub-groups for each of the priority projects will pursue steps to make them a reality, and IPICS will have another formal meeting attached to the PAGES OSM in July 2009. For further information, please see www.pages-igbp.org/ipics/

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ESF EuroCLIMATE Spring School: Late Quaternary timescales and chronology

Piran, Slovenia, 20-26 April 2008

Sébastien Bertrand[1], A. Burnett[2], K. Saunders[3], J. Striberger[4], Y. Axford[5] and S. Coulter[6] (Workshop Participants)

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Earth scientists are increasingly using natural archives to reconstruct past changes in climate, seismic activity, ocean circulation, glacial advances, atmospheric composition, etc. In order to compare geological records across space and time, they need to be properly dated and accurate chronologies need to be developed. Although several dating techniques have been in use for the last 50 years, the methods are still evolving and being refined.

To familiarize geoscientists with recent developments in dating techniques, the European Science Foundation sponsored a EuroCLIMATE Spring School on "Late Quaternary Timescales and Chronology" from 20 to 26 April 2008, in Piran, Slovenia. The Spring School, organized by Barbara Wohlfarth (Stockholm University, Sweden) and Bernd Kromer (University of Heidelberg, Germany) brought together 15 lecturers and 40 participants from 17 different countries, mainly from Europe but also Australia, New Zealand and the USA. The meeting included topical lectures, short participant presentations, and poster and discussion sessions. During the week, participants received various important insights regarding the development of accurate chronologies for their records. The most important issues discussed during the meeting are summarized hereafter.

First, every method requires appropriate samples. Selecting suitable samples may be tricky, especially for optically-stimulated luminescence (OSL) and cosmogenic radionuclide dating, and it is advisable to work in close collaboration with the lab running the samples. It is also essential to select appropriate dating techniques, apply them consistently, and clearly indicate the precision, methodological details and assumptions of the selected techniques in publications. For the radiocarbon technique, for example, authors should always clearly indicate the nature of the analyzed samples (e.g., bulk sediment vs macrofossil). Moreover, as numerous calibration and correction datasets have been published since the discovery of the radiocarbon dating technique, scientists should be extremely careful to select appropriate datasets for calibration and correction of radiocarbon results. Again, these choices should be clearly stated in the resulting publications. Several free software programs are available for download (BCal, Bpeat, OxCal, Calib), and they all feature the most common calibration and correction options.

