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Inconsistency between records of $\delta^{18}\text{O}$ and trace element ratios from stalagmites: Evidence for increasing mid-late Holocene moisture in arid central Asia

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Abstract

The interpretation of trace element/calcium ratios of speleothems as indicators of local hydroclimatic variability in the vicinity of caves has led to controversy in reconstructing the evolution of moisture conditions in arid central Asia (ACA) during the Holocene. Here we present records of Mg/Ca, Sr/Ca, Ba/Ca, and U/Ca from precisely dated stalagmites from Baluk cave in Xinjiang (northwest China), spanning the past 9370 years. The co-variations of the trace element ratios, together with the slopes of the regression lines of the corresponding logarithmically transformed data, suggest that they are dominated by prior calcite precipitation (PCP) and thus can be used as reliable proxies of changes in moisture/precipitation. The trace element ratios are relatively high during ~9 to 5 ka and lower from 5 ka to the present, indicating a trend of increasing mid-late Holocene moisture in ACA. The long-term trend of variation of the trace element ratios is correlative with two other records of speleothem trace element ratios from caves in ACA: Kesang cave (western Xinjiang) and Ton cave (Uzbekistan). This spatial coherency of the trend of inferred moisture conditions from three caves that are separated by hundreds of kilometers demonstrates that speleothem trace element ratios are indicative of large spatial scale rather than local hydroclimatic variability in ACA during the Holocene. However, the long-term trend of variation of the trace element ratios is the inverse of the corresponding oxygen isotope ($\delta^{18}\text{O}$) records from the same cave sites, which implies that Holocene speleothem $\delta^{18}\text{O}$ records do not represent changes in the precipitation amount in ACA; rather, they most likely reflect moisture sources and related water vapor transport controlled by Northern Hemisphere summer insolation (NHSI). Our findings provide new evidence for a 'westerlies-dominated climatic regime', which influenced hydroclimatic changes in ACA during the Holocene.

Keywords

arid central Asia, Holocene moisture/precipitation, inconsistent trends, speleothem $\delta^{18}\text{O}$, speleothem trace element ratios, westerlies circulation

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Introduction

Arid central Asia (ACA), comprising the region from the Caspian Sea in the West to the western Mongolian Plateau in the East (Figure 1), is climatically dominated by the mid-latitude westerlies circulation. The mid-latitude westerlies, forming in the upper troposphere (200–300 hPa), are concentrated in a narrow band ('jet-stream') within which large, wave-like perturbations are superimposed. The latitude of the jet shifts from day to day and the waves usually move from west to east ('west winds'). Because of the prevailing westerlies, the long-term mean water vapor fluxes influencing the ACA are mostly from the West (Huang et al., 2013; Yatagai, 2003), that is, North Atlantic Ocean, Mediterranean Sea, and Caspian Sea (Aizen et al., 2006; Bothe et al., 2012; Chen et al., 2008). As the largest azonal arid region in the world, ACA is characterized by scarce water resources and fragile ecosystems, making it sensitive to climate change (e.g. Liu et al., 2018; Shi et al., 2007; Yao et al., 2019; Zhang et al., 2016; Zhao et al., 2010). For example, in northwest (NW) China, with ongoing climatic warming, substantial increases in precipitation, glacier meltwater and river runoff, air temperature, the water level of

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Figure 1. Map showing the study area, with locations of cave records mentioned in the text. The blue dashed line indicates the northern limit of the modern Asian summer monsoon (Chen et al., 2008). The area enclosed by the white dashed line is arid Asia (Feng et al., 2014), and the area enclosed by the yellow dashed line indicates the core area of ACA (Huang et al., 2015). The arrows illustrate the main trajectories of the westerlies, East Asian summer monsoon (EASM), and Indian summer monsoon (ISM). [AQ: 1] 1: Baluk cave (this study); 2: Kesang cave (Cai et al., 2017; Cheng et al., 2012, 2016b); 3: Ton cave (Cheng et al., 2016b); 4: Dongge cave (Wang et al., 2005).

inland lakes, and the frequency of flood disasters have occurred during the past few decades (Shi et al., 2007). In addition, recent studies show that the vegetation in Xinjiang in NW China has been markedly influenced by regional hydroclimatic change over a similar interval (Liu et al., 2018; Yao et al., 2019; Zhang et al., 2016). Thus, climate change is a major factor affecting regional hydrology, vegetation, ecology, and sustainable development in ACA. In order to accurately evaluate the impact of future climate change on the ecological environment, a key step is to improve our understanding of the past hydroclimatic evolution of this arid region.

