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SPELEOTHEM DEPOSITION IN EOGENETIC CARBONATES: THE CONSEQUENCES FOR STRONTIUM

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ABSTRACT

Eogenetic carbonate rocks develop from allochems precipitated primarily as aragonite (e.g. green algae, mollusks, corals) and high-Mg calcite. (e.g. echinoderms, red algae). Relative to calcite, strontium (Sr) preferentially enters the orthorhombic aragonite crystal lattice, giving eogenetic carbonate rocks a higher Sr background than older teleogenetic rocks, in which the aragonite has inverted to calcite. Vadose speleothems (stalagmites) forming in caves in eogenetic carbonate rock should show a high Sr content. The Sr levels in the speleothems should decrease with rock age as the aragonite inverts and the Sr is lost. Climate can also affect aragonite inversion, with wetter climates leading to more rapid inversion. Strontium levels in speleothems have been used as a paleoclimate indicator, but the influence of eogenetic Sr has been under appreciated. This issue has become more important as oceanic island caves and speleothems hosted in eogenetic carbonates have recently been utilized as mid-ocean paleoclimate indicators.

Curacao, in the southern Caribbean, contains a series of tectonically uplifted reef terraces of eogenetic carbonate rock. These carbonate rocks contain flank margin caves in which speleothem growth is active. Surface samples and speleothems from each terrace show a progressive loss of Sr in each higher, and therefore older, terrace. The "Higher Terrace," older mid-Pleistocene age surface rock, is calcitic, less than 5% aragonite, with 0.44 ppm Sr; one speleothem from a cave in this rock contained 0.25 ppm Sr. The "Middle Terrace," younger mid-Pleistocene surface rock, is also calcitic, less than 5% aragonite, with 1.43 ppm Sr; two cave speleothems had Sr values of 0.30 and 0.31 ppm. The "Lower Terrace" surface rock, late Pleistocene in age, was 40% aragonite and 60% calcite, with a Sr value of 6.89 ppm; two cave speleothems had Sr values of 8.68 and 8.28 ppm. Utilizing caves from a single location removes any climatic differences in calcite inversion and speleothem deposition, leaving rock age as the major factor in the Sr variation in these speleothems. In The Bahamas, collection was done along a climatic gradient of decreasing rainfall, NW to SE, but the inability to accurately determine host rock age made the results here equivocal.

INTRODUCTION

The presence of strontium (Sr) in carbonate rocks has long been documented (Kulp et al., 1962) and has been recognized as a contributor to the trace element content of speleothems used for paleoclimate reconstruction (e.g. Goede et al., 1998). Strontium in speleothems can originate from multiple sources although for the majority of current research, the Sr is assumed to be infiltrated from a surface source (e.g. Roberts et al., 1998; Finch et al., 2003; White, 2004; van Beynen et al., 2008; Sinclair et al., 2012). In younger, eogenetic carbonates, the mineralogy is aragonite and the aragonite crystal structure accommodates Sr in replacement of Ca (White, 2004; Hill and Forti, 1997). Over time, aragonitic carbonate rocks will invert to the more stable polymorph mineral of calcite (Hill and Forti, 1997). This inversion releases Sr to the vadose zone, as the Sr cannot be accommodated in the calcite crystal lattice of the host rock.

The carbonate host rocks that are mineralogically aragonite will naturally have a greater Sr content than those rocks that are calcite. The older eogenetic rock units of the Caribbean typically have a higher calcite to aragonite ratio than those that are younger (Gaffey et al., 1995). It can then be surmised that younger carbonates in the Caribbean should have a greater amount of Sr contribution from the host rock into those speleothems than speleothems that form in older carbonates that are inverted to calcite. While Sr does not fit easily into the calcite crystal lattice, it can be incorporated into the growth bands on the growing stalagmite to leave a record of the dripwater geochemistry.

