

Speleothem Evidence for Changes in Indian Summer Monsoon Precipitation over the Last ~2300 Years

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Speleothems from a well-ventilated dolomitic cave in the Pokhara Valley, central Nepal, preserve a mineralogic record of Indian summer monsoon variability over the past 2300 yr. Annually deposited aragonite layers formed between 2300 and 1500 yr B.P., indicating reduced monsoon precipitation and increased cave aridity, whereas alternating calcite/aragonite laminae deposited after 1500 yr B.P. record elevated summer monsoon precipitation and increased cave humidity. Dense, optically clear calcite layers deposited from 450 ± 5 to 360 ± 20 yr B.P. (1550 to 1640 A.D.) indicate a less-evaporative cave environment and suggest moister and/or cooler conditions, possibly related to climatic change associated with the onset of the Little Ice Age. © 2000 University of Washington.

Key Words: speleothem; India; monsoon; Little Ice Age; late Holocene.

INTRODUCTION

The Indian summer monsoon is driven by development of atmospheric low pressure cells over southern Asia in response to elevated heating of the land surface relative to the Arabian Sea. These pressure gradients draw moist air masses landward during the summer, supplying most of the annual precipitation to India, Nepal, and the southern Himalaya. During the winter, flow patterns are reversed, and cool, dry air dominates over

south-central Asia (Gregory, 1989; Clemens *et al.*, 1991). Long-term fluctuations in monsoon intensity are linked to glacial/interglacial cycles and orbital forcing (Duplessy, 1982; Van Campo *et al.*, 1982; Clemens *et al.*, 1991; Overpeck *et al.*, 1996), and several continental climate proxies indicate pronounced shifts in Indian summer monsoon intensity throughout the Holocene (Fig. 1). These include studies of pollen (Liu *et al.*, 1998) and ice chemistry (Yao and Thompson, 1992; Thompson *et al.*, 1997) of the Tibetan Dunde ice cap, pollen and lake sediments from northern (Sharma and Singh, 1974; Mazari *et al.*, 1996) and western India (Bryson and Swain, 1981; Enzel *et al.*, 1999), alluvial sequences from western, eastern, and northeastern India (Williams and Clark, 1984), and multiproxy investigations of Tibetan lakes (Van Campo and Gasse, 1993; Fontes *et al.*, 1996; Hui *et al.*, 1996; Van Campo *et al.*, 1996).

Less-well understood is short-duration late Holocene monsoon variability (Gasse and Derbyshire, 1996; Overpeck *et al.*, 1996). Pollen sequences in northeastern India indicate that cool and dry conditions between ~2000 and 1000 cal yr B.P. were followed by a warmer, wetter interval from ~1000 to 400 cal yr B.P. (Mazari *et al.*, 1996) (Radiocarbon ages from previously reported studies have been converted to cal yr B.P. using the CALIB radiocarbon calibration program; Stuiver and Reimer, 1993). The pollen sequence from Dunde ice cap records wet intervals between 2700 and 2200, 1500 and 800, and 600 and 80 yr B.P. (Liu *et al.*, 1998). Peats from southern India

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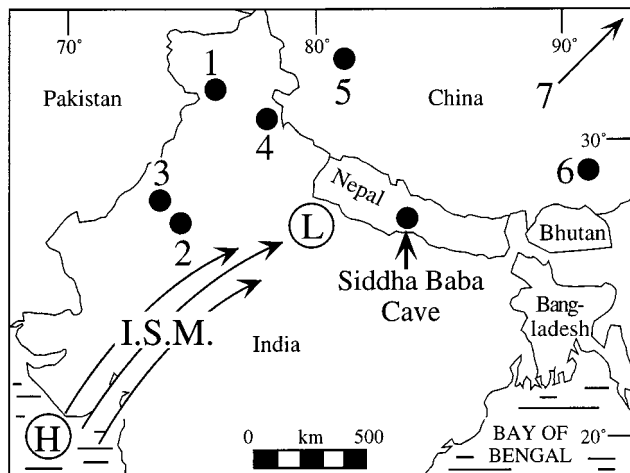


FIG. 1. Map of northern India and adjacent regions showing the position of Siddha Baba cave and other continental paleoclimate records containing evidence of Holocene monsoon activity. 1, Khajiar Lake and Rewalsar Lake; 2, Lake Didwana; 3, Lake Lunkaransar; 4, Takche Lake; 5, Sumxi Co. and Longmu Co.; 6, Qinghai-Xizang Plateau sediments; 7, Dunde ice cap (see text for references). The Indian summer monsoon (I.S.M.) develops in response to the development of low pressure systems (L) over the Tibetan Plateau relative to higher pressures (H) over the Arabian Sea.

