

CLIMATOLOGY

On the glacial-interglacial variability of the Asian monsoon in speleothem $\delta^{18}\text{O}$ recordsG. Liu^{1,2,3*}, X. Li⁴, H.-W. Chiang², H. Cheng⁴, S. Yuan^{1,2,3}, S. Chawchai⁵, S. He², Y. Lu², L. T. Aung^{2,6}, P. M. Maung^{2,7}, W. N. Tun⁶, K. M. Oo⁷, X. Wang^{2,3*}

While Asian monsoon (AM) changes have been clearly captured in Chinese speleothem oxygen isotope ($\delta^{18}\text{O}$) records, the lack of glacial-interglacial variability in the records remains puzzling. Here, we report speleothem $\delta^{18}\text{O}$ records from three locations along the trajectory of the Indian summer monsoon (ISM), a major branch of the AM, and characterize AM rainfall over the past 180,000 years. We have found that the records close to the monsoon moisture source show large glacial-interglacial variability, which then decreases landward. These changes likely reflect a stronger oxygen isotope fractionation associated with progressive rainout of AM moisture during glacial periods, possibly due to a larger temperature gradient and suppressed plant transpiration. We term this effect, which counteracts the forcing of glacial boundary conditions, the moisture transport pathway effect.

INTRODUCTION

The Asian monsoon (AM) affects the livelihoods of about half the global population. The AM variabilities have been widely inferred on the basis of oxygen isotope ratios ($\delta^{18}\text{O}$) in cave records, in particular those from Hulu, Sanbao, and Dongge caves in southern China, which all show $\delta^{18}\text{O}$ changed markedly on orbital time scales largely in response to the Northern Hemisphere summer insolation (NHSI), and abruptly on millennial time scales in close correspondence with North Atlantic climate events (1, 2). The Chinese cave records, unlike other monsoon proxy data (3–5) and global-scale paleoclimate records (6–9), are insensitive to global ice volume forcing, especially when considering their unexpectedly low $\delta^{18}\text{O}$ values during the Last Glacial Maximum [LGM; ~23 to 19 thousand years before present (ka B.P.; B.P. refers to 1950 A.D.) (10)] and relatively high $\delta^{18}\text{O}$ values during marine isotope stage 5e (MIS 5e) (Fig. 1) (1, 2).

This inconsistency has led to other interpretations, which instead suggest that the $\delta^{18}\text{O}$ records either reflect changes in the remote moisture source over the Indian Ocean (the so-called up-stream depletion mechanism) (11–13) or are a result of changing proportions of less ^{18}O -depleted moisture from the western Pacific that is mixed with moisture from the Indian Ocean (2, 4, 14, 15). To weigh in on the controversy, here we investigate cave $\delta^{18}\text{O}$ with a broader spatial coverage and report its changes over mainland Southeast Asia. Our records are based on stalagmites from three locations: southeastern Myanmar and western Thailand near the eastern coastal Bay of Bengal (CBoB), the western Shan Plateau in central Myanmar (CM), and southeastern Yunnan (SEY), China, close to the Myanmar-China border (Fig. 2 and figs. S1 and S2). These three locations form a SW-NE (southwest-northeast) transect along the moisture trajectory of the Indian summer monsoon (ISM), a major branch of the AM (16, 17).

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RESULTS

In total, for this study, we analyzed 19 stalagmites (fig. S3) from 11 caves (see Materials and Methods and table S1); we determined 164 dates using recently improved uranium/thorium dating techniques and constructed the chronologies of all samples (fig. S4) (18, 19); and we measured 2611 stable isotope values (figs. S5 and S6). The records from CBoB (fig. S7A) and CM (fig. S7B) cover a substantial part of the last 40 ka, including both the LGM and the most recent several hundred years. The spliced record from SEY extends almost continuously through the last 180 ka (fig. S7C).

Our three speleothem $\delta^{18}\text{O}$ records (fig. S7) broadly resemble the NHSI on orbital time scales, with more negative $\delta^{18}\text{O}$ values corresponding to higher insolation and vice versa. The records are also punctuated by abrupt $\delta^{18}\text{O}$ -positive shifts corresponding to the Younger Dryas and Heinrich event 1. However, the earlier millennial-scale events are less clearly shown, in part due to the coarse resolution of the records.

On glacial-interglacial time scales, our records show relatively more positive values during the glacial periods and more negative values during interglacial periods. For example, the CBoB $\delta^{18}\text{O}$ (fig. S7A) was ~-3 to ~-6‰ between ~18 and ~37 ka, but about -7.5‰ during the recent few hundred years. Similarly, the CM record (fig. S7B) presents $\delta^{18}\text{O}$ values ~-5‰ to ~-7‰ and ~-7.5‰ during the late glacial and late Holocene, respectively. The SEY record (fig. S7C) shows relatively more positive $\delta^{18}\text{O}$ values (~-8 to ~-11‰) during deep glacial periods (~160 to ~135 ka and ~75 to ~20 ka), compared with more negative $\delta^{18}\text{O}$ values (~-11 to ~-15‰) during interglacial periods (~125 ka and after ~10 ka).

DISCUSSION

The CBoB speleothem $\delta^{18}\text{O}$ as a proxy of ISM rainfall

Our speleothem $\delta^{18}\text{O}$ variability is similar to the trends in speleothem records from the ISM domain (figs. S8 and S9) (20–22), as well as broadly consistent with the records from the East Asian summer monsoon (EASM) domain (figs. S9 and S10) (1, 2), which, together with other lines of evidence (see Supplementary Text and table S2), suggests that the carbonate $\delta^{18}\text{O}$ in our samples primarily captured the rainfall $\delta^{18}\text{O}$ and cave temperature at the time of

