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The palaeoenvironmental significance of $\delta^{13}C$ of stalagmite BW-1 from Beijing, China during Younger Dryas intervals inferred from the grey level profile

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High-resolution records of carbon isotope composition and grey level were analysed from a stalagmite, BW-1, from Beijing, China, deposited between *c.* 14 and 10.5 ka BP, the $\delta^{18}O$ profile of which has been used to discuss the timing and structure of the Younger Dryas (YD) event in north China. The high grey level and low $\delta^{13}C$ match the milk-white coloured locations on the polished stalagmite surface and coincide with enhanced luminescent bands within which the concentration of both impurities and the total organic carbon (TOC) are high. Additionally, the fluorescence of speleothems was derived from organic acids that have been flushed onto the stalagmite surface along with impurities from the overlying soil by heavy summer rain and co-precipitated with the speleothem calcite. Thus, predominantly low $\delta^{13}C$ and high grey level values indicate increased summer precipitation that supports abundant vegetation and robust biological productivity. Consequently, three distinct time intervals are defined by the palaeoenvironmental conditions expressed in the $\delta^{13}C$ and grey level records of stalagmite BW-1: (i) a warm-humid stage (Pre-YD, 13.97 to 12.85 ka BP, including a hiatus from 12.99 to 13.21 ka BP reported before); (ii) a cool-arid stage (YD, 12.85 to 11.56 ka BP); and (iii) a warm-humid stage (Post-YD, 11.56 to 10.39 ka BP). The inferences based on our research are generally consistent with other regional vegetation and climatic records.

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Previous research on the $\delta^{18}O$ profile of a stalagmite, BW-1, from Kulishu cave, Beijing, China, has discussed the timing and structure of the Younger Dryas event (YD) in north China (Ma *et al.* 2012). However, the $\delta^{13}C$ of the stalagmite has not been utilized to interpret the palaeoenvironmental signals. Changes in $\delta^{13}C$ of stalagmite can be controlled by complicated processes including changes in vegetation C3/C4 ratio, which leads to changes in $\delta^{13}C$ of soil CO_2 (Dorale *et al.* 1992, 1998; Denniston *et al.* 2000), variation in the soil pCO_2 due to the density of vegetative cover and biomass (Hesterberg & Siegenthaler 1991; Amundson *et al.* 1998; Genty *et al.* 2003), the degree of mixing between atmospheric CO_2 and CO_2 derived from root respiration and microbial activity (Baker *et al.* 1997), variations in the amount of degassing due to changes in cave air pCO_2 (Spötl *et al.* 2005; Dreybrodt & Scholz 2011; Deininger *et al.* 2012), and the amount of prior calcite precipitation in the unsaturated zone of karstic aquifers (Baker *et al.* 1997; Verheyden *et al.* 2000). It is therefore necessary to utilize multiple proxies from the same stalagmite to precisely explore the significance of the $\delta^{13}C$ record. Time series of $\delta^{13}C$ can then be combined with other proxies, such as $\delta^{18}O$ (Dorale *et al.* 1998; Cruz *et al.* 2006; Cosford *et al.* 2009; Baker *et al.* 2011; Cui *et al.* 2012), trace elements (Johnson *et al.* 2006; Cruz *et al.* 2007; Cui *et al.* 2012), grey level (Cui *et al.* 2012; Gu & Wu 2012), and speleothem growth rates (Plagnes *et al.* 2002; Drysdale *et al.* 2004; Cruz *et al.* 2006) to discuss the climatic or environmental significance. Amongst these proxies of the stalagmite, the grey level is the easiest and cheapest to measure. The stalagmite grey level essentially reflects the changes in the ratio of impurities to pure calcite. High grey level values correspond to the milk-white coloured interval on the polished surface of the stalagmite (the impurity concentration is high), whereas low values match the transparent-brown interval (the impurity concentration is low) (Wu *et al.* 2006; Cui *et al.* 2012; Gu & Wu 2012). Furthermore, some previous studies have suggested that the impurities are flushed onto the stalagmite surface with dissolved organic carbon (DOC; the luminescent material), in most cases by heavy summer precipitation (Ban *et al.* 2008; Orland *et al.* 2012). Thus, the stalagmite grey level is related to impurity concentration and can be used to reflect the precipitation intensity. Accordingly, in this study, we have tried to interpret the palaeoenvironmental significance of the $\delta^{13}C$ data set from stalagmite BW-1 (Ma *et al.* 2012) by comparing it with the grey level record from the same stalagmite. Furthermore, the characters of

