

# *SISAL: bringing added value to Speleothem research*

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# SISAL: Bringing Added Value to Speleothem Research

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**Abstract:** Isotopic records from speleothems are an important source of information about past climates and, given the increase in the number of isotope-enabled climate models, are likely to become an important tool for climate model evaluation. SISAL (Speleothem Isotopes Synthesis and Analysis) have created a global database of isotopic records from speleothems in order to facilitate regional analyses and data-model comparison. The papers in this Special Issue showcase the use of the database for regional analyses. In this paper, we discuss some of the important issues underpinning the use of speleothems and how the existence of this database assists palaeoclimate research. We also highlight some of the lessons learned in the creation of the SISAL database and outline potential research going forward.

**Keywords:** SISAL; speleothems; database; data synthesis; isotopes

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## 1. Speleothems as Recorders of Past Climates

Speleothems are secondary cave deposits that form when water percolates through carbonate bedrock. Atmospheric CO<sub>2</sub> and CO<sub>2</sub> generated by root respiration and the decomposition of organic matter is dissolved by rainwater as it percolates through the soil, which produces carbonic acid that rapidly dissociates to produce weakly acidic water. In karst areas, this acidic water dissolves carbonate as it percolates through the bedrock until water becomes supersaturated with Ca<sup>2+</sup> and HCO<sub>3</sub><sup>-</sup>. Additional CO<sub>2</sub> contributions from microbial and root respiration occur in the vadose zone [1]. When the percolating waters emerge in a cave, CO<sub>2</sub> degassing from the drip water to the cave atmosphere induces CaCO<sub>3</sub> precipitation and forms stalagmites and stalactites [2]. CaCO<sub>3</sub> precipitation can be as either calcite or aragonite.

Speleothems are a rich archive of terrestrial palaeoclimate information. Although many different measurements can be made on speleothems, the most common types for palaeoclimate reconstructions are the stable isotopes of oxygen and carbon ( $\delta^{18}\text{O}$ ,  $\delta^{13}\text{C}$ ). Speleothem carbonate  $\delta^{18}\text{O}$  values are often more positive than would be expected from the temperature-dependent isotope fractionation during deposition. Changes in speleothem  $\delta^{18}\text{O}$  are primarily driven by changes in precipitation amount, temperature, or precipitation source linked to atmospheric circulation changes [3–5]. However, these records can also be affected by changes in drip water residence time in the overlying karst, the cave temperature, and ventilation dynamics or kinetic fractionation during carbonate deposition, all of which are linked indirectly to changes in climate. Changes in  $\delta^{13}\text{C}$  are generally interpreted to reflect the changing abundance of C<sub>3</sub> and C<sub>4</sub> plants above the cave and, hence, to provide an indirect signal of precipitation changes [6]. However, kinetic fractionation effects between the stalactite and the stalagmite due to evaporation or degassing of the groundwater within the aquifer can over-ride the C<sub>3</sub> versus C<sub>4</sub> signal [7]. Additionally, the  $\delta^{13}\text{C}$  of C<sub>3</sub> plants is inversely related to the drawdown of CO<sub>2</sub> during photosynthesis, which increases with environmental aridity [8]. Thus, changes in  $\delta^{13}\text{C}$  can be produced without requiring large shifts in vegetation type and, given that the availability of water for plant growth is not simply a function of precipitation inputs, could reflect complex climatic changes.

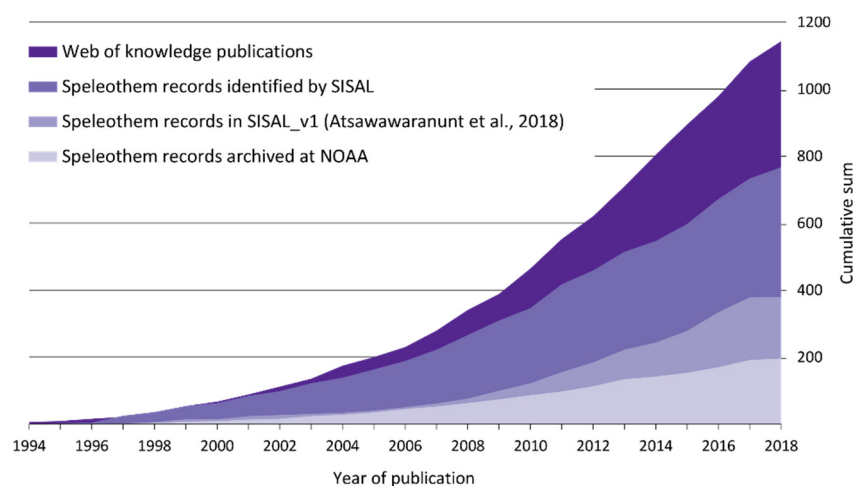
Furthermore, organic matter decomposition and mineral dissolution in the soil as well as at the soil-rock interface can also affect the  $\delta^{13}\text{C}$  signal [9].