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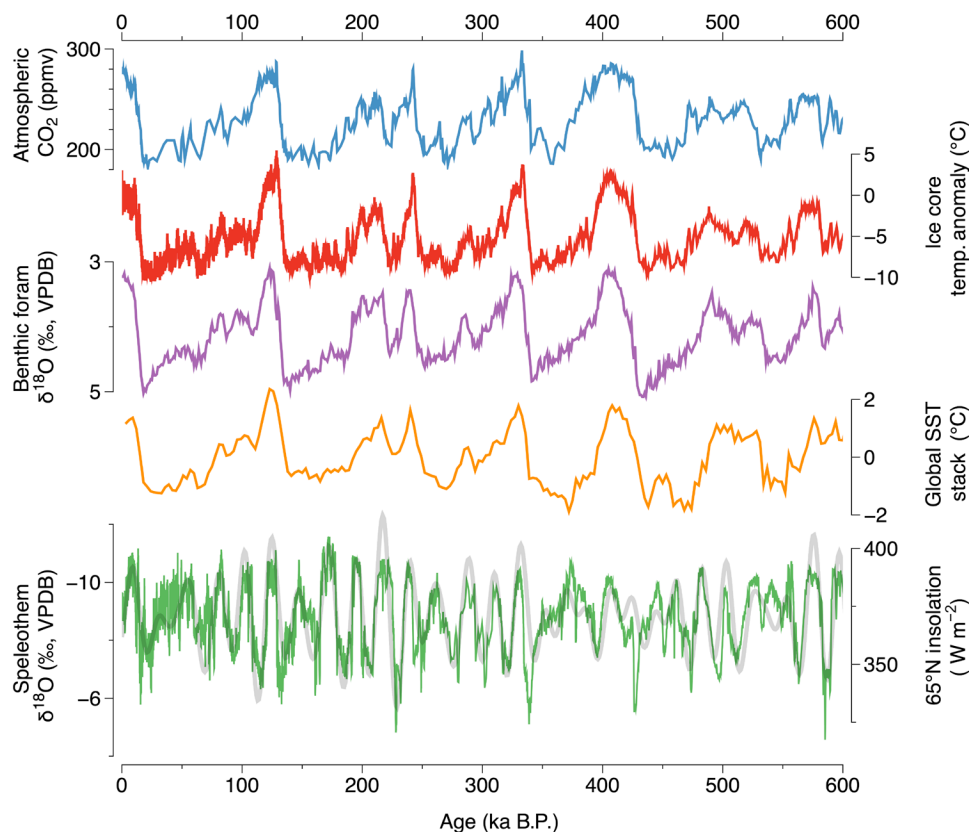


Fig. 1. Comparison of the Chinese speleothem $\delta^{18}\text{O}$ record and other major paleoclimate records. The compiled speleothem $\delta^{18}\text{O}$ record from southern China (green) (2) follows the 65°N summer insolation (gray) closely, and it is dominated by the 23-ka precessional cycle. However, variation in other major paleoclimate records, such as atmospheric CO_2 concentration (blue) (6) and Antarctic surface temperature anomaly (red) (7) retrieved from European Project for Ice Coring in Antarctica (EPICA) Dome C ice core, benthic foraminifera $\delta^{18}\text{O}$ LR04 stack (purple) (8), and global sea surface temperature (SST) stack (orange) (9), are dominated by the ~ 100 -kyr glacial-interglacial cycle. VPDB, Vienna Pee Dee belemnite; B.P., before the present.

carbonate precipitation. In addition, as the temperature in the region likely fluctuated $\sim 4^\circ$ to 5°C during the recent glacial cycles (23, 24), temperature-dependent fractionation accounts for only $\sim 1.0\text{‰}$ of the variability (25) in speleothem $\delta^{18}\text{O}$. A large portion of the variability in our samples must have resulted from changes in rainfall $\delta^{18}\text{O}$.

Rainfall $\delta^{18}\text{O}$ at our southern study site near the coastline is negligibly influenced by the continental effect and water recycling. $\delta^{18}\text{O}$ there is then primarily reflecting an isotope fractionation process close to that in the Rayleigh fractionation model. We, thus, argue that fluctuation of speleothem $\delta^{18}\text{O}$ at CBoB reflects AM rainfall or, more specifically, ISM rainfall.

The $\delta^{18}\text{O}$ fluctuation in our CBoB record closely matches the record from the Mawmluh Cave in northeastern India (fig. S8) (21, 26), which is also located close to the BoB. We therefore combined the two records by negatively shifting $\delta^{18}\text{O}$ in the Mawmluh record by $\sim 2.1\text{‰}$ to account for their difference in cave temperature (fig. S8). The combined record shows broad similarity to various planktonic foraminiferal records from the BoB (5, 27–29), in particular the foraminiferal *Globigerinoides ruber* $\delta^{18}\text{O}$ record from the northern BoB near the mouth of the Ganges-Brahmaputra rivers (Fig. 3 and fig. S1), which is also interpreted as fluvial input or ISM intensity (27). Hence, the resemblance further supports our interpretation of speleothem $\delta^{18}\text{O}$ as a proxy of ISM rainfall and, thus, its strength.

Reduced ISM rainfall during the LGM

The most prominent feature of the CBoB record is the 3.7‰ difference in $\delta^{18}\text{O}$ between the LGM and the recent several hundred years (Fig. 3, fig. S7A, and table S3)—one of the largest speleothem $\delta^{18}\text{O}$ differences in the AM region. A similar $>3\text{‰}$ difference is observable in the Mawmluh record (fig. S8) (21, 26). If we consider a 0.7‰ change resulting from temperature effect on isotope fractionation during cave calcite precipitation (table S3), the speleothem $\delta^{18}\text{O}$ shifts in both records are in accordance with a $>2\text{‰}$ increase in groundwater $\delta^{18}\text{O}$ recovered in Bangladesh and dates back to the LGM (30). If we further consider a $\sim 1\text{‰}$ change in source seawater $\delta^{18}\text{O}$ resulting largely from ice volume changes (fig. S11) (31), the remaining shift of $\sim 2\text{‰}$ in the CBoB record, together with an up to 2‰ increase in the BoB ice-volume-corrected surface seawater $\delta^{18}\text{O}$ (5, 23) during the LGM compared with the present, likely suggests a substantial decrease in monsoon rainfall amount during the LGM relative to the recent several hundred years.