Geochemical proxies from speleothems, such as trace element/calcium ratios (e.g. Mg/Ca, Sr/Ca, Ba/Ca, U/Ca, S/Ca), are regarded as reliable proxies of past hydroclimatic changes on various timescales (e.g. Baldini et al., 2002; Bernal et al., 2016; Hartmann et al., 2013; Li et al., 2005; McDonald et al., 2004; Springer et al., 2008; Zhang et al., 2018b; Zhou et al., 2009). However, the use of speleothem trace element ratios in paleoclimatic reconstruction in ACA was limited. For example, there is only one cave geochemical record from NW China – from Kesang cave in western Xinjiang (Cheng et al., 2016b). Notably, it was proposed that the trace element ratios from Kesang cave only document local hydroclimatic variability around the cave site (Cheng et al., 2016b), which has led to controversy regarding the reconstruction of the Holocene evolution of moisture/precipitation in this arid region (e.g. Chen et al., 2016; Cheng et al., 2016b). For example, Cheng et al. (2012, 2016b) and Cai et al. (2017) assumed that precipitation in ACA was relatively high during the early Holocene and then commenced a decreasing trend from the early to the mid–late Holocene, based on the speleothem $\delta^{18}\text{O}$ record from Kesang cave. However, this trend is the opposite to the long-term decreasing trend of speleothem trace element ratios from this cave, which was interpreted as a proxy of local hydroclimatic variability around the cave site (Cheng et al., 2016b). By contrast, based on lake sediment and loess–paleosol records, Chen et al. (2016) argued that the Holocene moisture/precipitation evolution of ACA was characterized by a relatively dry early Holocene and a wet mid–late Holocene, which is coherent with the long-term trend of variation of speleothem trace element ratios from Kesang cave (Cheng et al., 2016b). Therefore, the pattern of Holocene moisture/precipitation evolution in ACA remains unclear, partly because of the

paucity of speleothem geochemical records from the region. Consequently, more speleothem geochemical records are needed to resolve the pattern of evolution of moisture conditions in ACA during the Holocene.

In order to address this issue, we obtained records of Mg/Ca, Sr/Ca, Ba/Ca, and U/Ca from precise-dated stalagmites from Baluk cave in Xinjiang, NW China, spanning the last 9370 years. By comparing records of trace element ratios from three well-separated cave sites in ACA (Baluk, Kesang, and Ton), we attempt to determine whether records of speleothem trace element ratios in ACA can represent the Holocene evolution of moisture/precipitation on a large spatial scale. In addition, we compare the records of speleothem trace element ratios with speleothem $\delta^{18}\text{O}$ records from three caves in ACA, in order to explore the climatic significance of Holocene speleothem $\delta^{18}\text{O}$ records in the region. We then try to improve our understanding of the Holocene evolution of moisture/precipitation in ACA as a whole.

Materials and methods

Baluk cave (84°44'E, 42°26'N, 2752 m a.s.l.; Figure 1) is located ~120 km southeast of Bayanbulak Town, in Xinjiang Uygur Autonomous Region in NW China, an important part of ACA. The study area is located far inland, beyond the northern limit of the modern Asian summer monsoon (ASM; Chen et al., 2018), and is climatically dominated by the westerly circulation. Rainfall in the study area mainly occurs during boreal spring–summer (April–September). Stalagmites were collected in the central chamber of Baluk cave in September 2012. Powder subsamples were collected using an electrical dental drill in a class-100 laminar flow bench housed in a class-10,000 subsampling room. The weights of the subsamples ranged from 30 to 80 mg. U–Th dates from stalagmites BLK12A and BLK12B were reported previously (Liu et al., 2019). In this study, two additional intervals from these two stalagmites were used for dating: BLK12A-H (24 mm long) is the topmost part of BLK12A that grew above a hiatus (Figure 2a) and BLK12 C (87 mm long) is the topmost part of BLK12B which grew in a different direction from that of the main part of the stalagmite (Figure 2c). A total of 19 powder subsamples (6 for BLK12A-H and 13 for BLK12 C) were collected for U–Th dating.

The procedures of chemical preparation for U and Th separation and isotopic concentration measurement followed the methods described by Shen et al. (2003, 2012) and were conducted at the High-Precision Mass Spectrometry and Environment Change Laboratory (HISPEC) of National Taiwan University during January–April 2013. Decay constants and $^{238}\text{U}/^{235}\text{U}$ ratios reported in Jaffey et al. (1971), Cheng et al. (2013), and Hiess et al. (2012) were applied to calculate the stalagmite U–Th ages from Baluk cave.

For the measurement of speleothem element concentrations (Ca, Mg, Sr, Ba, and U), 203 powder subsamples (9 for BLK12A-H, 97 for BLK12A, 67 for BLK12B, and 30 for BLK12 C) were drilled along the growth axis of the stalagmite profiles. Powders were drilled at a 2.5-mm interval from BLK12A-H, from the interval of 17.0–182.0 mm from BLK12A, from the interval of 12.5–125.0 mm from BLK12B, and at a 3.0-mm interval for BLK12 C. In addition, powders were drilled at a 0.5-mm interval from the interval of 0–15.0 mm from BLK12A and from 0–10.0 mm for BLK12B. Determination of Ca, Mg, Sr, Ba, and U concentrations was conducted in the Chongqing Key Laboratory of Karst Environment, Southwest University, China, using an Optima 2100 DV inductively coupled plasma emission spectrometer (ICP-OES; PerkinElmer, USA) for Ca^{2+} and Mg^{2+} and a single-collector inductively coupled plasma mass spectrometry (SC-ICP-MS; Thermo Element XR, USA) for Sr^{2+} , Ba^{2+} , and U^{4+} . Carbonate powder of each subsample (~300 μg) was first dissolved with 3% redistilled (RD) HNO_3 and 1% hydrofluoric acid (HF) in a 15-mL