(Sukumar *et al.*, 1993) indicate sudden shifts toward wetter conditions at approximately 3000, 1500, and 900 cal yr B.P. Analysis of pollen in alluvial and lake sediments from southwestern China suggest dry and cold conditions from 3200 to 1350 cal yr B.P., mild conditions between 1350 and 300 cal yr B.P., and cold climates again from 300 cal yr B.P. to present (Fu-Bao and Fan, 1987).

While evidence exists for large-scale interrelated climate shifts, including a proposed teleconnection via the East Tropical Jet that links Tibet and northern tropical and subtropical Africa (Van Campo and Gasse, 1993), considerable variability exists in the nature, timing, and lateral extent of monsoon activity. Temporal and/or geographic differences in monsoon character are not unexpected and have been described from historical records (Gregory, 1989). In addition, regional paleoclimatic records also illustrate this variability. For example, Lunkaransar Lake and Didwana Lake in Rajasthan, northwestern India, underwent large-scale fluctuations in water levels throughout the Holocene. However, Didwana Lake decreased significantly in size during the first 60% of the Holocene while Lunkaransar Lake did not undergo such a clear decline. In addition, Lunkaransar Lake desiccated at ~5000 cal yr B.P. (Enzel *et al.*, 1999), whereas Didwana Lake became dry at ~2500 cal yr B.P. (Bryson and Swain, 1981).

The use of speleothem mineralogy for paleoclimatic reconstruction has been previously proposed (Bar-Matthews *et al.*, 1991; Railsback *et al.*, 1994; Genty and Quinif, 1996). Carbonate rocks host numerous caves across much of central Nepal (Baeumler and Gebauer, 1992); thus, speleothems pro-

vide excellent geographic coverage of an area with limited paleoclimatic records. Here we present a precisely dated, high-resolution mineralogic record from stalagmites that preserves Indian monsoon variability during the last ~2300 years. Two speleothems, SB-18 and SB-19, were collected from Siddha Baba cave near the town of Kheireni in the Seti River Valley of the Pokhara Valley, central Nepal (lat. is 27°59'N, long. is 84°04'E, elevation is ~2000 m), a region where the climate is dominated by the Indian summer monsoon (Fig. 1). The entrance to Siddha Baba has an area of ~4 m² and is located 10 m above a small stream. The cave is formed in the lower Paleozoic Dhading dolomite and is composed of two, quasi-circular rooms, the first of which is equal in elevation to the entrance and is approximately 40 m². A second, larger room is entered by ascending 4 m through a 1-m² subvertical passage. All areas of the cave were dry when visited in December, 1995, only 4 months after the termination of the summer monsoon, suggesting that the rocks hosting the cave do not act as significant reservoirs of summer monsoon moisture, and that infiltration to the cave is strongly seasonal.

ANALYTICAL METHODS

The stalagmites were sawed in half along the long axes of deposition and were polished. Samples for dating were drilled from along the vertical growth axis using a modified dental drill. Sample sizes approached 200 mg for thermal ionization mass spectrometry (TIMS) and ~800 mg for alpha spectrometry. Uranium-thorium TIMS analysis was performed at the Radiogenic Isotope Laboratory at the University of New Mexico. Samples were dissolved in HNO₃ and then spiked with ²²⁹Th, ²³³U, and ²³⁶U. Chemical separation was modified from Chen *et al.* (1986). The U and Th were measured on a Micro-mass Sector 54 thermal ionization mass spectrometer with a high-abundance sensitivity filter. All isotopes of interest were measured on an ion-counting Daly multiplier with abundance sensitivity in the range of 20 ppb at one mass distance in the mass range of U and Th, requiring very little background correction, even for samples with large ²³²Th content. Multiplier dark noise was about 0.12 counts per second. The NBL-112 U standard was measured during the course of this study and remained in the range of 0.1% of the accepted ²³⁴U/²³⁸U ratio.