We quantitatively estimate the reduction of rainfall amount using the Rayleigh distillation model, since the monsoonal moisture over the CBoB region comes dominantly from the tropical Indian Ocean, with almost no influence of the continental effect on rainfall $\delta^{18}\text{O}$. We assume that oxygen isotope exchanges remain in equilibrium state during water evaporation and condensation, and apply the

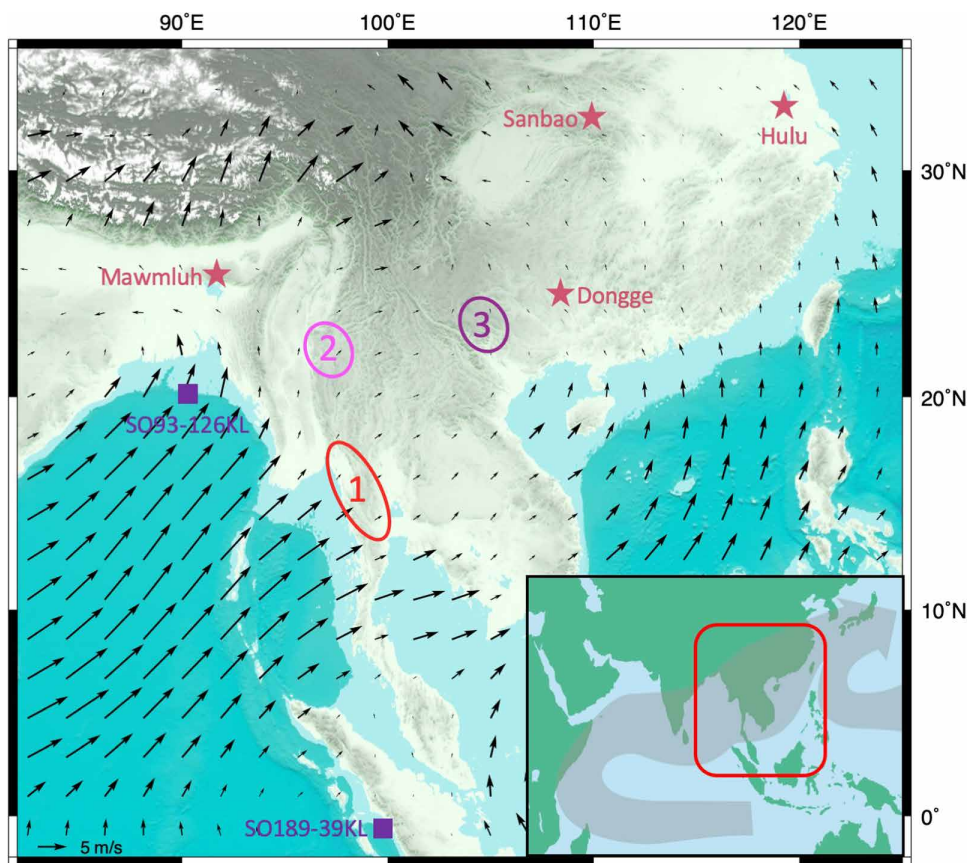


Fig. 2. Sample locations. The black arrows represent the present-day averaged June-July-August (JJA) wind pattern 10 m above sea level [data from the National Centers for Environmental Prediction/National Center for Atmospheric Research Reanalysis monthly mean (1981 to 2010) (58)]. The insert shows the AM system [modified after (59)]. The circled numbers mark the three strategic cave sites along the monsoon trajectory (1, CBoB; 2, CM; and 3, SEY). The stars mark the Mawmluh (21, 26), Hulu (60), Dongge (2, 61, 62), and Sanbao (1, 2) caves; and squares mark marine sediment cores SO93-126KL (27) and SO189-39KL (39) for reference.

following equation (32) to calculate the fraction of the original water vapor remaining in air masses (f)

$$(1000 + \delta^{18}O_p) / (1000 + \delta^{18}O_{sw}) = f^{\alpha(\text{liquid-vapor})-1} \quad (1)$$

where $\delta^{18}O_{sw}$ is seawater $\delta^{18}O$, $\delta^{18}O_p$ is the local precipitation $\delta^{18}O$, and $\alpha(\text{liquid-vapor})$ is the isotopic fractionation factor between water liquid and vapor phases. $\delta^{18}O_{sw}$ is assigned to be 0‰ [VSMOW (Vienna standard mean ocean water)] for today and 1‰ during the LGM (31). We calculate $\delta^{18}O_p$ based on the equation

$$\alpha(\text{calcite-H}_2\text{O}) = (1000 + \delta^{18}O_c) / (1000 + \delta^{18}O_p) \quad (2)$$

where $\alpha(\text{calcite-H}_2\text{O})$ is the isotopic fractionation factor between calcite and cave drip water and can be calculated from the equation (25)

$$1000 \times \ln \alpha(\text{calcite-H}_2\text{O}) = 17.66 \times 10^3 / T - 30.16 \quad (3)$$

$\delta^{18}O_c$ is a calcite $\delta^{18}O$ value, read from the CBoB speleothem record. $\alpha(\text{liquid-vapor})$ is the isotopic fractionation factor between water liquid and vapor phases, and we calculate it from the equation (33)

$$1000 \times \ln \alpha(\text{liquid-vapor}) = -7.685 + 6.7123 \times 10^3 / T - 1.6664 \times 10^6 / T^2 + 0.35041 \times 10^9 / T^3 \quad (4)$$

where T is the temperature (in kelvin).

Assuming that the temperature at CBoB was $\sim 4^\circ\text{C}$ lower in the LGM relative to modern temperature ($\sim 28^\circ\text{C}$), in concert with the observed and simulated sea surface temperature (SST) drop in the BoB (5, 23, 34, 35), our calculation shows that f changed from today's 54 to 68% during the LGM (table S3).

We then use the local precipitable water vapor (PWV; in kg/m^2 or mm) and the percentage of water vapor removed from air masses after reaching cave sites (i.e., $1 - f$) to calculate the precipitation amount at CBoB. We estimate PWV according to the equation (36)

$$\text{PWV} = 4932 \times \text{RH} \times e / T \quad (5)$$

where T is the temperature (in kelvin), RH is the relative humidity, and e is the saturation vapor pressure (in kPa), which we calculate from the equation (37)

$$e = 0.61121 \times e^{(17.368 \times T_c / (238.88 + T_c))} \quad (6)$$

where T_c is the temperature (in Celsius). Given a relative humidity of 100%, we can estimate the moisture removed from air masses relative to the modern value (set as a reference point at 100%) (table S3).