Figure 2. Polished sections and age–depth models based on combined stalagmite profiles from Baluk cave. Section views along the central growth axes of stalagmites BLK12A-H (a, 24 mm in length), BLK12 C (c, 87 mm in length), BLK12A (a, 184 mm in length), and BLK12B (c, 125 mm in length) (Liu et al., 2019) are shown. U–Th dates, including new dates from BLK12A-H (b) and BLK12 C (d) and published dates from BLK12A (b) and BLK12B (d), are plotted against depth. Age–depth models for all stalagmites are calculated using the StalAge algorithm, with the corresponding 95% confidence limits being shown in red (Scholz and Hoffmann, 2011). The yellow dashed lines indicate a hiatus within the combined stalagmite record of BLK12A/H.

centrifuge tube. Since no residue was found after dissolution, the solution of each subsample was diluted with 3% HNO₃, using a dilution factor of ~50,000, before the elemental concentrations were determined. The averaged analytical uncertainties were better than 2%. The trace metals are reported as ratios to Ca (i.e. Mg/Ca, Sr/Ca, Ba/Ca, and U/Ca).

In addition, we analyzed the stable oxygen isotope composition ($\delta^{18}\text{O}$) of stalagmites BLK12A-H and BLK12 C at low resolution in order to determine the long-term trends of variation. A total of 39 powder subsamples were collected, including 9 from BLK12A-H (at a 2.5-mm interval) and 30 from BLK12 C (at a 3.0-mm interval). The measurement procedures followed those of Liu et al. (2019) and were conducted at the Key Laboratory of Western China's Environmental Systems (MOE), Lanzhou University.

Results

Age–depth models

As shown in Table 1, the U–Th dates for BLK12A-H and BLK12 C are in stratigraphical order, within the error limits. For BLK12A-H, the six ²³⁰Th ICP-MS dates, with an averaged error of ± 145 years, range from 142 ± 109 yr BP (before present, where present is AD 1950) at 1.5 mm from the top to 1245 ± 188 yr BP at 24.0 mm depth. For BLK12 C, the determined age interval ranges from 18 ± 66 yr BP at 1.0 mm from the top to 2448 ± 330 yr BP at 82.5 mm depth, with an averaged error of ± 150 years

(Figure 2 and Table 1). The StalAge fitting method, a well-developed approach incorporating a stratigraphic factor (Scholz and Hoffmann, 2011), was used to build age models for all the stalagmites. The corresponding results are shown in Figure 2.

Based on the StalAge models, we produced the time series of Mg/Ca, Sr/Ca, Ba/Ca, and U/Ca and of $\delta^{18}\text{O}$ from Baluk cave. For comparison, we combined the time series of BLK12A-H and BLK12A into a single record, designated BLK12A/H, which spans the time intervals of 264–1180 and 2492–9378 yr BP, respectively. A hiatus is found during 1180–2492 yr BP in BLK12A/H (Figure 2). Similarly, we combined the time series of BLK12B and BLK12 C into a single record (BLK12B/C), which spans the interval from –8 to 2610 yr BP and from 2755 to 8302 yr BP. The mean time resolutions are 114.5 years for BLK12A-H; 21.0 years within the interval of 0–15.0 mm and 93.7 years within the interval of 17.0–182.0 mm for BLK12A; 90.3 years for BLK12 C; 20.5 years within the interval of 0–10.0 mm and 109.8 years within the interval of 12.5–125.0 mm for BLK12B.

Trace element ratios from Baluk cave

Although interrupted by hiatus, all of the time series of Baluk cave trace element ratios exhibit similar long-term temporal trends of decreasing values (Figure 3). For example, the Sr/Ca ratio of BLK12A/H fluctuates within the range of 42×10^{-3} to 55×10^{-3} during 9.3–4.4 thousand years ago (ka) and then rapidly decreases to 36×10^{-3} at ~2.7 ka (Figure 3b). The Mg/Ca ratio of

Table 1. Uranium and Thorium isotopic compositions and ^{230}Th ICP-MS dates of stalagmites BLK12A-H and BLK12 C from Baluk cave, NW China. **[AQ: 2]**