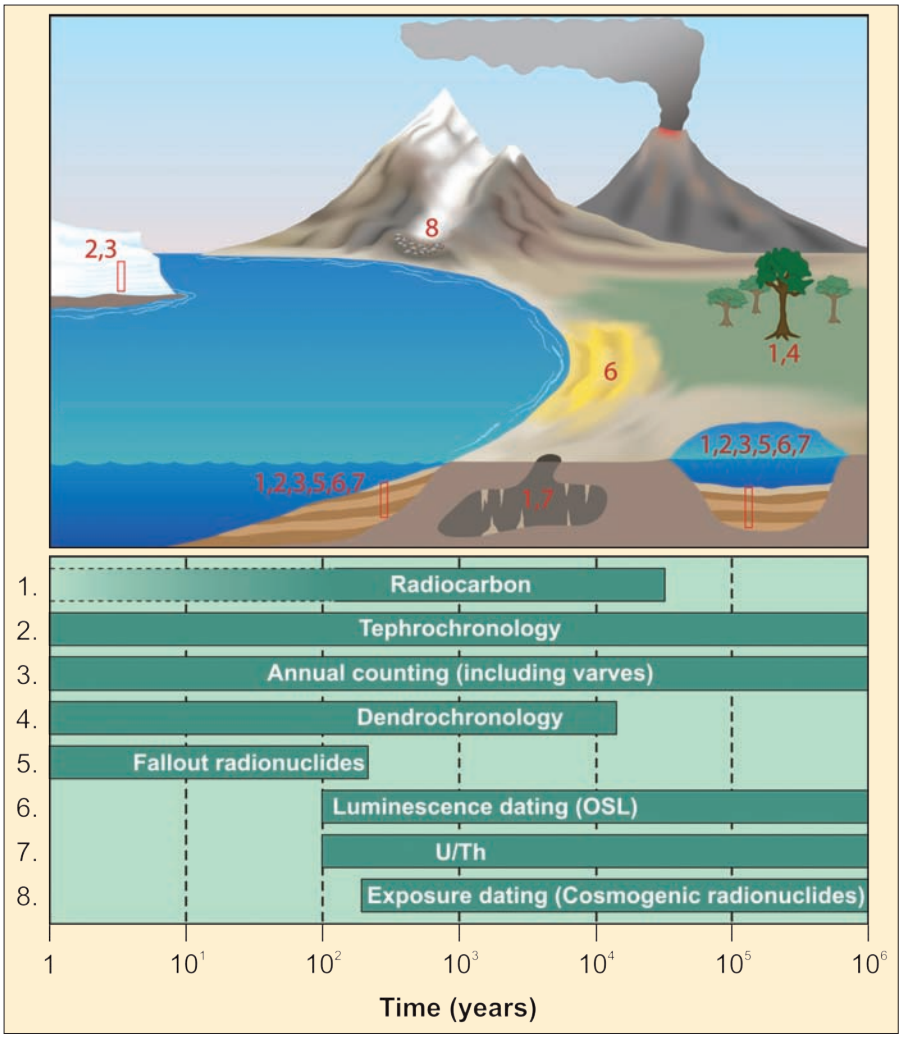

Figure 1: Late Quaternary depositional environments (ice caps, glacial moraine deposits, loess and sand dunes, trees, marine sediments, speleothems, lake sediments) and associated dating techniques discussed during the ESF Spring School. The lower part of the figure represents the effective dating ranges of the different techniques.

Second, it is common practice in geosciences to apply a linear age model between dates or to use a polynomial regression to model an age-depth relationship. Martin Blaauw, in particular, illustrated the fallacy in this approach. It is essential when constructing age models to consider prior knowledge of the archive, including visible sedimentological changes, hiatuses, density changes, etc.—all of which are likely to be associated with a change in sedimentation rate. A variety of tools based on Bayesian statistics (e.g., OxCal 4.0) are now available to help construct these models and should always be used when constructing chronologies.

Third, attendees discussed the rationale and best practices to tune and wiggle-match paleo-records. As the lead and lag relationship between paleoclimate events is an important issue in recent paleoclimate research, extreme care must be taken before tuning or wiggle-matching records. A distinction was made between the two terms, which are often used inappropriately. Tuning is changing the chronology of a record to match a known cyclicity, based on a known cause-and-effect relationship (e.g., orbital tuning). Wiggle-matching is matching events without a known mechanistic link and/or cyclicity. The main conclusion reached among the attendees was that a simple correlation without a cause-

and-effect explanation is not sufficient to tune or wiggle-match records. Scientists should first use independent chronologies to demonstrate that the records behave synchronously and there must be reason to believe the records are driven by the same mechanisms. Once such a comparison has been made, it is then permissible to tune or wiggle-match. It is recommended to use a reasonable number of tie-points but not more than two per distinct lithological unit, as too many tie-points would cause unnatural breaks in accumulation rates.

This Spring School was a great opportunity for senior and younger scientists to meet and discuss different chronological issues. The 17 lectures and the 40 participant presentations covered a wide range of dating techniques available for Late Quaternary archives, as well as various geographic areas. It is hoped that participants will use and circulate the information regarding best practices for constructing Late Quaternary chronologies—from sample selection in the field to publication of the results.

A new PAGES Working Group: Arctic2k - Arctic climate during the last 2 millennia

Boulder, USA, 8 March 2008

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Climate change in the Arctic is amplified, mostly due to ice- and snow-albedo feedback effects. Warming in the Arctic has occurred at about twice the rate of the global mean, both from the 19th to 21st centuries and from the late 1960s to present (Arctic Climate Impact Assessment, 2004). This trend has further accelerated during the past decade, as evidenced both by the dramatic decrease of summer sea ice cover and increased melt rates of glaciers (e.g., Kohler et al., 2007; Comiso et al., 2008). Observed increases in Arctic river discharge and freshening of Arctic water masses are consistent with human-induced Arctic moistening (Min et al., 2008). The effects of anthropogenic warming will continue to be superimposed on natural climate variability. Unfortunately, Arctic instrumental measurements only extend back to the mid-20th century, which limits our understanding of multi-decadal and -centennial spatial and temporal natural variability in this region.

Figure 1: Drilling site at Lomonosovfonna, Svalbard.