On the basis of our calculations, the 3.7‰ difference in speleothem $\delta^{18}O$ between the LGM and the recent several hundred years

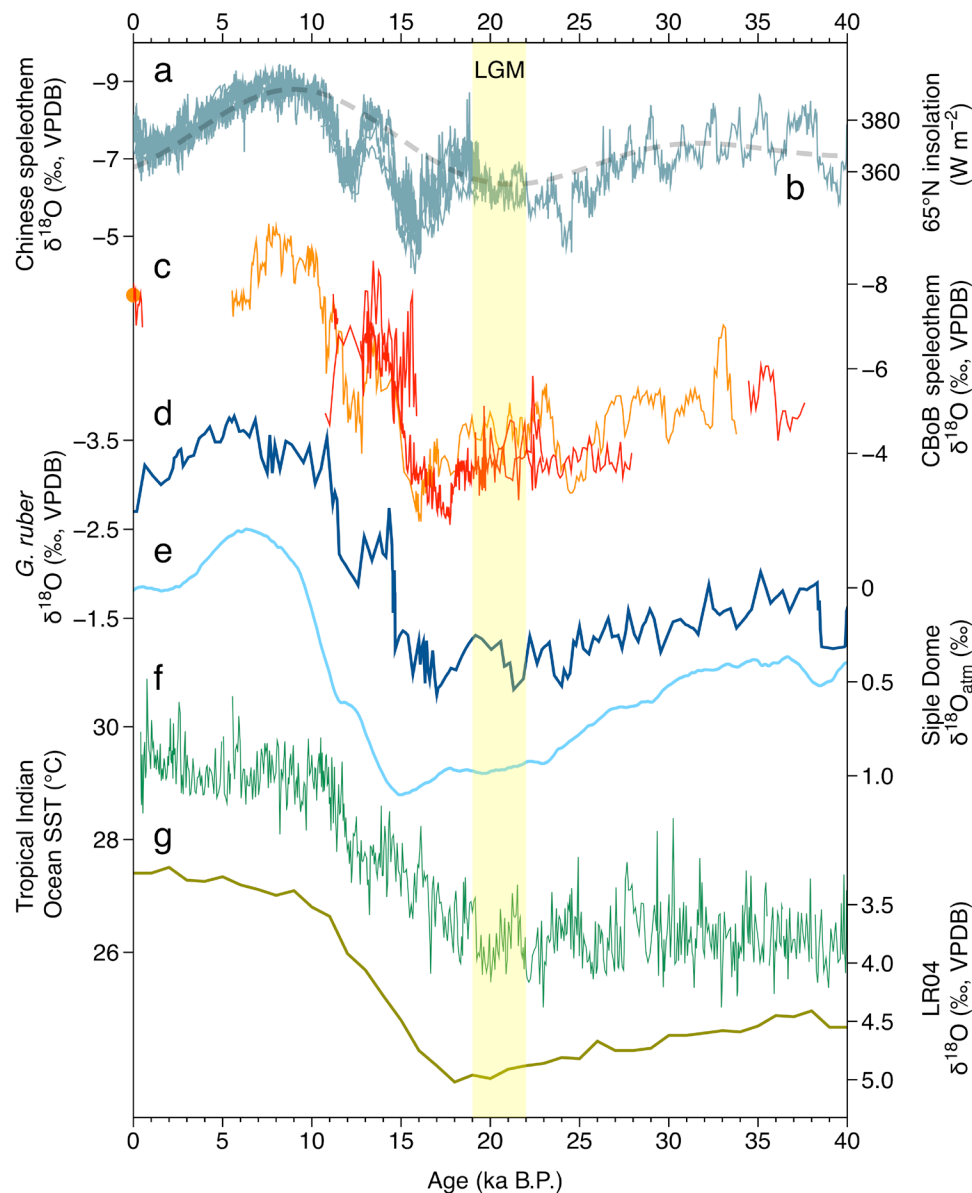


Fig. 3. Weakened ISM during the LGM and glacial boundary conditions. (A) The compiled speleothem $\delta^{18}\text{O}$ record from Hulu Cave (2, 60), Dongge Cave (61, 62), and Sanbao Cave, with the Sanbao record shifted positively by 1.6‰ to align it with the other two cave records from southern China (1). (B) The 65°N summer (JJA) insolation. (C) The combined speleothem records from CBoB (red) and Mawmluh (orange) (21, 26) caves. The Mawmluh record is shifted negatively by 2.1‰ after correcting it for temperature effect on calcite $\delta^{18}\text{O}$, and the orange dot marks the mean value of a modern stalagmite (see the Supplementary Materials). (D) *G. ruber* $\delta^{18}\text{O}$ record, retrieved from sediment core SO93-126KL, as a proxy for ISM intensity (27). (E) Atmospheric molecular oxygen $\delta^{18}\text{O}$ record ($\delta^{18}\text{O}_{\text{atm}}$), retrieved from the Antarctica Siple Dome ice core (38). (F) Tropical Indian Ocean SST reconstructed from sediment core SO189-39KL (39). (G) Benthic foraminifera $\delta^{18}\text{O}$ LR04 stack (8).

suggests that ISM rainfall during the LGM decreased to about 56% of today's value (table S3). Not only is this value comparable to the ~58% calculated for the eastern Amazon lowlands (32), but together, these values also imply a much weakened hydrological cycle in the deep tropics when Earth was in a full glacial state and, hence, are consistent with an increase in $\delta^{18}\text{O}$ of atmospheric O_2 during the LGM (Fig. 3) (38).

The reduced ISM strength during the LGM is attributable mainly to a combination of a lower SST over the tropical Indian Ocean and a larger continental ice/snow coverage (Fig. 3). Tropical SST dropped by ~3°C (23, 39), weakening convection over the region, decreasing

rainfall, and increasing $\delta^{18}\text{O}$ values of precipitation over the ISM domain. Meanwhile, the ice/snow expansion on the continent increased albedo and reduced sensible heating (35, 40); therefore, the expansion also weakened monsoonal rainfall. The exposure of the Sunda/Sahul shelf was shown by climate simulations as another key forcing of the tropical climate during the LGM, yet the same experiments were unable to capture large rainfall reduction in southern Myanmar (35).

Moisture transport pathway effect

The difference in $\delta^{18}\text{O}$ between the LGM and recent several hundred years decreases landward following the moisture trajectory, with ~2 and

<1‰ at CM and SEY, respectively (Fig. 4 and fig. S7, B and C). Since the NHSI values during the LGM were very close to those of today's, such a spatial-temporal pattern clearly shows that speleothem $\delta^{18}\text{O}$ is more sensitive to glacial forcings in coastal regions compared with those inland. We highlighted the spatial pattern by calculating the mean $\delta^{18}\text{O}$ values for every thousand years in each record and corrected locations other than CBoB for temperature effect (see Materials and Methods and fig. S11). The corrected records show a broad ^{18}O -depletion trend inlandward, consistent with the progressive rainout or Rayleigh distillation effect on water isotopes, implying a relatively stable trajectory of monsoon moisture for at least the recent ~40 ka (Fig. 4 and fig. S12). However, the offset between the records, which essentially describes the gradient of rainfall $\delta^{18}\text{O}$ across mainland Southeast Asia, was much larger (~8 to 10‰) during the last glacial period, particularly the LGM, compared with today or broadly the Holocene (~5 to 7‰), suggesting a stronger-than-today isotope fractionation of rainfall during the last glacial period (total difference ~3‰).