Sample ID	Depth (mm)	^{238}U (ppb) ^a	^{232}Th (ppt)	$\delta^{234}\text{U}$ (measured) ^a	$^{230}\text{Th}/^{238}\text{U}$ (activity) ^b	$^{230}\text{Th}/^{232}\text{Th}$ (ppm) ^c	^{230}Th age (years) (uncorrected)	^{230}Th age (yr BP) (corrected) ^{b,d}	$\delta^{234}\text{U}_{\text{initial}}$ (corrected) ^e
BLK12A-H									
A1	1.5	591.2 ± 0.5	5906.5 ± 16.0	229.0 ± 1.3	0.00473 ± 0.00019	7.8 ± 0.3	421 ± 17	142 ± 109	229.2 ± 1.3
A2	2.5	624.2 ± 1.2	8252.9 ± 35.3	226.8 ± 4.1	0.00714 ± 0.00021	8.9 ± 0.3	637 ± 19	288 ± 144	227.0 ± 4.1
A3	7.5	535.4 ± 0.5	10,301.1 ± 36.5	226.8 ± 1.1	0.01230 ± 0.00037	10.5 ± 0.3	1099 ± 33	620 ± 211	227.2 ± 1.1
A4	14.5	556.6 ± 0.6	1379.0 ± 5.8	230.0 ± 1.5	0.01009 ± 0.00013	67.1 ± 0.9	898 ± 11	781 ± 29	230.6 ± 1.5
A5	19.0	564.5 ± 1.0	9745.0 ± 38.4	235.8 ± 2.2	0.01697 ± 0.00034	16.2 ± 0.3	1507 ± 31	1074 ± 188	236.5 ± 2.2
A6	24.0	593.9 ± 0.6	1260.3 ± 27.2	236.5 ± 1.4	0.01889 ± 0.00033	18.0 ± 0.3	1678 ± 30	1245 ± 188	237.4 ± 1.4
BLK12 C									
C1	1.0	713.8 ± 0.8	4685.8 ± 12.5	319.0 ± 1.5	0.00257 ± 0.00011	6.5 ± 0.3	213 ± 9	18 ± 66	319.1 ± 1.5
C2	2.5	691.1 ± 2.0	3301.6 ± 14.7	322.6 ± 3.3	0.00244 ± 0.00009	8.4 ± 0.3	201 ± 7	43 ± 48	322.7 ± 3.3
C3	7.0	755.6 ± 2.0	1962.5 ± 10.8	315.6 ± 3.1	0.00347 ± 0.00007	22.0 ± 0.5	288 ± 6	173 ± 27	315.8 ± 3.1
C4	12.5	724.0 ± 1.8	3294.5 ± 13.2	319.1 ± 2.7	0.00604 ± 0.00011	21.9 ± 0.4	500 ± 9	346 ± 47	319.5 ± 2.7
C5	23.5	629.0 ± 0.9	804.3 ± 9.9	317.4 ± 2.0	0.00803 ± 0.00010	103.6 ± 1.9	667 ± 9	578 ± 16	317.9 ± 2.0
C6	26.5	790.4 ± 0.5	3011.0 ± 7.2	318.3 ± 1.2	0.00936 ± 0.00012	40.5 ± 0.5	777 ± 10	638 ± 39	318.9 ± 1.2
C7	32.5	728.8 ± 1.1	7001.5 ± 22.4	325.5 ± 2.0	0.01214 ± 0.00020	20.8 ± 0.3	1003 ± 16	748 ± 97	326.2 ± 2.0
C8	37.5	768.8 ± 1.2	18,351.3 ± 62.1	324.7 ± 2.1	0.01881 ± 0.00041	13.0 ± 0.3	1559 ± 34	1018 ± 242	325.7 ± 2.1
C9	44.5	893.0 ± 1.3	4704.5 ± 11.7	306.3 ± 2.5	0.01600 ± 0.00019	50.1 ± 0.6	1344 ± 17	1174 ± 56	307.4 ± 2.5
C10	56.5	907.6 ± 1.3	32,075.0 ± 142.0	309.5 ± 2.2	0.03014 ± 0.00058	14.1 ± 0.3	2537 ± 50	1758 ± 362	311.1 ± 2.2
C11	65.5	900.0 ± 1.2	14,994.9 ± 70.3	319.1 ± 1.6	0.02824 ± 0.00045	27.9 ± 0.5	2357 ± 38	1960 ± 172	321.0 ± 1.6
C12	77.5	875.6 ± 1.2	38,462.5 ± 276.9	312.8 ± 2.1	0.03779 ± 0.00079	14.1 ± 0.3	3182 ± 68	2228 ± 452	314.8 ± 2.1
C13	82.5	861.2 ± 1.4	27,412.1 ± 136.6	303.1 ± 2.2	0.03724 ± 0.00073	19.3 ± 0.4	3158 ± 63	2448 ± 330	305.3 ± 2.3

ICP-MS: inductively coupled plasma mass spectrometry; NW: northwest; MC-ICP-MS: multi-collector inductively coupled plasma mass spectrometry. Uncertainties in the U–Th isotopic data and ^{230}Th dates (yr BP; before AD 1950) are given at the two-sigma (2σ) level or as two standard deviations of the mean ($2\sigma_m$). Chemical analysis was performed from 25 to 30 January in 2013 (Shen et al., 2002, 2003) and MC-ICP-MS analysis from 6 to 25 February, 2013 (Shen et al., 2012). The values shown are for a material at secular equilibrium, with the crustal $^{232}\text{Th}/^{238}\text{U}$ value of 3.8. The errors are arbitrarily assumed to be 50%. Analytical errors are 2 sigma of the mean.

^a $[\delta^{234}\text{U}] = [^{235}\text{U}] \times 137.818 (\pm 0.65\%)$ (Hess et al., 2012); $d^{234}\text{U} = ([^{234}\text{U}/^{238}\text{U}]_{\text{activity}} - 1) \times 1000$.