Uplifted island carbonates, such as the carbonate units in the study area of Curaçao in the Netherlands Antilles (Figure 1A and 1B), are often terraced; where the oldest unit is at higher elevations and the youngest at lower elevations (Schellmann et al., 2004). These units are composed of coral reef carbonates that were originally composed of aragonite (Schellmann et al., 2004). The inversion of aragonite to calcite is not only time dependent, but is also dependent on the flux of meteoric water through the limestone and thus a climatic signature. Abaco Island in The Bahamas is the wettest island in the Bahamian archipelago (Figure 1C); Long Island and San Salvador, The Bahamas are two of the drier islands in the archipelago, having negative water budgets (Sealey, 1990). However, the islands are assumed to have carbonates of the same approximate age, so any difference in aragonite inversion degree should be a climatic signal as opposed to an age signal as for the island of Curaçao. This interpretation also assumes past climates are similar to those seen today.

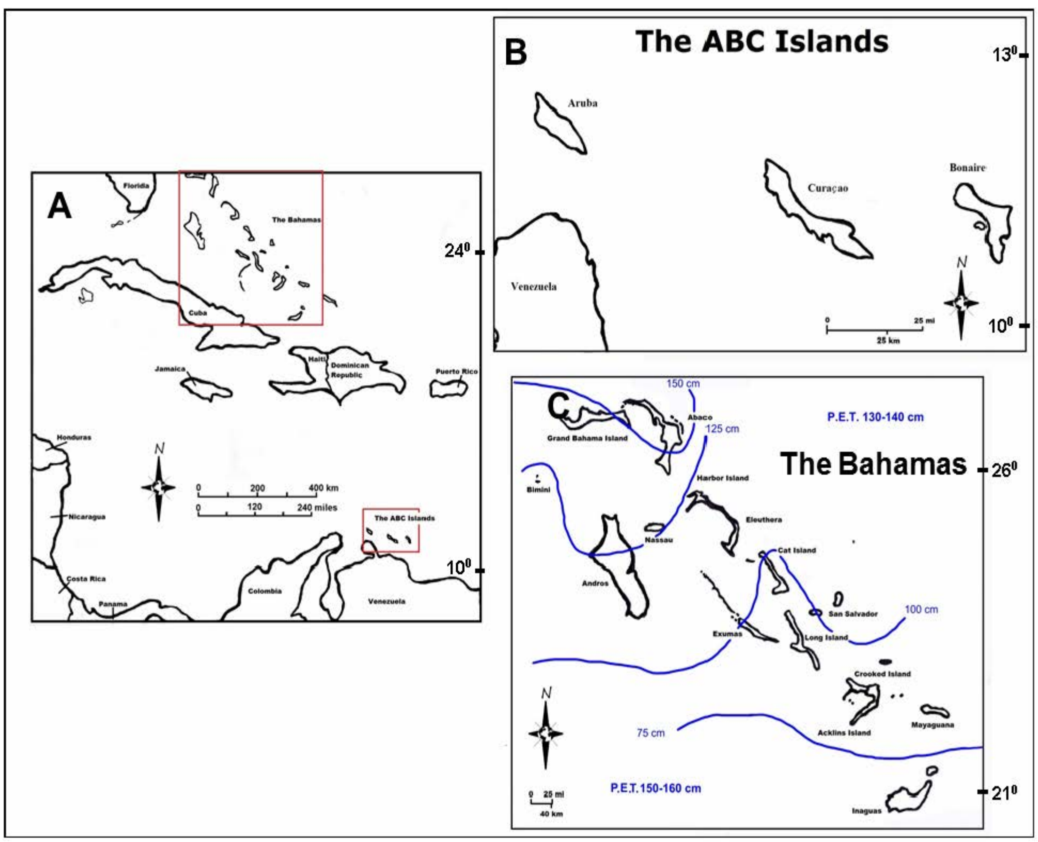

Figure 1: A. Caribbean and North Atlantic locations of the study sites. B. Curaçao is the middle of the three ABC Islands. C. Abaco, San Salvador, and Long Island trend across a climatic gradient (modified from Whitaker and Smart, 1997).

Speleothem research has become an integral part of paleoclimate reconstruction because cave deposits preserve the conditions that were present at the time of speleothem deposition. As cited earlier, most speleothem research seeks to interpret the local Earth surface conditions at the time of deposition, such as temperature, rainfall, vegetation. However, the state of the host rock itself can also be preserved in the speleothem record.