Following an early study (41), we term this change in isotope gradient through time the moisture transport pathway effect. We argue that the pathway effect reasonably explains the apparently different sensitivities of speleothem $\delta^{18}\text{O}$ to glacial forcings.

Changes in glacial forcings have led to a large negative shift in precipitation $\delta^{18}\text{O}$ between the LGM and the present, such as the >3‰ difference recorded in speleothems from the CBoB and Mawmluh

caves. However, as a result of the pathway effect, the negative $\delta^{18}\text{O}$ shift becomes gradually muted inland, to as low as <1‰ at SEY. Underestimating this pathway effect could lead to inconsistent conclusions on the $\delta^{18}\text{O}$ sensitivities to climate forcings. For example, the CBoB $\delta^{18}\text{O}$ record suggests that glacial boundary conditions strongly forced the ISM (Fig. 3 and fig. S7A), while the SEY record (fig. S7C) implies a strong influence of the NHSI but insensitivity to glacial forcings, which is, in fact, similar to the characteristic Chinese records (fig. S9).

Mechanism for the pathway effect

What may have caused the pathway effect? Here, we propose that it is temperature, possibly together with suppressed plant transpiration.

During the last glacial period, the global temperature was much lower (by ~4°C or more) than today's, in particular on the continent, due to an enhanced ice albedo and lower atmospheric CO_2 (10, 42), while the latitudinal temperature gradient was larger (43). The lower global temperature and larger temperature gradient led to a stronger-than-modern isotopic distillation and, hence, progressively lower $\delta^{18}\text{O}$ values of rainfall toward the ISM downstream.

To test how much temperature effect accounts for the observed $\delta^{18}\text{O}$ gradient changes, we calculate the approximate rainfall $\delta^{18}\text{O}$ gradient based on a temperature-dependent Rayleigh distillation model, largely following an early study (44). Regardless of any moisture

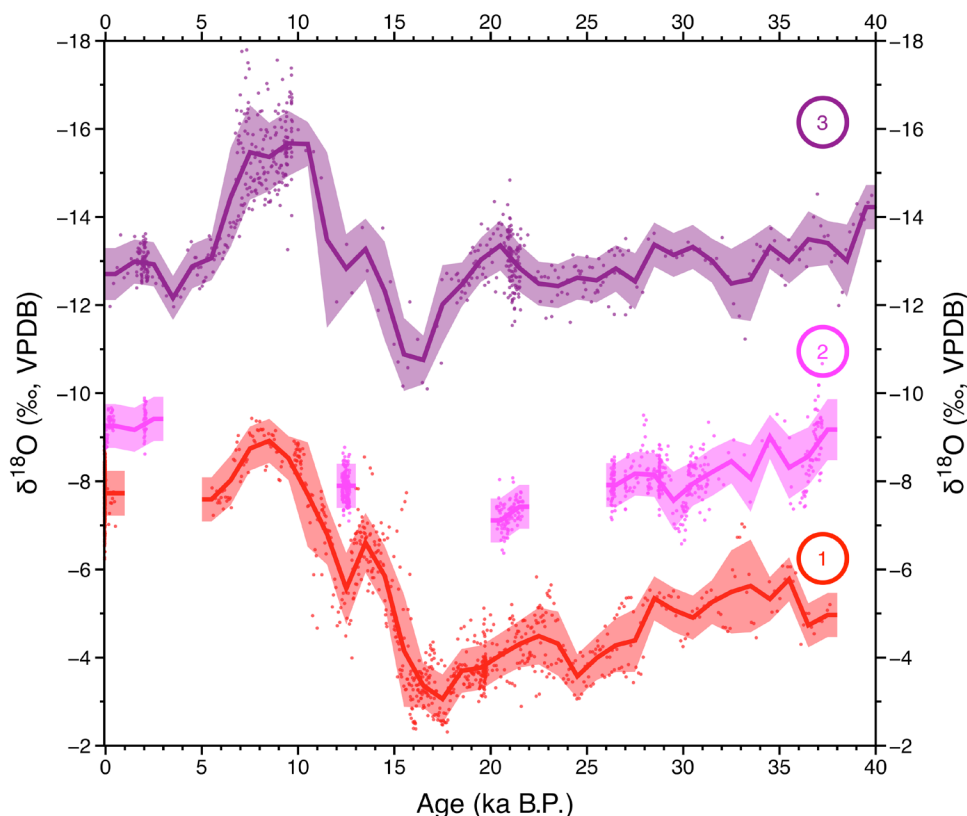


Fig. 4. Spatial-temporal comparison of speleothem $\delta^{18}\text{O}$ records from mainland Southeast Asia over the past 40 ka. The records obtained from the CBoB and Mawmluh (27, 26) (site 1), CM (site 2), and SEY (site 3) caves are shown in red, pink, and purple, respectively. Note that the $\delta^{18}\text{O}$ in those records from locations other than CBoB has been corrected for temperature effect during calcite precipitation (see details in Materials and Methods). We further smoothed each record using 1000-year averages (thick lines). The shaded envelopes indicate the range of 1σ uncertainty of the $\delta^{18}\text{O}$ values. The comparison shows a broad ^{18}O -depletion trend from coastal sites to inland, associated with the progressive rainout effect on water isotopes. In addition, the isotopic gradient was larger during the LGM compared with today.

loss en route to CBoB, we assign the moisture remaining at CBoB as the initial state. We then calculate $\delta^{18}\text{O}$ of moisture and precipitation at SEY using the equation

$$(1000 + \delta^{18}\text{O}_v)/(1000 + \delta^{18}\text{O}_{vi}) = f^{(\alpha(\text{liquid-vapor})-1)} \quad (7)$$

where $\delta^{18}\text{O}_v$ is the remaining water vapor $\delta^{18}\text{O}$, and $\delta^{18}\text{O}_{vi}$ is its initial value, which we calculate with

$$(1000 + \delta^{18}\text{O}_{pi})/(1000 + \delta^{18}\text{O}_{vi}) = \alpha(\text{liquid-vapor}) \quad (8)$$

where $\delta^{18}\text{O}_{pi}$ is the speleothem inferred rainfall $\delta^{18}\text{O}$ at CBoB and can be read from table S3, $\alpha(\text{liquid-vapor})$ is temperature dependent (Eq. 4), and f is the fraction of the initial water vapor remaining in air masses and is calculated by dividing PWV values (assuming RH = 100%) at different cave sites, which roughly decrease exponentially with temperature (Eqs. 5 and 6).