^b $[^{230}\text{Th}/^{238}\text{U}]_{\text{activity}} = 1 - e^{-\lambda^{230}\text{Th}T} + (d^{234}\text{U}_{\text{measured}}/1000)[^{230}\text{Th}/(^{230}\text{Th} - ^{234}\text{Th})](1 - e^{-(\lambda^{230}\text{Th} - \lambda^{234}\text{U})T})$, where T is the age. BP means before present, where present is AD 1950.

Decay constants are $9.1705 \times 10^{-6} \text{ yr}^{-1}$ for ^{230}Th , $2.8221 \times 10^{-6} \text{ yr}^{-1}$ for ^{234}U (Cheng et al., 2013), and $1.55125 \times 10^{-10} \text{ yr}^{-1}$ for ^{238}U (Jaffey et al., 1971).

^cThe degree of detrital ^{230}Th contamination is indicated by the $[^{230}\text{Th}/^{232}\text{Th}]$ atomic ratio instead of the activity ratio.

^dAge corrections for samples were calculated using an estimated atomic $^{230}\text{Th}/^{232}\text{Th}$ ratio of 4 ± 2 ppm.

^e $d^{234}\text{U}_{\text{initial (corrected)}}$ was calculated based on the ^{230}Th age (T), that is, $d^{234}\text{U}_{\text{initial}} = d^{234}\text{U}_{\text{measured}} \times e^{\lambda^{234}\text{U}T}$, and T is the corrected age.

BLK12B/C varies from 7×10^{-3} to 12×10^{-3} during 8.3–4.5 ka, with large centennial-scale oscillations superimposed, and then decreases gradually during 4.5–0.5 ka (Figure 3e). The records of the Ba/Ca and U/Ca ratios from BLK12A/H and the Sr/Ca, Ba/Ca, and U/Ca ratios from BLK12B/C exhibit persistent decreasing trends with time, except the abruptly increased trace element ratios during the hiatus around 3.0 ka (Figure 3c, 3d, and 3f–h). The Mg/Ca record of BLK12A/H however exhibits extremely low ratios during 5.0–3.1 ka and overall increased Mg/Ca ratios after 1.18 ka (Figure 3a). We calculated the linear correlation coefficients for these ratios and the results are listed in Table 2. Significant positive correlations are particularly evident between Sr/Ca and Ba/Ca ($r = 0.65$, $p < 0.001$) and between Sr/Ca and U/Ca ($r = 0.84$, $p < 0.001$) for BLK12A/H, and between Mg/Ca and Sr/Ca ($r = 0.61$, $p < 0.001$), Sr/Ca and Ba/Ca ($r = 0.71$, $p < 0.001$), and Sr/Ca and U/Ca ($r = 0.85$, $p < 0.001$) for BLK12B/C. Moreover, all the trace element ratios from Baluk cave exhibit a pronounced increase at ~3.0 ka, indicating the occurrence of coherent abrupt changes on the centennial scale. This evidence indicates that the Baluk cave trace element ratios co-vary substantially during the Holocene.

Discussion

Climatic significance of the Baluk cave trace element ratios

A previous study showed that the co-variation of speleothem trace element ratios (Mg/Ca, Sr/Ca, Ba/Ca, and U/Ca) from Kesang

cave in western Xinjiang and Ton cave in Uzbekistan in ACA mainly reflects the precipitation–evaporation (P–E) conditions at each cave site (Cheng et al., 2016b), which is dominated by prior calcite precipitation (PCP; Allan et al., 2018; Baldini et al., 2006; Cruz et al., 2007; Fairchild and Treble, 2009; Fairchild et al., 2000; Griffiths et al., 2010; Johnson et al., 2006; Treble et al., 2003; Verheyden et al., 2000). The co-variation of the Baluk cave trace element ratios (Figure 3 and Table 2) resembles the long-term trends of the records from Kesang and Ton caves (Figure 5), and the co-variation is most likely attributed to PCP during the past 9370 years. In order to verify the domination of these records by PCP, we applied the logarithmic calculation method proposed by Sinclair (2011) and Day and Henderson (2013) to our results. As shown in Figure 4, the slope of the regression line of $\ln(\text{Sr/Ca})$ versus $\ln(\text{Ba/Ca})$ for BLK12A/H perfectly matches the slope expected for PCP dominance (1.02 ± 0.08 ; Sinclair, 2011). However, the correlation of $\ln(\text{Mg/Ca})$ versus $\ln(\text{Ba/Ca})$ of BLK12A/H is quite weak (Figure 4a), which is likely attributed to the hiatus (lack of data) during 2.8–1.1 ka. For BLK12B/C, the regression line slopes for $\ln(\text{Mg/Ca})$ versus $\ln(\text{Sr/Ca})$ and $\ln(\text{Sr/Ca})$ versus $\ln(\text{Ba/Ca})$ are very close to the expected slopes of 0.88 ± 0.13 (Sinclair, 2011) and 1.02 ± 0.08 (Day and Henderson, 2013), respectively, which indicates that the variations of Mg/Ca, Sr/Ca, and Ba/Ca are dominated by PCP. In addition, the variations in the profiles of the U/Ca ratios of BLK12A/H and BLK12B/C are also likely related to PCP (Cheng et al., 2016b), as the U/Ca ratios are positively correlated with Sr/Ca and Ba/Ca at a high confidence level ($r > 0.6$, $p < 0.001$) (Table 2). Because the regression line slopes of $\ln(\text{Sr/Ca})$ versus $\ln(\text{Ba/Ca})$ of BLK12A/H and

Figure 3. Comparison of profiles of trace element ratios from two combined stalagmites from Baluk cave: (a–d) BLK12A/H and (e–h) BLK12B/C. The dashed gray line shows the shift (at ~5 ka) from relatively high values (light orange shading) to relatively low values (light blue shading).