In recent years, some high-resolution Arctic paleoclimate data have been obtained from ice cores (Figs. 1 and 2), and lake and ocean sediments. PAGES Working Group (WG) on Arctic climate during the last two millennia (Arctic2k) is a new initiative that will generate additional records and synthesize these high-resolution paleoclimate data to assess and elucidate both the timing and variability of Arctic climate change during this period. In particular, the group will contribute to regional reconstructions of the last 2 kyr, under the new PAGES Focus 2 "Regional Climate Dynamics". Forty paleoclimatologists from both the data and modeling communities, gathered for the first Arctic2k workshop at NCAR, Boulder, USA on 8 March 2008, following the 38th International Arctic Workshop at INSTAAR (5-7 March 2008).

During this one-day kick-off, workshop participants gave a preliminary overview of national and regional activities from the different paleocommunities. The future research issues for the Arctic 2k WG were discussed extensively. These included emphasizing the need to assess sensitivities and thresholds in the Arctic system, spatial and temporal modes of climate signals, persistence of anomalies, frequencies of extreme events, rates of change, and irreversibility and feedback mechanisms. Some central questions emerged:

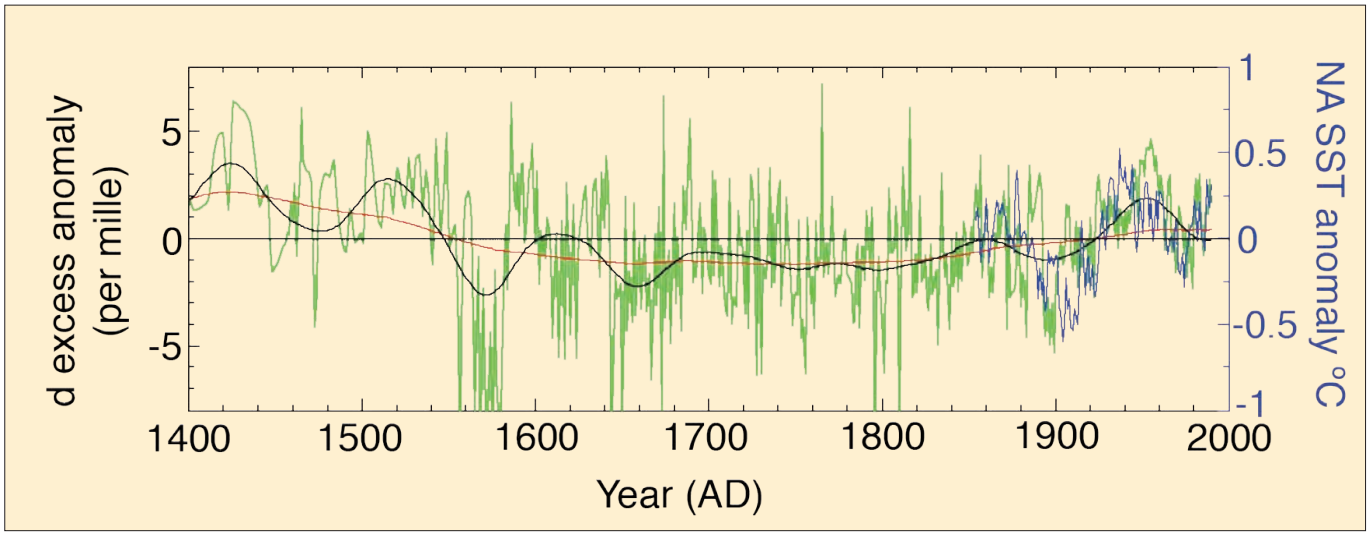

*Figure 2: **Left axis**: Annual mean deuterium excess anomalies for the Lomonosovfonna ice core (green); non-linear trend (red) and non-linear trend with superimposed centennial component (black), both retrieved using Singular Spectrum Analysis. Deuterium excess ($d = \delta D - 8 * \delta^{18}O$) is a sensitive indicator of sea surface temperature (SST) of the area where the precipitated moisture originally evaporated. **Right axis**: Annual mean SST anomalies in mid-latitude North Atlantic between 20°-45°N (blue) (Divine et al., submitted).*

a) Is the 20th century warming of the Arctic unprecedented in the last 2 kyr? b) What is the multi-decadal- to century-scale variability in Arctic climate? c) What portion of the Arctic-wide and regional temperature changes during the last 2 kyr can be explained by changes in solar irradiance and volcanic activity, and what portion is related to internal adjustments of the climate system? In addition, improvement of our confidence in the interpretation of the proxies and the geochronology were identified as important underlying goals.