Applying the idealized Rayleigh distillation model, we find that the magnitudes of rainfall $\delta^{18}\text{O}$ decrease through the ISM transport trajectory are 6.0 and 7.7‰ for today and the LGM, respectively (fig. S13). Thus, temperature alone can explain ~1.7‰ out of the total ~3‰ difference in the $\delta^{18}\text{O}$ gradient between the LGM and the present across mainland Southeast Asia (Fig. 4).

We argue that the remaining ~1.3‰ results largely from changes in moisture recycling effect through suppressed plant transpiration but enhanced evaporation during the LGM. Different from evaporation, plant transpiration does not induce water isotope fractionation (45). Therefore, the proportion change of the two processes in moisture recycling can regulate the isotopic offset in rainfall between the last glacial period and the present (32). Various studies have indicated a reduction in forest coverage in now-heavily-forested Southeast Asia (46, 47), which enhanced moisture recycling through evaporation but reduced plant transpiration during glacial periods, and likely also contributed to a steeper isotopic gradient in rainfall. Moisture transport by eddy diffusion is another physical process that influences the continental rainfall $\delta^{18}\text{O}$ gradient (45). For a given degree of rainout, eddy diffusion results in shallower gradients in precipitation isotopes. However, such influence in the humid tropics is rather weak (45).

The pathway effect in Chinese speleothem $\delta^{18}\text{O}$ records

The pathway effect, which in the end counteracts the forcing of glacial boundary conditions, is probably the key to the lack of glacial-interglacial variations in the inland speleothem $\delta^{18}\text{O}$ records. To further test our hypothesis, we compare speleothem $\delta^{18}\text{O}$ records from the ISM region with those from southern China (Fig. 5), where the records generally show weak glacial-interglacial variations. We focus specifically on MIS 5 and select three speleothem $\delta^{18}\text{O}$ records from sites broadly along the moisture pathway landward [i.e., SEY caves, Dongge Cave (2), and Sanbao Cave (1)]. Rainfall over southern China comes remotely from the tropical Indian Ocean by the ISM (17) or adjacently from the South China Sea and Western Pacific (4). The moisture from the latter source is isotopically heavier, which can lead to relatively higher $\delta^{18}\text{O}$ values in the Dongge and Sanbao cave records (4), thus explaining why the speleothems there do not record a landward ^{18}O -depletion trend.

Notable differences exist among the three cave records when comparing their amplitudes of $\delta^{18}\text{O}$ minima during MIS 5e (the last interglacial) to two subsequent interstadials MIS 5a and 5c: SEY $\delta^{18}\text{O}$ values were prominently the lowest during MIS 5e; Dongge

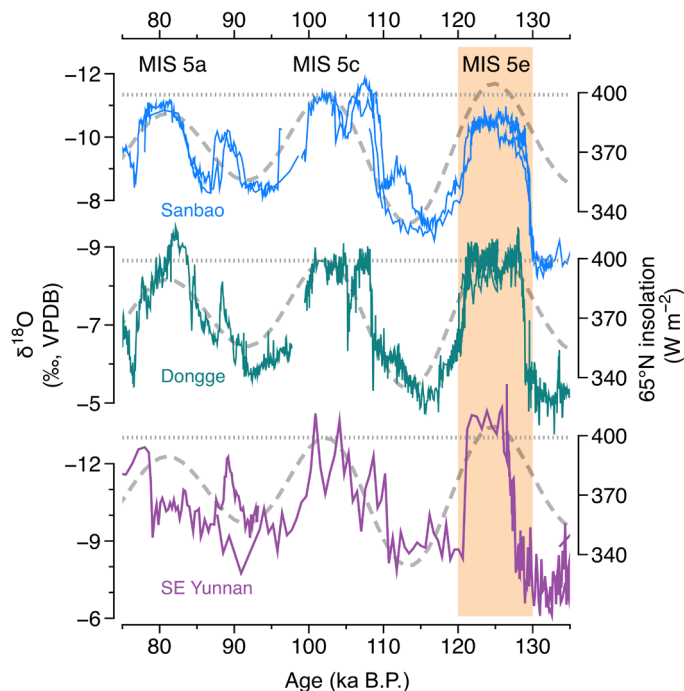


Fig. 5. Spatial-temporal comparison of speleothem $\delta^{18}\text{O}$ records from the SEY, Dongge (2) and Sanbao (1) caves in southern China during MIS 5. The summer (21 July) insolation at 65°N (the gray curve) is also plotted for comparison. To facilitate the comparison, we aligned the speleothem $\delta^{18}\text{O}$ values during MIS 5c with the insolation peak. The dashed lines, drawn from the MIS 5c insolation peak, indicate the difference among the three records in their amplitudes of $\delta^{18}\text{O}$ minima during MIS 5.

Cave $\delta^{18}\text{O}$ values during MIS 5e were similar to those during MIS 5a and 5c (2, 48); and further inland, Sanbao Cave $\delta^{18}\text{O}$ values were the highest during MIS 5e (1). The $\delta^{18}\text{O}$ values during MIS 5e in both Dongge and Sanbao records are clearly at odds not only with an enhanced insolation and stronger interglacial climate boundary conditions but also with the possibly reduced relative contribution of isotopically heavier moisture from the Pacific to the annual total precipitation (20). We hereby contend that the relatively positive $\delta^{18}\text{O}$ values in Dongge and, particularly, Sanbao speleothems can mainly be attributed to weakened isotopic distillation during MIS 5e, in line with the influence of the pathway effect. As the temperature differences among the three time periods are not prominent (48, 49), the weakened isotopic distillation is possibly due to an enhanced plant transpiration effect during MIS 5e.