Despite potential uncertainties in the interpretation of the isotopic records, speleothems have been used to infer shifts of the Intertropical Convergence Zone [10], glacial-interglacial transitions, and millennial-scale variability of tropical atmospheric circulation in response to Dansgaard-Oeschger cycles [11,12] as well as the timing of climate-related migrations across the Saharan-Arabian region and how these enabled the migration of humans out of Africa [13].

## 2. The Rationale Behind the SISAL Database

The extensive distribution of karst landscapes (see papers in this Special Issue for regional maps with carbonate lithologies and speleothem coverage) means that speleothems are studied worldwide. There has been a marked increase in speleothem-based publications over the last few decades (Figure 1), which was facilitated by the development of spectrometers that can deliver high-precision U-Th dates and improved techniques for high-resolution sampling. However, analysis of spatio-temporal patterns of isotopic changes, which is necessary to facilitate reconstructions of large-scale changes in atmospheric circulation, is not possible unless the > 700 published speleothem records are documented in a standardised way with adequate metadata in a database. There have been attempts to compile speleothem data globally from publicly available data in the last few years, particularly in the model evaluation context [14,15]. However, repositories such as the NOAA/World Data Center Paleoclimatology Program (<https://www.ncdc.noaa.gov/data-access/paleoclimatology-data>) have an uneven and incomplete representation of the published data. In particular, the NOAA/WDCPP archive contains only 196 speleothem records (Figure 1) and the key data and metadata needed to assess uncertainties are often missing, which prevents quality control or screening of the records.

SISAL (Speleothem Isotopes Synthesis and Analysis, <http://pastglobalchanges.org/ini/wg/sisal/intro>, last access 26 September, 2018), which is an international working group under the auspices of the Past Global Changes (PAGES) project, is seeking to redress this situation by creating a systematic global synthesis of speleothem data. The first version of the SISAL database [16,17] includes 381 speleothem isotope records either from public repositories or provided by the original investigators. This database includes an exhaustive set of key metadata such as information on the age-models, the geology of the cave site or speleothem mineralogy, to facilitate quality control and to ensure the reusability of the data.



**Figure 1:** Evolution of speleothem palaeoclimate research. Data from top to bottom: “Web of Knowledge” are the publications that resulted from querying the keywords “speleothem” and “isotope” in <http://www.webofknowledge.com> on 10 June, 2018 (may include publications only using the records). Speleothem records identified by SISAL working group members. Records in the SISAL database [16,17]. Speleothem records lodged in the NOAA repository as of 5 October, 2108 (<https://www.ncdc.noaa.gov/data-access/paleoclimatology-data>). Compilations have been excluded.

The SISAL database, as showcased in this Special Issue, is an important tool for exploring past climate changes. We also envision that it will be used for evaluating state-of-the-art climate models, especially those that explicitly simulate water and carbon isotopes as a tool for characterizing and diagnosing the atmospheric hydrological cycle [18–20]. These models are evaluated against modern observations of the isotopic composition of rainwater [21,22], but modern data only document late 20th century climates. Speleothem records can provide a test of model performance over a wider temporal range, including intervals when the climate change was as large and/or as fast as that projected for the 21st century [23]. Lastly, by contributing to improved geographical coverage of palaeodata, SISAL will improve the reliability of data-assimilation techniques used in both model evaluation and climate reconstruction [24–26]. This Special Issue provides eight examples of the use of the SISAL database for regional syntheses and palaeoclimate interpretation. In showcasing the usefulness of the speleothem data, this Special Issue provides a motivation for the planned expansion of the SISAL database to include all of the 767 records that we have identified (Figure 1).