The research presented here sought to answer three questions: 1) does the Sr content of Caribbean speleothems have a direct relationship with the age of the host rock at the time of current speleothem precipitation? 2) do speleothems from older host rock contain less Sr than speleothems from younger host rock in the same climatic setting? and, 3) will speleothems record the change in Sr concentration of eogenetic carbonates as a faster depletion in climates of higher precipitation as opposed to drier climates?

Traditional work on speleothems has been concentrated on those located in much older geologic settings where the limestones are all calcite and Sr is more likely to be an infiltrate from an external source. The work on speleothems in younger, Sr-enriched host rocks will need to consider the possibly significant amount of host-rock sourced Sr within the speleothem record. This could affect the interpretations of paleoclimatic data from the use of speleothems in young carbonates. By determining the correlation of host-rock age with strontium concentration in the speleothem in contrast with the variation of strontium in differing climatic settings, a calibration could be developed to better interpret earlier paleoclimatic studies in young carbonates.

Recently, speleothem research has been shifting to oceanic islands to obtain speleothem climate records in mid-ocean areas not available on continents (e.g. Sinclair et al., 2012). The rocks hosting the speleothem-containing caves are commonly young and aragonitic, so the research for this project is timely and important. The work described here is preliminary. Other elements, such as Mg, were measured, but that work is still in progress. No age dating of sampled stalagmites is yet available, so time constraints remain unknown. Future work plans to complete a more robust data set and provide better interpretations using additional trace elements and age constraints.

METHODS

Fieldwork on the island of Curaçao took place in December, 2012, while the fieldwork on the islands of Abaco, Long Island, and San Salvador took place in June, 2013. Prior fieldwork on Curaçao conducted by Kambesis et al. (2015) and Sumrall et al. (2016) provided information on caves in these field areas that allowed for planning of sample collection. Due to the delicate nature of collecting and exporting cave samples, permission for six stalagmites (two each from three caves in each of the terrace levels) was obtained from the Carmabi Foundation for Curaçao which is equivalent to their national park service. In addition, permission for collecting multiple stalagmites from caves on Abaco Island, San Salvador Island, and Long Island was obtained from the Bahamas Environment, Science and Technology (BEST) Commission.

The Curaçao caves identified for collection were Lardem Cave in the Upper Pleistocene (MIS 5e) lower terrace; Raton Cave in the middle terrace of mid-Pleistocene age; and Hato Cave within the older and uppermost high terrace, also of mid-Pleistocene age. These caves are all flank margin caves that developed in the freshwater lens (Mylroie and Mylroie, 2007; Kambesis et al., 2015). For each terrace, the time window of speleogenesis was restricted to when the terrace was elevated and exposed to allow meteoric catchment to create a freshwater lens, but before the terrace was further uplifted and the freshwater lens was left behind. The stalagmites were carefully selected as the most optimum specimens within the caves (Figure 2A) and with growing apical tips (Frappier, 2008). They were collected from deep within the caves identified as primary candidates for this study to ensure humidity was high and airflow was minimal. Rock samples were collected from within the caves close to the speleothem sample sites. For comparison purposes, rock samples were also taken from exposed rock surfaces above each cave. All speleothem and rock samples from within the caves were collected from hidden locations to preserve the caves' internal appearance and aesthetics.

The Bahamian Archipelago is dominated by eolian calcarenites with subtidal deposits from the last interglacial (MIS 5e, ~120 ka) exposed occasionally at elevations below 6 m (Carew and Mylroie, 1995). The specimens from The Bahamas were collected from three islands that are located in areas that have differing water budgets measured by the balance between precipitation and evaporation (Figure 1C). Abaco, the northernmost island in the study area, has a positive water budget with the highest annual precipitation rate of the three islands. Long Island has a negative water budget with the least amount of annual precipitation. San Salvador has a negative water budget and annual precipitation that falls between values at Ababo and Long Island. The two caves that were identified as prime candidates for collection on Long Island are Salt Pond Cave and Hamilton Cave. Hole in the Wall Cave and Roadside Cave from the southern end of Abaco were sampled. And Lighthouse Cave was the San Salvador Island sample site.