$\ln(\text{Mg}/\text{Ca})$ versus $\ln(\text{Sr}/\text{Ca})$ of BLK12B/C are closest to the expected slopes of PCP dominance (Figure 4), we consider that the variations in the Sr/Ca ratio of BLK12A/H and the Mg/Ca ratio of BLK12B/C are most strongly influenced by PCP. All of the aforementioned evidence supports our interpretation that PCP dominates the long-term variations of the Baluk cave trace element ratios during the Holocene.

As suggested in previous studies, PCP is a process whereby degassing in the vadose zone during periods of lower flow causes deposition of calcite and the disproportionate loss of Ca^{2+} ions (relative to Mg^{2+} and Sr^{2+}) along the flow path prior to reaching the stalagmite (Cruz et al., 2007; Griffiths et al., 2010; Johnson et al., 2006). Thus, under drier climatic conditions, water availability is reduced, PCP is enhanced, and CO_2 degassing is intensified,

resulting in the progressive enrichment of Mg, Sr, and Ba, relative to Ca in the seepage water. In contrast, under wetter climatic conditions, PCP is weakened and Mg, Sr, and Ba concentrations relative to Ca in the seepage water are decreased (Fairchild and Treble, 2009; Fairchild et al., 2000). In addition, under drier climatic conditions, seepage water residence time and water–rock interaction (WRI) in the aquifer are prolonged, leading to selective leaching of Mg and Sr from the dolomite host rock (Fairchild et al., 2000) and elevated Mg and Sr concentrations. Therefore, we infer that the intervals of higher trace element ratios of the Baluk cave stalagmites correspond to intervals of enhanced PCP, increased WRI, prolonged drip water residence time, and decreased moisture/precipitation in the study area, and vice versa. Notably, the interpretation of speleothem trace element ratios as a proxy of paleo-moisture/precipitation changes is demonstrated by

Table 2. Linear correlations of Mg/Ca, Sr/Ca, Ba/Ca, and U/Ca time series for stalagmites BLK12A/H and BLK12B/C from Baluk cave, northwest China.

	Mg/Ca	Sr/Ca	Ba/Ca	U/Ca
<i>BLK12A/H</i>				
Mg/Ca	1			
Sr/Ca	−0.17	1		
Ba/Ca	0.28**	0.65***	1	
U/Ca	−0.24*	0.84***	0.62***	1
<i>BLK12B/C</i>				
Mg/Ca	1			
Sr/Ca	0.61***	1		
Ba/Ca	0.51***	0.71***	1	
U/Ca	0.46***	0.85***	0.68***	1

*** $p < 0.001$; ** $p < 0.01$; * $p < 0.05$.

the results of cave monitoring studies (e.g. Fairchild et al., 2000; Johnson et al., 2006; Musgrove and Banner, 2004; Wong et al., 2011; Zhang and Li, 2019). For example, a recent 12-year (2005–2016) monitoring study from Furong cave in Chongqing, southwest China, demonstrated that trace element ratios (i.e. Mg/Ca, Sr/Ca, and Ba/Ca) mainly reflect changes in the precipitation amount (Zhang and Li, 2019). Therefore, our Baluk cave speleothem trace element ratios can be regarded as reliable proxies of past changes in moisture/precipitation.

Comparison with records of speleothem trace element ratios from two other caves in ACA

In Figure 5, the Sr/Ca ratio of BLK12A/H (Figure 5b) and the Mg/Ca ratio of BLK12B/C (Figure 5c) from Baluk cave are compared with equivalent records from Kesang cave in western Xinjiang and Ton cave in Uzbekistan (Cheng et al., 2016b). The Sr/Ca and Mg/Ca ratios of Baluk cave are high during ~9.0–5.0 ka and low from 5.0 ka to the present. This long-term decreasing trend of Baluk cave trace element ratios (ignoring the hiatus and superimposed suborbital fluctuations) is broadly consistent with the record of Sr/Ca ratios from both Kesang (Figure 5d) and Ton (Figure 5e) caves. For instance, the Sr/Ca ratio of Kesang cave is relatively high before ~7.6 ka with a major peak at ~8.1 ka, decreases gradually during 7.6–5.2 ka, and maintains low values after 5.0 ka (Figure 5d). Similarly, the profile of Sr/Ca ratios from Ton cave decreases gradually during 10.0 to ~4.0 ka with minor suborbital fluctuations superimposed, and slightly increased values thereafter (Figure 5e). Overall, the records of speleothem trace element ratios from the three caves in ACA – Ton cave in Uzbekistan (Central Asia), Kesang cave in western Xinjiang, and Baluk cave in central Xinjiang (NW China) – all exhibit a coherent long-term decreasing trend during the Holocene. This strongly