### 3. What is SISAL Bringing to the Table?

Current practices of data reporting vary extensively among publications and the reported data are usually incomplete. In addition, many of the pioneers in speleothem research are now retiring, which makes it more difficult to access their unpublished data (or published data not logged in public repositories). Both of these situations present an important “data rescue” challenge and, in some cases, resulted in published speleothem records not being included in the SISAL database. For example, old and unpublished records account for 54% of the speleothem records that we have identified from Australia, including important sites such as Little Trimmer cave [27,28], Victoria cave [29,30], Frankcombe cave [31,32], and Royal cave [33,34]. By providing a template for what data is required and working with individual scientists, SISAL is engaging in this data rescue.

The absence of community-agreed data-reporting standards prevents improvements to the original study and hinders data re-usability in the context of wider studies even though there have been attempts to outline guidelines and protocols for documenting speleothem records [35]. The template provided by the NOAA/WDCPP is the one predominantly used by the community to make data publicly available. However, beyond geographical location and the dating and isotope measurements, the metadata required is limited. The depth associated with each isotope sample, which is needed to construct age-depth models with alternative or new methodologies, is often unreported. As Fairchild and Baker [1] said, “collected speleothem records are scientifically a non-renewable resource” and this makes it vital to preserve as much information as possible through appropriate documentation and archiving so that future research can build on these non-renewable resources. The community-endorsed metadata in SISAL is a step towards providing guidelines on what information needs to be made available for scrutiny, verifiability, re-use, and re-purposing of speleothem data since it is increasingly required by funding agencies in support of open science in line with the FAIR (Findable, Accessible, Interoperable, Reusable) principles for scientific management and stewardship [36]. The template used to upload speleothem records to the first version of the SISAL database can be found in the Supplementary Material.

The analyses of the SISAL database in this Special Issue allowed us to identify spatial and temporal gaps in the coverage of speleothem records. This has permitted us to identify regions in which more work would be beneficial. For example, enormous scope for further work has been identified in southern Africa to assess the robustness of  $\delta^{18}\text{O}$  signals between 11–6 ka BP ([37], this issue). Additionally, a ~20,000 year period centered around 100 ka BP that lacks speleothem records has been identified in the Eastern European/Turkey region ([38], this issue). Comparing the distribution of speleothem records with global or regional maps of carbonate lithologies (e.g. WOKAM, [39]) helps identify gaps in spatial coverage due to limited research or difficulties of access rather than the absence of karst. Kaushal et al. ([40], this issue) use such a comparison to advocate for an improved spatial coverage to allow a detailed examination of the regional teleconnection patterns associated with the Indian Summer Monsoon. Along similar lines, Zhang et al. ([41], this issue) argue that, despite the

existence of well-developed karst in NW China and in the Tibetan plateau, the low number of speleothem records from these regions is due to difficulties of access. Identification of biases in temporal coverage, facilitated by having a global database, has led to the identification by Lechleitner et al. ([42], this issue) and Burstyn et al. ([43], this issue) of a positive bias in the abundance of speleothem records from Europe and the Middle East after the Last Glacial Maximum (LGM). This is likely a combined result of less natural attrition (which makes it more suitable for geochemical analyses because it is less chemically altered/weathered), as well as a research focus on the recent past to facilitate calibration with instrumental data or to investigate human migrations.

A significant advantage of a global database of speleothem data is the potential for documenting regional and/or large-scale changes through time either by presence/absence of records (e.g. [37], this issue) or by their isotopic values (e.g. [44], this issue). For example, although there is an uneven spatio-temporal coverage of speleothem records in North Central America ([45], this issue), the observed more negative and variable  $\delta^{18}\text{O}$  at high latitudes during the LGM is consistent with studies that suggest a more variable temperature during this period at higher latitudes [46].