All rock samples were carefully cut into blanks for the creation of thin sections (Figure 2B and 2C). The rock blanks were each cut with a wet saw with a diamond tipped blade manufactured for the purpose of cutting tile and stone. They were cut to specifications set by Spectrum Petrographics—the company chosen to create the thin sections. Each blank was notched with the wet saw to preserve the upright direction previously marked on the samples in the field. Rock samples from Curaçao were analyzed for composition using laser ablation and ICP-Mass Spectometry at the Rensselaer Polytechnic Institute. The Bahamas rock samples, as well as additional samples of the Curaçao rocks, were prepared in powder form and analyzed using the ICP-Optical Emission Spectrometer at Western Kentucky University. The preparation of the stalagmites was conducted at Western Kentucky University. Stalagmites were cut in half parallel to the growth axis, perpendicular to the growth layers. They were then evaluated for feasibility of study. Analyses, interpretation, and data appendices of all samples for this study can be found in Ridlen (2014).

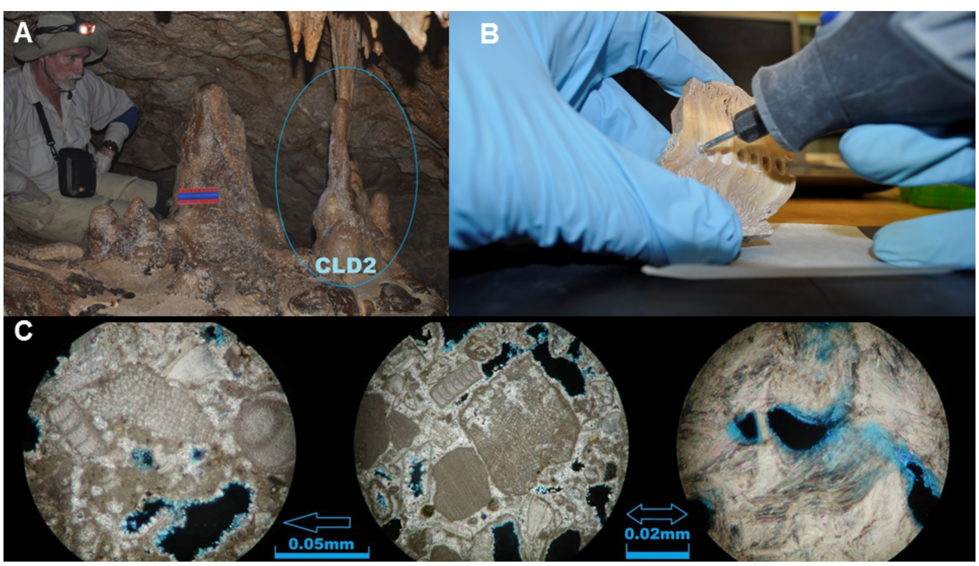

Figure 2: A. Collection site for sample CLD2 in Lardem Cave, Curaçao. Each sample site has a before and after image taken. B. Collection of powder sample every 0.5 cm from a stalagmite for analysis. C. Lardem thin sections. From left to right: cave wall rock, cave ceiling rock, and surface rock. Note the two thin sections on the right have a different scale from the thin section on the left.

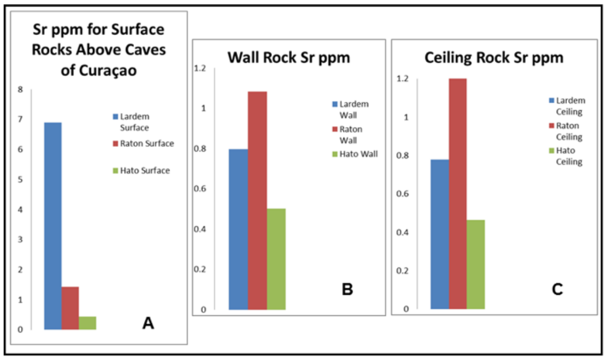

Figure 3. Results of strontium concentrations from rocks on Curaçao. A) Surface rocks, showing loss of Sr with increasing rock age, from youngest (Lardem) to oldest (Hato). B) Rock samples from cave wall rock, and C) Rock samples from ceiling rock within the caves, proximal to stalagmite collection sites. Lardem and Hato Cave follow the expected Sr trend, but Raton Cave is high, perhaps as a result of secondary speleothem aragonite which may have enriched Sr.