Our proposed moisture transport pathway effect reasonably explains the lack of glacial-interglacial variability in Chinese speleothem $\delta^{18}\text{O}$ records. During glacial periods, AM rainfall amount was significantly lower due to glacial climate boundary forcings, and meanwhile, transported moisture endured stronger isotope fractionation over the continent. In comparison, during interglacial periods, AM rainfall increased, but a weaker isotope fractionation of transported moisture led to apparently high speleothem $\delta^{18}\text{O}$ values in southern China.

MATERIALS AND METHODS

Cave settings and sample descriptions

We have analyzed 19 stalagmite samples from 11 different caves from three sites broadly located along the trajectory of the ISM (table S1).

While the climatology at our three sites differs, the overall rainfall in the region comes predominantly from the ISM (figs. S1 and S2). For example, although the annual rainfall varies from >2000 mm at CBoB to ~1000 mm at SEY, over 90% of the rainfall comes from the BoB and precipitates between late May and early November. The mean annual temperature is ~28°C at CBoB, dropping to ~15°C at SEY, largely due to combined latitudinal and altitudinal effects.

The CBoB contains four caves, including three in Mon State [Phabaung Gu (PHA)] and Kayin State [Padamya Gu (PDM) and Ya Thea Pyan Gu (YTP)], Myanmar, ~180 km to the southeast of Yangon, and one [Hin Tum No. 2 Cave (HT2)] in Uthai Thani province, Thailand, ~240 km to the northwest of Bangkok. The three Burmese caves, all located less than 100 m above sea level (asl), formed in tower karst, and the ages of the host carbonate rocks are unknown. The PHA (~1000 m long) and YTP (~400 m in length) caves contain multiple chambers. The PDM cave (~100 m long) has only one chamber but with a narrow entrance, which limits air ventilation. The HT2 cave (~200 m asl and ~300 m long) contains multiple chambers in argillaceous limestone that dates back to the Ordovician Period.

Our CM site includes the Lin Noe Twin (LNT) cave (~1300 m asl), located on the eastern side of the Shan Plateau. The cave formed within partially dolomitized Permian-Triassic limestone formations (50). The LNT cave consists of both an inner and an outer chamber. The outer chamber is small and narrow since its roof has collapsed; it also has a steep slope extending ~20 m downward. At its end, a narrow vertical drop of ~15 m leads to a large inner chamber, which is ~100 m in diameter and has poor ventilation.

In Yunnan, China, our six caves are located around 23.5°N, 104°E in Wenshan Prefecture (~2000 m asl), ~220 km to the southeast of Kunming. All SEY caves contain multiple chambers, likely longer than 1000 m.

To minimize the potential kinetic effect on carbonate stable isotope ratios, we only collected stalagmites from the inner chambers of the caves with very limited air ventilation and relative humidity close to 100%. All collected samples are made of calcite except for stalagmite A14, whose top 8 mm is calcite, while the rest is composed of aragonite. Moreover, in consideration of cave preservation, we almost exclusively collected broken stalagmites. The only standing stalagmite we collected was PHA02, with active dripping water. We used PHA02 to evaluate the equilibrium conditions of carbonate precipitation. Details about the caves, samples, and related regional climatology are in figs. S1 to S3 and table S1.

U-Th ages and stable isotopes

We halved all the stalagmites along the growth axes and polished their surfaces. We then drilled a total of 164 subsamples along the growth axes for ^{230}Th dating (fig. S4). We separated and purified uranium and thorium, following similar chemical procedures as described in (51) and (52) and analyzed all the CBoB, CM, and part of SEY samples in the Environmental Geochemistry Laboratory (EGL) at the Earth Observatory of Singapore (EOS)/Asian School of the Environment (ASE), Nanyang Technological University, Singapore, while we analyzed the remaining SEY samples at the Institute of Global Environmental Change (IGEC), Xi'an Jiaotong University, Xi'an, China. In both laboratories, the ^{230}Th dating techniques were identical, performed on Thermo-Finnigan Neptune Plus multi-collector inductively coupled plasma mass spectrometers using recently improved techniques (18, 19).

We drilled a total of 2611 subsamples along the growth axes of the stalagmite samples at 1- to 10-mm increments for stable isotopic analysis ($\delta^{18}\text{O}$ and $\delta^{13}\text{C}$) (figs. S5 and S6). We completed the measurements at three laboratories with different analytical systems: the EGL, EOS/ASE, Nanyang Technological University, Singapore; the Speleothem Isotope Laboratory (SIL), Nanjing Normal University, Nanjing, China; and the IGEC, Xi'an Jiaotong University, Xi'an, China. We report our results in per mil (‰), relative to the Vienna PeeDee Belemnite (VPDB) standard. At the EGL, we analyzed stable isotope samples using an online carbonate preparation system (Gasbench II) interfaced with a Thermo Fisher Scientific Delta V isotope ratio mass spectrometer (IRMS). We measured the stable isotope at the SIL on a Thermo-Finnigan MAT-253 IRMS equipped with a Kiel Carbonate Device III, and at the IGEC on a Thermo-Finnigan MAT-253 IRMS equipped with a Kiel Carbonate Device IV. At all the three laboratories, measurements of international carbonate standard NBS19 and in-house TTB1 standard showed a long-term reproducibility of ~0.1‰ or better (1σ). Cross-laboratory calibration confirms that stable isotope data are well reproduced among the three laboratories (e.g., fig. S5).

U and Th isotopic compositions and ^{230}Th dates are presented in data S1. ^{238}U concentrations range from 20 to 200 parts per billion (ppb), with a few exceptions (~3 to 13 parts per million) mainly in the aragonite portion of sample A14. ^{232}Th concentrations mostly range from fewer than 10 parts per trillion to the ppb level. We calculated ^{230}Th ages assuming an initial $^{230}\text{Th}/^{232}\text{Th}$ atomic ratio of $(4.4 \pm 2.2) \times 10^{-6}$, which is the value for a material at secular equilibrium, with the bulk Earth $^{232}\text{Th}/^{238}\text{U}$ value of 3.8. We considered it reasonable to adopt this value for initial ^{230}Th correction, as most samples had relatively high $^{230}\text{Th}/^{232}\text{Th}$ ratios, and initial correction was negligible. In addition, at the tip of modern stalagmite PHA02, $^{230}\text{Th}/^{232}\text{Th}$ atomic ratios are well within uncertainties of $(4.4 \pm 2.2) \times 10^{-6}$, which supports the assumption. The typical relative errors (2σ) in ages after the initial ^{230}Th correction for other samples are better than 1%, and all ^{230}Th dates are in stratigraphic order within 2σ errors, further confirming the data robustness (53). We used the StalAge method (54) to establish the chronology for samples with large errors in their age dating due to low U and high Th contents (e.g., HT2-1), or for samples whose ages have minor inversions (i.e., age inversions still within uncertainties, for example, XR05). We assumed that the top ~1 cm above a hiatus in sample PDM02 grew steadily between 11.2 and 11.5 ka. We used linear interpolation to establish the chronology for the rest of the samples (fig. S4).