Past changes in precipitation amount and/or local vegetation types would affect evapotranspiration rates that, in turn, modulate the  $\delta^{18}\text{O}$  signal. These processes cannot always be inferred solely from speleothem stable isotope records and complementary data needs to be integrated from other climate archives, such as pollen records from lakes, peat, or ocean sediments. The availability of a database where all palaeo-records are consistently formatted enables such comparisons and ultimately contributes to a better understanding of the processes controlling the isotopic signals registered in speleothems.

#### 4. Enabling Quality Control Assessment of the Records

Comprehensive databases allow analyses to be based on a large number of records and, thus, avoid biases that might arise by selecting sites from particular or atypical localities. It has been shown, for example, that the reconstruction of a global cooling trend during the Holocene [47], which caused controversy because climate models are unable to reproduce such a trend, [48] was largely a result of geographically biased site selection favoring marine records from the North Atlantic and is not seen in terrestrial records [49]. Nevertheless, there are situations in which it is important to screen records to assess their suitability to answer specific research questions.

For example, low-resolution records or records with comparatively poor dating are clearly inadequate to examine the duration of short-lived events such as the Little Ice Age (LIA), but could be adequate for documenting the large changes from glacial to interglacial states. For this reason, Oster et al. ([45], this issue) only included records with at least decadal resolution for their assessment of speleothem  $\delta^{18}\text{O}$  variability in North and Central America for the last 2,000 years. The metadata fields in the SISAL database are designed to enable the screening of records prior to any cross-comparison on a regional or global scale.

The precision and accuracy of the ages obtained with U/Th dating methods depend on the analysed samples and the technique used. Low concentrations of U and Th require larger samples, usually implying that material of varying ages is mixed, which results in larger age uncertainties. This is particularly relevant for slowly growing speleothems, where U- and Th-concentrations may significantly vary across depositional layers. Another source of uncertainty is the correction of detrital  $^{230}\text{Th}$ .  $^{232}\text{Th}$  is used as an index of the initial amount of  $^{230}\text{Th}$  but, as the relationship between both is normally estimated, an uncertainty is transferred to the U-series ages. This is the case especially for 'dirty' samples with high  $^{232}\text{Th}$  concentration and low Uranium. These sources of uncertainties may be behind the age reversals found by Oster et al. ([45]; this issue) when analysing the precision of the age models constructed for the North Central America region.

The ability to analyse multiple records of screened raw speleothem isotopes from the same region permits the identification of records that are anomalous within their regional climatic context likely because the speleothem did not grow under equilibrium conditions. Such anomalous records need to be removed from reconstructions of regional isotope patterns and/or trends through time. Screening

records according to their mineralogy is also important when comparing  $\delta^{18}\text{O}$  values across records because the isotope fractionation between water (or the aqueous carbonate species) and  $\text{CaCO}_3$  is larger for aragonite than for calcite. There have been several estimates of this aragonite-calcite fractionation offset for  $\delta^{18}\text{O}$ , varying from 0.3‰ to 0.8‰ [50–52]. This inconsistency between estimates, given that the amplitude of the isotopic excursions linked to some global-scale climate events such as the Younger Dryas (YD) and the LIA is typically only ~0.5 and 2‰, means it is crucial that the speleothem records can be screened for mineralogy. Such screening has been applied by Lechleitner et al. ([42], this issue) when comparing the SISAL records with the Global Network of Isotopes in Precipitation (GNIP/IAEA) and by Kaushal et al. ([40], this issue), who highlight two examples (Valmiki Cave and Munagamanu Cave) in which this screening is required to derive robust regional patterns.

Changes associated with the infiltration of rainwater and water-soil-rock interactions such as degassing and prior calcite precipitation in the cave environment can also cause changes in  $\delta^{13}\text{C}$ ,  $\delta^{18}\text{O}$ , and trace element to Ca ratios [53–55]. These processes also modify speleothem growth rates by means of changes in the saturation state of the drip water [56]. Thus, another way to assess the suitability of individual speleothem records is by comparing the stable isotopes with other geochemical variables sampled along the same transect. Although the first version of the SISAL database only contains  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  measurements, it does indicate whether other types of measurements have been made on a specific speleothem to facilitate such comparisons. Kaushal et al. ([40], this issue), for example, used this information to retrieve cave monitoring data and data from measurements of bacterially derived branched Glycerol dialkyl glycerol tetraethers (GDGTs) for Mawmluh cave to help assess the quality of the record and to identify climate events.