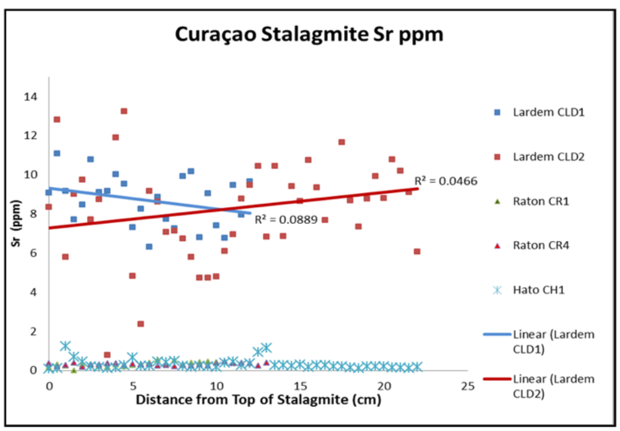

Figure 4. Stalagmite strontium concentrations, showing high levels in Lardem Cave but low levels in both Raton and Hato Caves on Curaçao.

RESULTS

Curaçao

Rock samples from within the lower terrace of Curaçao contained well-preserved fossils from inside the Lardem Cave (Figure 2C), whereas the rock specimen collected from the surface was highly altered by surficial weathering processes. In both cases, crystals growing into the pores of the rock from all directions indicated vadose conditions during subsequent post-deposition emergent crystallization. The caves in the higher terraces (middle and upper terraces) had progressively less well preserved fossils and altered pore spaces. The rock samples from within Raton Cave in the middle terrace revealed predominantly broken fossil pieces. The surface rocks showed lower Sr concentrations in the high terrace compared to the lower terrace; however, rock samples from within the cave tell a different geochemical story (Figure 3). Hato rocks seem overall depleted of Sr in comparison to the younger Raton and Lardem rocks. The Lardem cave rock samples, both wall and ceiling, seem to be overall lower in Sr concentrations than Raton.

The levels of Sr in the stalagmites from the Lardem Cave, located in the youngest eogenetic carbonate of this study, were notably higher than the Sr of Raton or Hato caves (Figure 4). The stalagmites in Raton and Hato caves were close to the same with very low Sr values (Figure 4). These data disagree with the high host rock values of Sr for Raton Cave, which suggests that the stalagmite-producing drip water comes from a surface rock source, and if secondary speleothem aragonite is present, it is not contributing to the drip water.

The majority of the stalagmites from Curaçao show a variation of Sr and Mg concentrations (Figure 5) progressing down the axis (from young apical to older basal material). The co-varying Sr and Mg concentrations are a topic of current research. The overall time span of stalagmite growth is not constrained, the degree of internal variation of Sr and Mg cannot be assumed to be a long-term trend. The data summary for Curaçao is shown in Figure 6.

The Bahamas

The increased meteoric water flux (Figure 1C) of the farthest north study area on Abaco island should deplete the speleothem Sr levels faster than

Long Island located in the southernmost part of the Bahamian study area. The concern for this part of the study was an accurate age for the host rock.