The stable isotope ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) compositions are in data S2. $\delta^{18}\text{O}$ shows large shifts, ranging from -8.6 to -2.3‰, -9.0 to -4.7‰, and -16.1 to -6.1‰ in the CBoB, CM, and SEY speleothem samples, respectively (figs. S5 to S7).

Rainfall $\delta^{18}\text{O}$ gradient across mainland Southeast Asia

Carbonate $\delta^{18}\text{O}$ in our samples reflected mostly the rainfall $\delta^{18}\text{O}$ and cave temperature at the time of carbonate precipitation. To better portray the spatial pattern of rainfall $\delta^{18}\text{O}$, we corrected the speleothem calcite $\delta^{18}\text{O}$ values from sites other than the CBoB for temperature-dependent isotope fractionation resulting from the differences in cave temperatures. The modern cave temperatures are similar to the mean annual temperatures of nearby cities, that is, 28°C in the CBoB caves (near Yangon and Bangkok), 18°C in Mawmluh Cave (near Shillong), 20°C in CM LNT cave (near Taunggyi), and 15°C in SEY caves (near Kunming). We assumed

that the temperature differences among these caves have not changed through time.

The temperature differences among the cave sites were in fact likely larger during glacial periods, since the global meridional temperature gradient was generally larger (43) and the tropical lapse rate was possibly steeper (55, 56). For example, during the LGM, the temperature likely dropped between 2° and 4°C at CBoB (5, 20, 27, 29, 34) and between 4° and 8°C at SEY (20, 24, 57). Model simulations also suggest an increase of ~2°C/1000 km in the regional temperature gradient during the LGM (20). Note that we considered here only the overall temperature gradient across mainland Southeast Asia from CBoB to SEY (~1000 km apart), as to date, we have only limited knowledge of the climatology of CM. An amplified temperature gradient during the LGM would have shifted speleothem $\delta^{18}\text{O}$ even more positively in the downwind region relative to local rainfall $\delta^{18}\text{O}$. Therefore, the aforementioned assumption provides a conservative estimate of the rainfall $\delta^{18}\text{O}$ gradient in our study area during the LGM. The rainfall $\delta^{18}\text{O}$ gradient was possibly larger than estimated here but would not have differed substantially because of the small temperature-dependent fractionation factor of oxygen isotopes between speleothem calcite and cave drip water (~-0.21‰/°C) (25).

After we remove site-specific temperature effect on speleothem $\delta^{18}\text{O}$ (figs. S8 and S11), the difference between the contemporaneous speleothem $\delta^{18}\text{O}$ values at CBoB and SEY is essentially equivalent to the difference in rainfall $\delta^{18}\text{O}$ values in the region (Fig. 4 and fig. S12B), as cave temperature effect and ocean reservoir effect in speleothem $\delta^{18}\text{O}$ through time cancel each other (32). The speleothem $\delta^{18}\text{O}$ gradient obtained by dividing the speleothem $\delta^{18}\text{O}$ offset by the length of the transect (~1000 km) is, therefore, a good approximation of the overall rainfall $\delta^{18}\text{O}$ gradient across the region following the monsoon trajectory. Again, we considered only the overall gradient across mainland Southeast Asia from the CBoB to SEY sites.

We further highlighted the spatial pattern by calculating the mean $\delta^{18}\text{O}$ values and their SDs for every 1000 years, assuming 0.5‰ as the minimum 1 σ SD, as smaller SDs could also reflect relatively sparse data points. During the most recent 1000 years, the speleothem $\delta^{18}\text{O}$ values, after the site-specific temperature correction, have decreased by $5.2 \pm 0.7\text{‰}$ across mainland Southeast Asia (Fig. 4). This ~5.2‰/1000 km gradient reflects to a large extent the temperature gradient (~13°C/1000 km) across the transect, which is due to a combination of altitude and latitude effects. Our records show that the $\delta^{18}\text{O}$ gradient was much larger during the last glacial time (~8 to 10‰/1000 km or more if we consider the aforementioned amplified temperature effect) relative to today or broadly the Holocene (~5 to 7‰/1000 km), suggesting a stronger-than-today isotope fractionation of precipitation $\delta^{18}\text{O}$ during the last glacial period (total difference ~3‰) (Fig. 4).

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at <http://advances.sciencemag.org/cgi/content/full/6/7/eaay8189/DC1>

Supplementary Text

Fig. S1. Locations of the study sites.

Fig. S2. Climatology of relevant cities.

Fig. S3. Images of stalagmite samples.

Fig. S4. Age models.

Fig. S5. Replication test on stable isotope data from PDM02.

Fig. S6. Scatter plot of $\delta^{18}\text{O}$ versus $\delta^{13}\text{C}$.

Fig. S7. The three obtained speleothem $\delta^{18}\text{O}$ records.

Fig. S8. Comparison of the CBoB and Mawmluh cave records.

Fig. S9. Comparison of orbital AM records and Vostok atmospheric molecular oxygen $\delta^{18}\text{O}$ record.

Fig. S10. Spatial-temporal comparison of speleothem $\delta^{18}\text{O}$ records from the coastal Indian Ocean and southern China.

Fig. S11. Ice volume and temperature effects.

Fig. S12. Spatial-temporal comparison of speleothem $\delta^{18}\text{O}$ records from mainland Southeast Asia over the past 40 ka.

Fig. S13. Calculation of rainfall $\delta^{18}\text{O}$ gradient across mainland Southeast Asia.

Table S1. A list of the studied speleothem samples and their cave locations.

Table S2. Rainfall oxygen isotopic compositions across mainland Southeast Asia.

Table S3. Calculations of water vapor loss over the CBoB site.

Data S1. ^{230}Th dating results, with errors within 2σ (95% in confidence).

Data S2. Stable isotope compositions.

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