## 5. Important Aspects to Consider When Interpreting Speleothem Records

A major advantage of speleothems is that they can be dated very precisely by U-series disequilibrium methods [57–59], which allows dating back to ca 500–600 ka. This is an advantage over other archives such as sediment cores, where the chronology is based on  $^{14}\text{C}$ -dating (and, therefore, only extends to about 50 ka) or on the calibration of individual events such as tephra layers occurring in different records via “wiggle matching” [60]. However, one of the main challenges encountered when analysing the SISAL data has been to incorporate temporal uncertainties into our calculations. Unfortunately, age uncertainties are frequently unreported or omitted in the original publications. This prevented an assessment of the 8.2 ka event in Western Europe where uncertainties were available only for two records ([42], this issue). Inclusion of age-uncertainty information is particularly worthwhile for older records so that they can be updated using currently standard age-depth model approaches. In addition, robust age uncertainties consistent with what is actually attainable from U/Th dates (i.e. 1%, 2-sigma) are not always reported. This issue is highlighted in Lechleitner et al. ([42], this issue), who found that, in many cases, the U-Th age uncertainties are larger than the uncertainties from the constructed isotope chronology, which indicates that these are often underestimated at least in the Western European records.

Infiltration of meteoric water from the surface to the cave can occur slowly by seepage and diffuse flow, or more rapidly by conduit flow. Depending on the preferred pathway, the mean drip discharge may vary and, as a result, drip sites within the same cave gallery may exhibit large variability [61]. Consequently, replication of speleothem records can be difficult. One example of this, pointed out by Kaushal et al. ([40], this issue), is the three records from Mawmluh Cave that have very different  $\delta^{18}\text{O}$  values for the 4.2 ka interval. The cross-comparison of records facilitated by the SISAL database allows non-replicating records to be identified so that inferred changes can be treated with caution.

Speleothem deposition is often continuous and high-resolution micro milling or laser ablation sampling techniques [62,63] allow measurements to be made at annual or even seasonal resolution. Such high-resolution measurements have been used to infer the speed of climate transitions. Chinese stalagmite records, for example, suggest that the Asian monsoon transition into the YD lasted 380 years [64], while the shift out of it took less than 38 years [65]. However, highly resolved samples do not necessarily record annual or seasonal climate conditions above the cave [66]. For this to happen, there

needs to be a strong climate signal at the surface, such as a seasonal monsoon, that is rapidly transferred to the speleothem. This can occur through direct interactions between surface and cave environments, such as seasonal cave ventilation affecting cave air  $p\text{CO}_2$  levels, or indirectly through water transfer of the surface signal to the speleothem e.g. via rapid conduit flow. In addition, the location of the speleothem is important. If it is too deep beneath the surface, mixing of waters within the karst can modify the surface signal and shift it in time [67], but if it is too shallow, speleothem growth may be inhibited because of the saturation state of the drip water. Since these conditions are rarely fulfilled, many annually resolved records in reality provide a signal that is mixed over considerably longer periods. Comparisons with local meteorological records [68] or cave monitoring programs [6,69] provide insights into the dynamics of a specific cave environment helpful in determining the likelihood that a speleothem record provides annually-resolved climate information [70]. However, while there are a growing number of such monitoring programmes, they are not ubiquitous, and it is, therefore, important to recognise the potential temporal distinction between registration and climate signaling.

For stalagmites to be straightforward archives of past rainfall  $\delta^{18}\text{O}$ , their isotopic composition must show a predictable relationship with infiltrated rainfall  $\delta^{18}\text{O}$  [71] despite the low-pass filter that is effectively applied to the speleothem series as a result of the water residence time in the subsurface. This is often referred to as “isotopic equilibrium,” which assumes a constant and predictable change in oxygen fractionation between water and carbonate with temperature. However, the complex controls and variability of drip water composition, even under constant climatic conditions [1], make it difficult to estimate the relationship between carbonate and water  $\delta^{18}\text{O}$  and cave palaeo-temperatures. In addition, the relationship between the measured  $\delta^{18}\text{O}$  or  $\delta^{13}\text{C}$  and the surface climate variable on which it primarily depends can be complex and can be modified before capture by the stalagmite [4,72,73]. Speleothem records, therefore, reflect a complex interplay between hydro-climate, temperature, circulation, and cave influences. The use of speleothem records for climate model evaluation will require the development of novel techniques and approaches for data-model comparisons.