Figure 4. Scatter plots of logarithmically transformed Baluk cave trace element ratios. Correlations of $\ln(\text{Mg}/\text{Ca})$ versus $\ln(\text{Sr}/\text{Ca})$ and $\ln(\text{Sr}/\text{Ca})$ versus $\ln(\text{Ba}/\text{Ca})$ are shown for BLK12A/H (a, b) and BLK12B/C (c, d). The red lines show the linear regressions and the slopes are indicated by numbers, enabling comparison with the expected slopes based on thermodynamic and kinetic calculations if PCP is solely responsible for modulating the variability of the trace element ratios (Day and Henderson, 2013; Sinclair, 2011).

Figure 5. Comparison of speleothem trace element ratios from three well-separated caves in ACA. (b) Mg/Ca ratio of stalagmite BLK12B/C from Baluk cave (this study); (c) Sr/Ca ratio of stalagmite BLK12A/H from Baluk cave (this study); (d) Sr/Ca ratio from Kesang cave, NW China (Cheng et al., 2016b); (e) Sr/Ca ratio from Ton cave, Uzbekistan (Cheng et al., 2016b). For comparison, $\delta^{18}\text{O}$ record from Baluk cave is also plotted (a) (Liu et al., 2019, and this study). The dashed arrows indicate the long-term decreasing trends of speleothem trace element ratios at each cave site and the increasing trend of $\delta^{18}\text{O}$ from Baluk cave.

suggests that speleothem trace element ratios are reliable proxies of long-term moisture/precipitation variations in ACA during the Holocene. This pattern is characterized by a long-term trend of increasing mid-late Holocene moisture in this arid region. Thus, we argue that the spatially coherent long-term variation trends of speleothem geochemical records from caves separated by hundreds of kilometers represent moisture/precipitation variability on a large spatial scale, rather than local hydroclimatic variability, as was previously thought (Cheng et al., 2016b).

Inconsistency between the Holocene trends of speleothem trace element ratios and $\delta^{18}\text{O}$ records in ACA

As concluded above, speleothem trace element ratios from the caves of Baluk, Kesang, and Ton in ACA, which are separated by

distances of hundreds of kilometers, exhibit a consistent long-term decreasing trend, which represents a regional signal of increased mid-late Holocene moisture/precipitation in ACA (Figure 6a–c). However, the speleothem $\delta^{18}\text{O}$ records from the three caves exhibit different long-term trends during the Holocene (Cai et al., 2017; Cheng et al., 2012, 2016; Liu et al., 2019) (Figures 5a and 6e–g). For instance, the Baluk cave $\delta^{18}\text{O}$ records from stalagmites BLK12A and BLK12B (Liu et al., 2019) (Figure 6e) exhibit a gradually increasing trend from the early Holocene to the mid-late Holocene, with substantial centennial-scale oscillations superimposed. Similarly, the $\delta^{18}\text{O}$ records from Kesang cave (Figure 6f) are characterized by relatively low $\delta^{18}\text{O}$ values (–12‰ to –9‰) before ~3.0 ka with minor suborbital-scale fluctuations superimposed and relatively high $\delta^{18}\text{O}$ values (–9‰ to –6‰) after ~3.0 ka, with large-amplitude millennial- to centennial-scale oscillations superimposed (Cai et al., 2017; Cheng et al., 2012). In

Figure 6. Comparison of records of Holocene speleothem trace element ratios and $\delta^{18}\text{O}$ from three well-separated caves in ACA: (a) Speleothem Sr/Ca ratio from Baluk cave (this study); (b) speleothem Sr/Ca ratio from Kesang cave (Cheng et al., 2016b); (c) speleothem Sr/Ca ratio from Ton cave (Cheng et al., 2016b); (e) speleothem $\delta^{18}\text{O}$ records from Baluk cave (Liu et al., 2019); (f) speleothem $\delta^{18}\text{O}$ records from Kesang cave (Cai et al., 2017; Cheng et al., 2012); and (g) speleothem $\delta^{18}\text{O}$ record from Ton cave (Cheng et al., 2016b). Synthesis of Holocene variation in moisture conditions in the core area of ACA (Chen et al., 2016) (d), speleothem $\delta^{18}\text{O}$ record from Dongge cave, southern China (Wang et al., 2005) (h), and the variation of July NHSI at 65°N (Berger and Loutre, 1991) (i) are included for comparison. The dashed arrows illustrate the long-term trends of the records.

addition, the speleothem $\delta^{18}\text{O}$ from Ton cave in Uzbekistan increases gradually from -8‰ to -5‰ with minor centennial-scale fluctuations superimposed (Figure 6g) (Cheng et al., 2016b). This evidence strongly suggests that the long-term trend of speleothem $\delta^{18}\text{O}$ variation during the Holocene is approximately the inverse of the trend of variation of trace element ratios at the same site. Thus, the long-term trend of Holocene speleothem $\delta^{18}\text{O}$ variation in ACA is spatially inconsistent with that of speleothem trace element ratios. [AQ: 3]