Alpha spectrometry was performed at the University of Iowa. Samples were dissolved and spiked with ²²⁸Th and ²³²U. Chemical separation techniques were modified from Chen *et al.* (1986). U and Th were electroplated on stainless steel disks using ammonium chloride and ammonium oxalate plating solutions and were counted for 4 to 8 days depending on activity levels. Isotopic compositions determined by alpha spectrometry agree well with isotopic ratios obtained with mass spectrometry (Table 1). All U-series ages are reported with 2σ uncertainties.

TABLE 1
Uranium and Thorium Isotopic Ratios and $^{230}\text{Th}/^{234}\text{U}$ Ages

Sample (mineral) ^a	mm from bottom ^b	^{238}U ($\mu\text{g/g}$)	^{232}Th (ng/g)	$\delta^{234}\text{U}^{c,d}$ measured	$^{230}\text{Th}/^{238}\text{U}$ activity	$^{230}\text{Th}/^{232}\text{Th}$ atomic	Age ^e	Dating method
SB-18 (a)	460	12.4	0.30	-58.5 (14)	3.14×10^{-3} (20)	2.16×10^{-3} (17)	360 (20)	TIMS
SB-18 (a)	440	21.4	1.14	-56.3 (32)	3.92×10^{-3} (5)	1.21×10^{-3} (2)	450 (7)	TIMS
SB-18 (a)	182	6.70	b.d. ^f	-39.3 (11)	1.42×10^{-2} (21)	b.d.	1590 (240)	alpha
SB-18 (a)	135	12.6	b.d.	-8.40 (20)	1.71×10^{-2} (23)	b.d.	1890 (250)	alpha
SB-18 (a)	2	16.8	b.d.	-32.6 (65)	2.32×10^{-2} (20)	b.d.	2640 (300)	alpha
SB-19 (l)	321	6.94	15.84	-76.9 (20)	2.52×10^{-3} (9)	1.82×10^{-5} (6)	230 (30)	TIMS
SB-19 (a)	307	6.73	b.d.	-35.8 (30)	1.10×10^{-2} (26)	b.d.	1240 (310)	alpha
SB-19 (l)	283	2.93	20.67	-42.2 (25)	1.67×10^{-2} (5)	3.89×10^{-5} (13)	1490 (120)	TIMS
SB-19 (a)	277	8.67	1.28	-41.4 (15)	1.32×10^{-2} (2)	1.48×10^{-3} (3)	1510 (20)	TIMS
SB-19 (a)	50	5.21	b.d.	-43.0 (12)	1.71×10^{-2} (9)	b.d.	1960 (320)	alpha
SB-19 (l)	5	8.51	8.40	-19.4 (19)	2.07×10^{-2} (1)	3.45×10^{-4} (2)	2290 (20)	TIMS

^a Mineralogy abbreviations: a, aragonite; c, calcite; l, aragonite/calcite laminae.

^b Total length of SB-18 = 512 mm; SB-19 = 331 mm.

^c $\delta^{234}\text{U}_{\text{measured}} = [(^{234}\text{U}/^{238}\text{U})_{\text{measured}} / (^{234}\text{U}/^{238}\text{U})_{\text{eq}} - 1] \times 10^3$, where $(^{234}\text{U}/^{238}\text{U})_{\text{eq}}$ is the secular equilibrium atomic ratio: $\lambda_{238}/\lambda_{234} = 5.472 \times 10^{-5}$.

^d Values in parentheses represent 2σ errors in the last significant figure. In all cases, age uncertainties are equal to or less than the number of annual bands sampled for dating.

^e Unsupported ^{230}Th was subtracted using an initial $^{230}\text{Th}/^{232}\text{Th}$ ratio of 8.5×10^{-6} ($\pm 2.125 \times 10^{-6}$). This value was obtained by correcting SB-19-283 in order to place it in correct stratigraphic sequence relative to the precisely dated SB-19-277 which underlies it.

^f b.d., below detection using alpha spectrometry.

Mineral phases were identified using optical, chemical (Feigl's solution) (Friedman, 1959), and X-ray diffraction (XRD) techniques. XRD analyses were performed on ~ 3 mg of powder using a Philips APD 3720 X-ray diffractometer and were used for qualitative assessment of speleothem mineralogy.