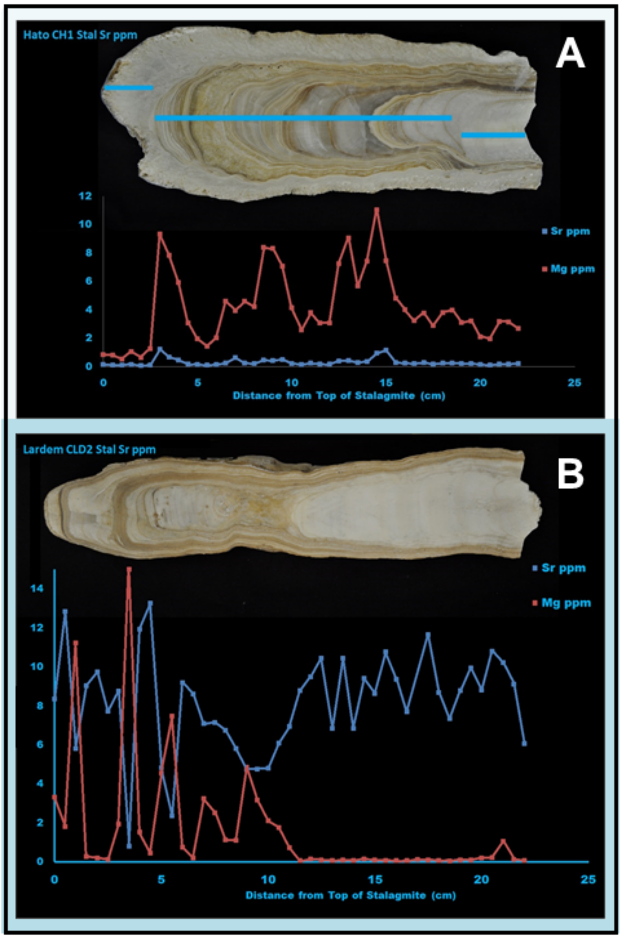

Figure 5. The Mg and Sr concentration from the stalagmites of Curaçao. A) Stalagmite (CH1) from Hato Cave in the oldest, highest terrace; Mg and Sr appear correlated. B) Stalagmite (CLD2) from Lardem Cave in the youngest, lowest terrace; Mg and Sr appear anti-correlated. Without age constraints, short-term variation in Sr and Mg cannot be separated from long-term trends.

The Bahamian Sr data for the stalagmites are variable (Figure 7). The stalagmite from Lighhouse Cave on San Salvador (SL4) had very high Sr concentrations in comparison to the stalagmites from Abaco from Hole in the Wall Cave (AH1) and Roadside Cave (AR1). The samples closest to the top of the SL4 stalagmite were likely to be contaminated by the tidal influx of modern seawater in Lighthouse Cave. The Sr content of the stalagmite from Hamilton Cave on Long Island (LH1) had the lowest Sr content of all the Bahamian stalagmites.

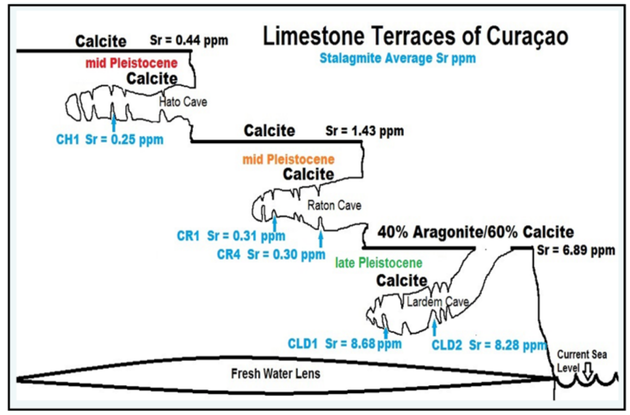

Figure 6. Summary data for Curaçao showing the decrease in Sr values with higher terrace levels and older surface rocks (which are the Ca and Sr source for current stalagmite growth in the caves). The stalagmites show a similar Sr decrease with higher elevations and thus increase in age of the host rock. Also shown are the locations of current sea level and the modern fresh water lens.

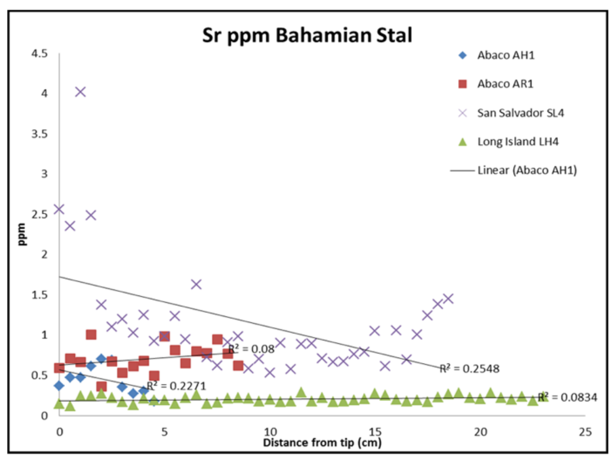

Figure 7. Results of strontium concentrations from stalagmites in The Bahamas. Abaco and San Salvador show expected Sr results, but the extremely low Sr concentrations for Long Island were not expected, perhaps as a result of age uncertainties for the host rock.