Given that the record of speleothem trace element ratios is a reliable proxy of changes in moisture/precipitation at a large spatial scale, we propose that the apparent inconsistency mentioned above is mainly caused by the misinterpretation of speleothem $\delta^{18}\text{O}$ records in ACA. Notably, the climatic significance of speleothem $\delta^{18}\text{O}$ records in ACA has been thoroughly discussed in recent years (Cai et al., 2017; Chen et al., 2016; Liu et al., 2015b; Zhang and Jin, 2016; Zhang et al., 2017). Variations in moisture sources and related water vapor transport paths (Liu et al., 2015b; Zhang and Jin, 2016; Zhang et al., 2017), rather than the precipitation amount (Cheng et al., 2012), have been considered to be the primary factors determining long-term changes in precipitation $\delta^{18}\text{O}$ ($\delta^{18}\text{O}_p$) and speleothem $\delta^{18}\text{O}$ values in ACA. We assume that, during the early Holocene, in accord with the peaks in Northern Hemisphere summer insolation (NHSI; Figure 6i) (Berger and Loutre, 1991) and Northern Hemisphere temperature (Marcott et al., 2013), there was a weakening of both the latitudinal temperature gradient (Routson et al., 2019) and a northward shift of the mid-latitude westerlies (Jin et al., 2012). These changes led to the cave sites receiving an increased proportion of water vapor derived from distant high-latitude regions, with relatively low $\delta^{18}\text{O}_p$ values during the early Holocene, which was reversed when NHSI and Northern Hemisphere temperature subsequently decreased during the mid-late Holocene. This would indicate that the speleothem $\delta^{18}\text{O}$ records do not represent changes in the precipitation amount in ACA during the Holocene. Furthermore, the long-term trend of variation of Holocene speleothem $\delta^{18}\text{O}$ records in ACA is coincident with speleothem $\delta^{18}\text{O}$ records in the ASM region, for example, Dongge cave (Wang et al., 2005) (Figure 6h). This implies that Holocene speleothem $\delta^{18}\text{O}$ records in both ACA and the ASM region document a supra-regional pattern of climatic variability of atmospheric circulation-determined $\delta^{18}\text{O}_p$ values controlled by NHSI on the orbital scale (Cheng et al., 2016a, 2016b; Liu et al., 2015a; Sun et al., 2018).

Numerous studies have shown that the environment in ACA was relatively dry during the early Holocene and wet during the mid-late Holocene (e.g. An et al., 2011; Chen et al., 2008, 2016; Hong et al., 2014; Huang et al., 2009; Jiang et al., 2012; Leroy et al., 2014; Liu et al., 2008; Long et al., 2017, 2014; Ran and Feng, 2014; Wang and Feng, 2013; Wang et al., 2013; Xie et al., 2018; Zhang et al., 2018a). This pattern has been called a 'westerlies-dominated climatic regime' in terms of its influence on Holocene climate change in ACA (Chen et al., 2008, 2009). The spatially coherent trend of increasing mid-late Holocene moisture conditions, suggested by the speleothem geochemical data from our Baluk cave and the two other caves, provides new evidence for a 'westerlies-dominated climatic regime', especially the persistent wetting trend in the core region of ACA (Huang et al., 2015) during the Holocene (Chen et al., 2016) (Figure 6d).

Conclusion

Based on precise U–Th age controls, we have obtained profiles of speleothem trace element ratios (Mg/Ca, Sr/Ca, Ba/Ca, and U/Ca) from Baluk cave in Xinjiang, NW China, which document the long-term evolution of moisture conditions in the arid Xinjiang region during the past 9370 years. The main conclusions of the study are as follows:

1. Our records from Baluk cave reveal the long-term co-variation of trace element ratios. In addition, in biplots of logarithmically transformed element ratios, the slopes of the regression lines suggest that the trace element ratios are dominated by PCP. Therefore, trace element ratios can be used as reliable proxies of changes in moisture/precipitation. In addition, the trace element ratios from Baluk cave indicate a trend of increasing mid-late Holocene moisture in ACA.
2. The spatial coherence of long-term variations in speleothem trace element ratios from three caves that are separated by hundreds of kilometers (Baluk, Kesang, and Ton caves) suggests that the speleothem trace element ratios represent a supra-regional signal of increasing mid-late Holocene moisture in ACA, rather than local hydroclimatic variability as was previously thought.
3. The long-term trend of variation of the speleothem trace element ratios at the three caves is inconsistent with their respective $\delta^{18}\text{O}$ records. This indicates that the speleothem $\delta^{18}\text{O}$ records do not represent changes in the precipitation amount in ACA during the Holocene; rather, they represent changes in moisture sources controlled by NHSI on an orbital timescale. The spatial coherence of a long-term wetting trend revealed by the speleothem trace element ratios from three different cave locations provides new evidence for the importance of a 'westerlies-dominated climatic regime' in influencing Holocene climate change in ACA.

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Data availability

The data from Baluk cave referenced in this study can be accessed from <https://figshare.com/s/f5b92b3b66396fce8340>.

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