CLIMATIC CONTROLS ON SPELEOTHEM MINERALOGY

Siddha Baba stalagmites are largely aragonite. Aragonite speleothems occur in caves associated with dolomite (González and Lohmann, 1988) and are linked to arid cave conditions (Murray, 1954; Thrailkill, 1971; Bar-Matthews *et al.*, 1991; Railsback *et al.*, 1994). Dissolution of dolomite bedrock produces drip waters with initial Mg/Ca ratios ranging from ~ 0.6 to 0.9 (depending on the stoichiometry of the host dolomite) (Thrailkill, 1976; Evens *et al.*, 1986; González and Lohmann, 1988; Turin, 1995) and can exceed 1.0 in response to preferential removal of Ca^{2+} during precipitation of calcium carbonate mineral phases (e.g., along stalactites or in the vadose zone overlying the cave) (Möller and Kubanek, 1976; González and Lohmann, 1988). Numerous studies have demonstrated that Mg inhibits calcite crystallization while not affecting aragonite crystallization (e.g., Bischoff, 1968; Berner, 1975; Fernández-Díaz *et al.*, 1996; Deleuze and Brantley, 1997).

Recent work on speleothems, springs, and hydrothermal systems (e.g., travertines) has demonstrated that at temperatures consistent with cave drip waters, Mg/Ca molar ratios in excess of ~ 1.0 are required to precipitate aragonite (González and Lohmann, 1988; Folk, 1994; Morse *et al.*, 1997). In order

for aragonite to become the dominant mineral phase, however, higher Mg/Ca molar ratios are required; Möller and Kubanek (1976) report ratios of ~ 1.5 , González and Lohmann (1988) report ratios of 2.5, and Fischbeck and Müller (1971) found that a ratio of 4.4 is necessary. Only at temperatures in excess of $\sim 40^\circ\text{C}$ is aragonite favored over calcite at low Mg/Ca ratios (Folk, 1994; Deleuze and Brantley, 1997). In addition, evaporation increases the degree of saturation of the solution with respect to calcite, thus raising calcite precipitation rates and accelerating removal of Ca^{2+} from solution (Railsback *et al.*, 1994). However, while Ca^{2+} concentrations decrease, Mg^{2+} is concentrated in the remaining fluid. Thus, under evaporative conditions, Mg/Ca ratios increase more rapidly than during humid (less evaporative) conditions and aragonite tends to be favored (González and Lohmann, 1988; Bar-Matthews *et al.*, 1991; Railsback *et al.*, 1994).

Crystallization rates also affect the precipitation of aragonite and calcite. Growth rate is a function of degree of supersaturation (Chernov, 1984; Morse and Mackenzie, 1990; Fernández-Díaz *et al.*, 1996), and as the supply of CO_3^{2-} determines the degree of calcite supersaturation in many natural environments, when CO_3^{2-} is readily available the solution is more highly saturated and often favors aragonite as the predominant calcium carbonate mineral phase (Given and Wilkinson, 1985; Fernández-Díaz *et al.*, 1996). Because infiltration rate can impact drip-water chemistry (Bar-Matthews *et al.*, 1996; Baker *et al.*, 1997), shifts in cave hydrology can affect speleothem mineralogy. In addition, sudden loss of CO_2

can result in instantaneous aragonite supersaturation so that aragonite potentially can precipitate from non-Mg-bearing fluids (Chafetz *et al.*, 1991). At very high saturation states, growth rates can be sufficiently elevated so as to override the inhibitory effects of Mg^{2+} on calcite crystallization (Fernández-Díaz *et al.*, 1996). Thus, under extremely high precipitation rates, Mg-calcite (6 to 10 mol% MgCO_3) or high-Mg-calcite (>10 mol% MgCO_3) can crystallize.

In monsoonal climates, calcite crystallization is favored in many speleothems during the wet season because cave humidity is increased in response both to elevated infiltration and to less evaporative loss to the outside atmosphere, as well as to potentially lower calcite saturation states. Aragonite is favored during relatively dry intermonsoon months when the cave becomes more evaporative and slower infiltration rates allow for potentially higher calcite saturation states (Railsback *et al.*, 1994). A decrease in the intensity of precipitation during the monsoon period could therefore result in aragonite precipitation throughout the entire year.

RESULTS

The stalagmites were dated with precision 4 to 10 times better than radiocarbon or ice layer counting dates of similar age, or similarly sized calcite samples, because aragonite incorporates U more readily than does calcite; we observed U concentrations as high as 21 ppm. In addition, throughout the majority of the aragonitic portions of these speleothems, numbers of visible growth bands agree to within the error of radiometric dates, demonstrating that they are annually deposited—likely in response to monsoon cycles. Annual layering has been previously identified from speleothems in temperate regions based on annual laminae (Broecker and Olson, 1960; Genty and Quinif, 1996) and fluorescent banding (Baker *et al.*, 1993) and from monsoonal climates based on calcite–aragonite mineralogy (Railsback *et al.*, 1994). Age determinations are limited in the more calcite-rich portions of SB-19 (and to a much lesser degree, SB-18) by abundant illuviated clays with poorly constrained $^{230}\text{Th}/^{232}\text{Th}$ ratios.