The rocks of the Bahamian study areas are equally as chaotic as the stalagmite data (Figure 8). As with Curaçao, the surface rocks followed the expected pattern, with Sr concentrations lowest in the wet northern Bahamas, and the highest Sr values for the dry southern Bahamas (Figure 8A). In contrast, the rocks from within Hamilton Cave on Long Island were relatively low in Sr concentration (Figure 8B and 8C). Sr concentrations of speleothems from caves on Abaco and San Salvador were as expected relative to one another. Abaco has a lower Sr concentration in the cave rocks than San Salvador, hypothetically due to the higher annual precipitation and larger water flux on Abaco. The data for the cave rocks of Long Island show depleted Sr.

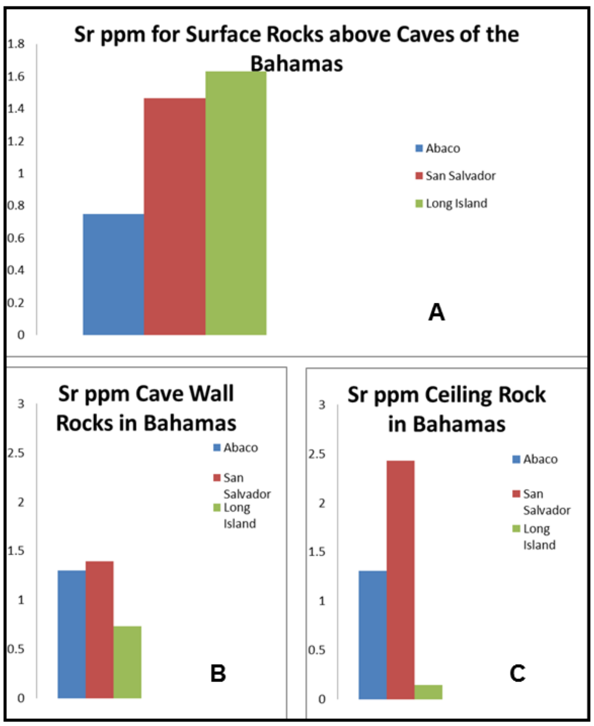

Figure 8. Results of strontium concentrations from Bahamian rocks. A) Surface rocks showing retention of Sr in the drier localities of San Salvador and Long Island. B and C) Wall and ceiling rocks show the expected increase in Sr for dry San Salvador versus wet Abaco. However, the Long Island data show unexpected loss of Sr; rock age is uncertain but the high Sr values for surface rocks makes these results confusing.

DISCUSSION

This study sought to find evidence of the Sr concentrations in speleothems deposited in eogenetic carbonate caves in relation to the relatively young mineralogy of the rocks. Three hypotheses with regards to Sr concentrations in speleothems of these settings were posed and described below.

Hypothesis 1. The Sr content of southern Caribbean speleothems has a direct relationship with the age of the host rock at the time of speleothem precipitation.

The stalagmites collected from within the youngest cave rock unit on Curaçao had a significantly higher concentration of Sr than currently growing stalagmites collected from the older carbonate terraces. The growing stalagmites in the caves of older rock units had lower Sr values indicating that the Sr had already leached out of the bedrock as a result of aragonite to calcite inversion.