## 6. Recommendations and Conclusions

Drawing from the experience with SISAL, we have identified several issues that, if taken on board by the community, would help improve the quality and re-usability of future speleothem studies (Table 1) as well as enable more robust science across research disciplines.

**Table 1.** Summary of opportunities for improvement identified during the construction and consequent analyses of the SISAL database.

Need/Issue	What Would This Enable Us to Do?
<i>Age Control</i>	
Better chronologies (with more radiometric dates well distributed across speleothem length to reduce an increased uncertainty in between dates). For example, if age-depth model is linear, the errors are not properly propagated in between U/Th ages — this occurs in c. 40% of the speleothem records in SISAL.	Avoid small biases in the age control of a time-series adversely affecting estimates of the timing and duration of short events such as YD, the 4.2 ka event, or the LIA.
Age-depth model uncertainties not always made available or used in comparisons with climate data or records from other archives.	Incorporate uncertainties in our climate reconstructions and/or calibrations with instrumental data in a sound manner.
<i>Mineralogy</i>	
Speleothem’s mineralogy is not always purposely reported – if mixed mineralogy, the percentages of aragonite versus calcite are not always given. Isotope correction from aragonite to calcite is not straightforward.	Examination of small isotope excursions linked to some events requires a robust isotopic record. A thorough assessment of the mineralogy would enable accurate comparisons of $\delta^{18}\text{O}$ across records.
<i>Interpretation</i>	

Assumption that speleothem laminae that have been postulated to form annually can be interpreted as an annual climate signal.	Laminae-based speleothem records may not correspond to annual climate signal due to the water transit time between surface of the cave and the drip site. This needs to be taken into account when using speleothem data.
The accuracy of the final chronology depends on the density of U/Th dates and the approach followed to construct the age-depth model.	Interpretation of $\delta^{18}\text{O}$ time series strongly depends on their temporal resolution and the accuracy of the age-depth model/chronology.
<i>Transparency and Reusability of Data (Metadata)</i>	
Depths and widths of measurements are usually not reported.	If they were, we would be able to reuse the data and re-calculate age-depth models with approaches that may not have been able at the time of the original publication.
Lamina counts are very difficult to get hold of.	If this information was made available, we would get a direct record of laminae thickness that could be used to complement that of $\delta^{18}\text{O}$ .
Most laminae counts have no uncertainties associated to them, even though this dating method is susceptible to missing or false bands.	The uncertainties of high-resolution palaeoclimate records are important for studies of annual-to-interannual dynamics of climate systems.
The definition of “BP” should be clearly stated. Some authors refer to the year when the U/Th chemistry column was done, while some others refer to 1950 CE.	Avoid adding extra errors when using already available data. Most important for young records where a ~50-year difference may make a big difference in their interpretation when the record is used by researchers other than the original authors.

As the regional papers in this Special Issue demonstrate, SISAL has made a major contribution to our ability to use speleothem records. Nevertheless, all the studies show that there are opportunities to improve the spatial and temporal coverage of speleothems records and, thus, the SISAL database. The inclusion of these missing sites as well as the creation of new stalagmite-based records in areas where these are lacking would strengthen our ability to draw strong conclusions about past climate changes as well as facilitate more robust comparisons with climate model simulations. There is a commitment from the SISAL working group to continue expanding the speleothem database in order to improve its coverage and to include other types of data from speleothems.

The SISAL initiative has shown there is a need for the adoption of common reporting practices, including metadata standards, to facilitate speleothem data being open, scrutinisable, accessible, reusable, and for quality control. The community-endorsed template used to upload speleothem records to the first version of the SISAL database (Supplementary Material) provides a model for this.

**Supplementary Materials:** The Excel workbook used for data entry to the SISAL database is available online at [www.mdpi.com/xxx/s1](http://www.mdpi.com/xxx/s1).

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