SB-19 is composed of annually banded aragonite ranging in age from 1500 to 2300 yr B.P. Laminae were deposited in cycles, probably associated with the summer monsoon. A sharp, easily recognized transition from laminae composed exclusively of aragonite to laminae defined by distinct alternating layers of calcite and aragonite exists at 1500 yr B.P. (Figs. 2 and 3). No evidence for interrupted growth or recrystallization is observed at the aragonite to calcite/aragonite transition, either in hand samples or in thin sections. A small amount of calcite is visible below the transition from banded aragonite and calcite/aragonite laminae.

SB-18 is characterized by a complex growth geometry resulting from numerous changes in drip position. The last 1500 yr of SB-18 is more aragonitic than SB-19, and as a result,

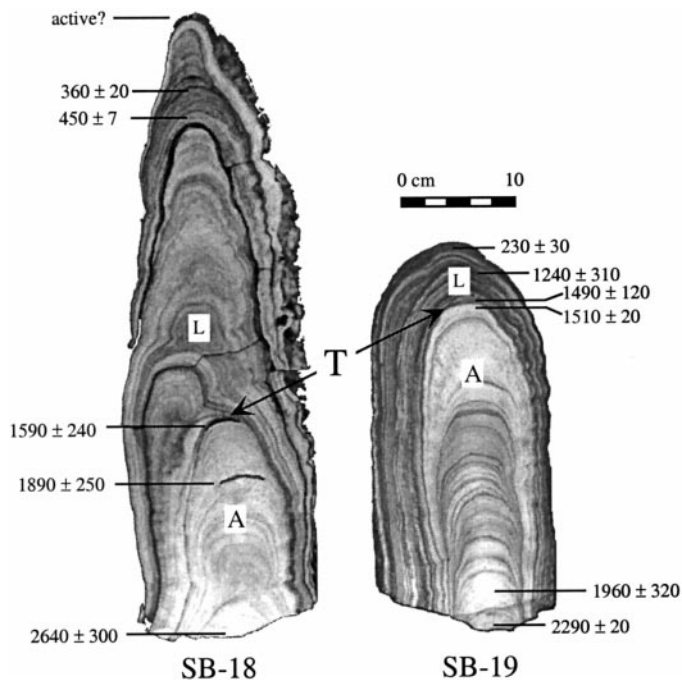


FIG. 2. Cross-sectional view of SB-18 (left) and SB-19 (right) demonstrating mineralogical changes and radiometric ages in years B.P. T, transition between aragonite (A) and calcite/aragonite laminae (L).

growth rates, and temporal resolution, are higher than in SB-19. Three dense, optically clear calcite layers exist in the upper 20% of SB-18 at 443 to 453, 455 to 457, and 460 to 462 mm. Two mass spectrometry dates bracket this part of the stalagmite and place these calcite intervals between 450 and 360 yr B.P. (1550 to 1640 A.D.). It is extremely difficult to assign tight age constraints to specific calcite laminae in SB-19, however, because the same time interval is compressed into about 10% of the distance in the vertical growth direction, and high ^{232}Th abundances limit the precision available by U/Th dating techniques. X-ray diffraction analysis demonstrates that the calcite portions of these speleothems are low-Mg calcite (<5 mol% MgCO_3).

DISCUSSION

Between ~2300 and 1500 yr B.P., Siddha Baba speleothems formed almost entirely as aragonite, indicating reduced drip rates and/or arid conditions throughout the year. The overall lack of alternating calcite laminae suggests that summer monsoon precipitation was diminished relative to the period after 1500 yr B.P. Sporadic calcite layers are present during this interval and may record anomalously wet summer monsoons in a period of overall elevated aridity. The mineralogic transition about 1500 yr B.P. records increased summer monsoon precipitation and heightened infiltration into Siddha Baba that elevated cave humidity during the monsoon season and dimin-