The host rock Sr data is a bit confusing. For Curacao, the surface samples have Sr levels that decrease with increasing age of the rock, as expected. However, the cave wall and ceiling rock samples show Sr levels higher in the Raton Cave terrace rocks than in either the older Hato Cave terrace rocks or the younger Lardem Cave terrace rocks. Speculating, this might be the consequence of speleothem aragonite formation (Onac et al., 2001) in rock pores in the Raton Cave terrace rocks as a result of elevated Mg/Ca ratios in vadose water, with the Mg sourced from sea spray. This secondary speleothem aragonite could have trapped Sr that otherwise would have left the host rock. This speculation does not address the Sr depletion in the Hato Cave terrace rocks; it appears that the secondary aragonite precipitated in the host rock must later mobilize and be removed with the Sr. The same pattern holds for The Bahamas, but the age constraints there are weak so no conclusions can be drawn nor viable speculations made. The fact that the stalagmites show a Sr concentration pattern consistent with the host rock surface samples is an indication that the drip water is sourced from the surface and near-surface epikarst.

Hypothesis 2. Growing speleothems in older rock contain less Sr than speleothems in younger rock in the same climatic setting.

The stalagmites collected from Curaçao were all obviously from within the same climate with no variation in precipitation or temperature. The only difference in these stalagmites are the times at which they were deposited and the age of the rocks that hosted the cave they were deposited in. The Lardem stalagmite could not be older than 115 ka at the end of the MIS 5e interglacial (Thompson, et al., 2011) and the last time the fresh-water lens was at an elevation to permit speleogenesis of the cave. Dating would be needed to ascertain if the age of the Hato and Raton stalagmites are younger than those in the Lardem Cave, but they did have the opportunity to begin depositing long before Lardem Cave even existed. Their currently growing apical tips should record the same time record conditions as Lardem Cave's currently growing apical tips. In any case, the host rock is older. The Lardem stalagmite clearly has more Sr than the Raton and Hato stalagmites, but further investigation is necessary to determine the relative basal age of the two stalagmites from Raton Cave and the stalagmite from Hato Cave.

Hypothesis 3. Eogenetic carbonates within climates of higher precipitation will lose Sr content faster than those in climates of lower precipitation and the speleothems deposited in the caves within these rocks will record this trend.

The stalagmites from caves on the island of Abaco clearly show a lower Sr content than the stalagmite collected from the island of San Salvador. However, Long Island shows extremely low Sr concentrations in stalagmites, contrary to expectations (Figure 7). The cave rock (Figure 8) and stalagmite from Long Island seemed to be leached of Sr despite its dry climatic setting. Given that the surface rocks on Long Island have high Sr values, these results are unexpected. Unlike Curaçao, the high Sr values in surface rocks on Long Island do not appear to contribute Sr to the stalagmites in Hamilton Cave. These Long Island Sr values could be speculatively explained by either an unknown difference in age of the rock or the thickness of the rock unit where the cave is located, or perhaps a shift in glacial to interglacial climate. Carbonate rocks from the last interglacial (MIS 5e) deposited as the Grotto Beach Formation are easily identified based on their relationship with datable subtidal facies in The Bahamas (Carew and Mylroie, 1995). The eolianites of the Owls Hole Formation date from MIS 7, 9, 11 and possibly older, and are very hard to date, especially in the field (Carew and Mylroie, 1995). As the flank margin caves in The Bahamas are formed in these Owls Hole Formation eolianites, the host rock age cannot be constrained (Mylroie and Mylroie, 2013). Therefore, the climate-induced differences in aragonite (and hence Sr preservation) cannot be separated from a possible age-induced aragonite differences. The third hypothesis remains unproven.

SUMMARY

The study indicates that stalagmites growing in eogenetic rocks on Curaçao contain a Sr record that reflects the age of the host rocks at the land surface. Older, eogenetic rocks that have inverted mostly to calcite, create stalagmites with less included Sr compared to stalagmites in caves in the youngest rock. As the caves are all in the same climatic setting, the stalagmite Sr values reflect the age of the eogenetic host rock. These data have implications for studies using speleothem Sr values for paleoclimate reconstructions in young oceanic carbonate islands. Analysis of host rock Sr values within the caves indicates some storage mechanism, perhaps re-precipitated speleothem aragonite at the Raton Cave site. The Bahamian data, while similar to the Curaçao data, are not properly constrained as to host rock age, and therefore, the age versus climatic control of Sr flux cannot be established.

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