The group decided to limit the study area to north of 60°N latitude, which includes Greenland, Iceland and Alaska. A website for the WG has been launched, which lists the researchers and subprojects involved (www.pages-igbp.org/science/arctic2k/). Furthermore, a metadatabase is being developed there to assist the compilation of Arctic records. An immediate task is to map the available data and to assess regions with data gaps. The Arctic2k WG will be organizing three regional reconstruction workshops in 2009 (Scandinavia-Nordic Seas-Svalbard; Baffin Bay-Greenland-Iceland; Northern Pacific-Alaska). All paleoclimate scientists working in the Arctic with high-resolution data and/or modeling are welcome to join and contribute to the PAGES Arktic2k WG.

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Serbia

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PAGES PAST GLOBAL CHANGES

The famous Serbian scientist Milutin Milanković made a fundamental impact on modern paleoclimate research with his theory of the ice ages and the relationship between variations of the Earth's orbit and long-term climate change (Milankovitch cycles). In spite of this, and despite their potential significance, Serbian paleoclimatic and paleoenvironmental archives are still relatively understudied. The present Serbian paleoenvironmental community is predominantly focused on investigation of local Mesozoic and Cenozoic formations, with some of the most significant advances being made in late Cenozoic loess research. Indeed, the Serbian loess-paleosol sequences have been established as some of the oldest, thickest and most complete in Europe.

In publishing the Serbian national PAGES, we hope to motivate the PAGES community to help us to further improve the understanding of past climate and environmental evolution in our region.

National highlight

A unique Middle Pleistocene European loess-paleosol record

Serbian loess deposits are among the oldest and most complete loess-paleosol sequences in Europe. In particular, the thick loess-paleosol sequences in the Vojvodina region contain a detailed paleoclimatic record from the late-early Pleistocene (Fig. 1). The current stratigraphic model of the Vojvodinian loess-paleosol chronostratigraphic units follows the Chinese loess stratigraphic system, using the prefix "V" to refer to the standard Pleistocene loess-paleosol stratigraphy in Vojvodina (Marković et al., 2008). Variations in sedimentological, magnetic and geochemical proxies, dust accumulation rates, and intensity of pedogenesis provide clear evidence for the Middle Pleistocene climatic and environmental transition in the region. The onset of loess deposition in the Vojvodina region indicates a direct link to the progressive aridification of the interior of Eurasia from the early Pleistocene. Multidisciplinary results provide valuable data for detailed reconstruction of natural processes and climatic change over the last c. 850 kyr, and allow correlation with equivalent records from Central Europe and the Black Sea coast to Central Asia and China.

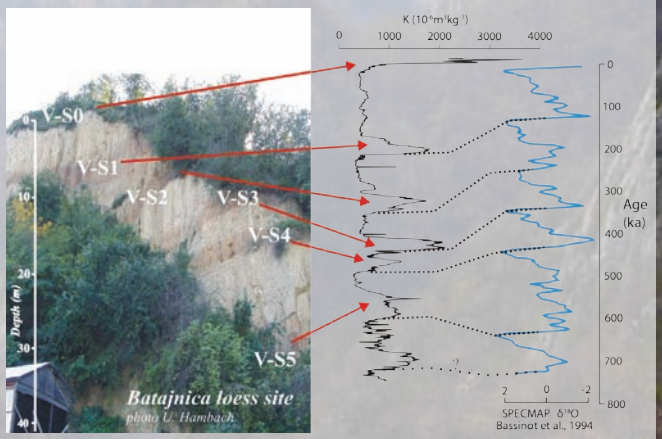

Figure 1: Correlation between Batajnica loess-paleosol sequence and SPECMAP paleoclimatic timeseries (modified from Marković et al., submitted at Quaternary International)

Research Institutions

Chair of Physical Geography, University of Novi Sad; www.ig.ns.ac.yu/fizgeo.htm
Institute of Lowland Forestry and Environment, Novi Sad; www.ilfe.org/
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Call for contributions to the PAGES newsletter

All PAGES newsletters have an open section for general contributions. If you would like to contribute a "Science Highlight", "Workshop Report", "Program News", or an amusing "Tales from the Field" story, please contact Louise Newman (newman@pages.unibe.ch). The next deadline is 30 September 2008.

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