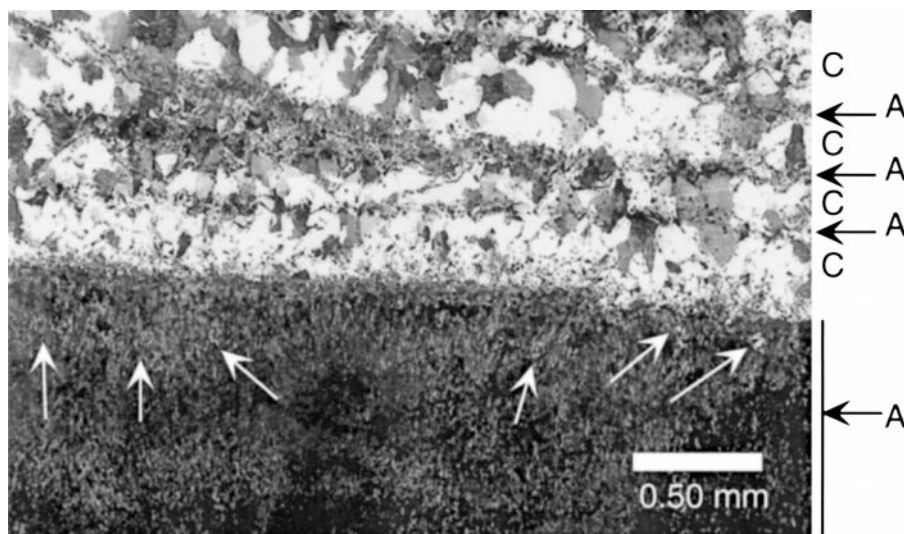


FIG. 3. Photomicrograph of SB-19 showing the transition from banded aragonite (bottom) to alternating calcite (C) and aragonite (A) laminae (top). Field of view is 3.2 mm $\times$ 2.1 mm. Arrows denote calcite intergrown with aragonite prior to primary mineralogic transition at 1500 yr B.P.

ished evaporation to the point that calcite crystallized from cave drip water. The sharp contact between mineralogic domains suggests that summer monsoon precipitation increased suddenly at $\sim$ 1500 yr B.P. The association of illuviated clays with calcite/aragonite laminae also argues for rapid changes in infiltration rate. However, it also is possible that infiltration rates and/or cave humidity were increasing gradually over a long time prior to 1500 yr B.P., but conditions appropriate for formation of calcite were not reached until 1500 yr B.P. Non-climatic factors that could also have influenced cave mineralogy, such as changes in infiltration hydraulics or cave geometry (e.g., closing off of the cave atmosphere by landslide blockage of the cave entrance), are not likely because (1) aragonite continued to crystallize during the relatively dry intermonsoon months and (2) illuviated clays are rare in aragonitic layers but are abundant in calcite/aragonite laminae, suggesting more rapid water flux through the overlying soil and bedrock.

During the late Holocene, monsoon circulation responded to climatic changes, including Neoglacial cooling (Liu *et al.*, 1998). Pollen and oxygen isotope studies of the Dundee ice cap in Tibet suggest that decreased summer temperatures played a more significant role than increased summer precipitation in elevating effective moisture during the late Holocene (Thompson *et al.*, 1997; Liu *et al.*, 1998). Our data indicate that in the Pokhara Valley, the last 1500 years witnessed greater monsoon precipitation than the preceding millennium, with wetter conditions resulting from a stronger summer monsoon. Pollen evidence from the Dundee ice cap also suggests that during the Little Ice Age (1660–1910 A.D.), the Tibetan Plateau was characterized by cool and humid conditions (Liu *et al.*, 1998). Our findings of dense calcite beginning at 450 ± 5 yr B.P.

(1550 A.D.) argue for cave conditions that were sufficiently cool and/or moist to suppress evaporation and allow calcite crystallization all year. This supports a climatic scenario similar to that proposed by Liu *et al.* (1998), but these speleothem data indicate that Little Ice Age climate change apparently began $\sim$ 100 yr earlier in the Pokhara Valley than at the Dundee ice cap.

CONCLUSIONS

Changes in speleothem mineralogy record shifts in the intensity of the Indian summer monsoon during the late Holocene. While the forcing mechanisms behind these changes remain unclear, gross similarity exists between regional climate records that suggest a shift toward more humid conditions $\sim$ 1500 yr B.P. Well-developed karst across northern India and the southern Himalaya may allow increased geographic coverage of monsoon variability as recorded in speleothems, and thus may help us assess the synchrony of changes in summer monsoon precipitation on a regional scale.

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