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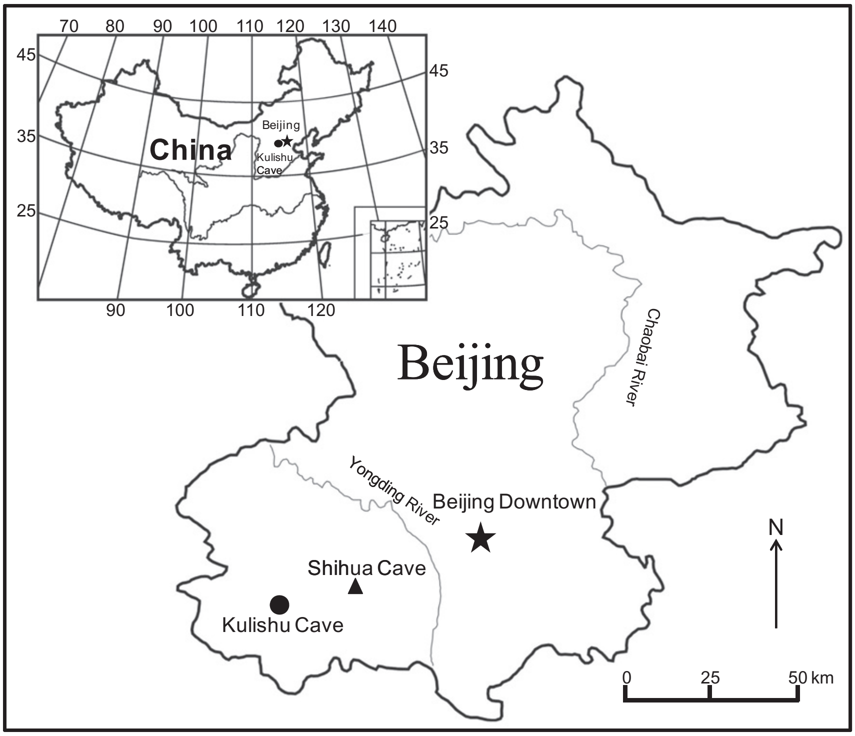

Fig. 1. Location of Kulishu Cave.

the stalagmite fluorescence and the total organic carbon (TOC) concentrations in the milk-white coloured interval and transparent-brown interval, respectively, of the stalagmite have been qualitatively analysed.

Cave location and sample

Kulishu Cave (latitude 39°41′N, longitude 115°39′E, altitude 610 m a.s.l.) is located about 80 km SW of downtown Beijing (Fig. 1). The cave developed at a depth of nearly 60 m below the surface in Middle Proterozoic dolomite. A stalagmite, BW-1, was collected from Kulishu Cave about 40 m from the entrance in October 1999 and the sample is 19.5 cm high and 10 cm wide (Ma *et al.* 2012). Over the cave, the vegetation is dominated by secondary-growth deciduous broadleaf trees and shrubs. Located within the northern boundary of the East Asian monsoon in the northwestern arid-semiarid zone, the area around Kulishu Cave typically has cold/dry winters and warm/wet summers and about 74% of the total annual precipitation falls during summer (June to August). The mean annual temperature and precipitation are 12.3°C and 570 mm, respectively (1971–2000 averages, according to the Chinese Meteorological Administration data at http://www.cma.gov.cn/) (Ma *et al.* 2012).

Methods

The chronology and stable isotope ($\delta^{18}O$ and $\delta^{13}C$) analysis of stalagmite BW-1 from Kulishu Cave have already been stated in detail (Ma *et al.* 2012).

The grey level was measured on a scanned image of the polished stalagmite surface taken by a high-resolution scanner (Microtek) under RGB/3200 dpi conditions. The data were obtained from every 8 μm along the growth axis using IMAGE-PRO PLUS 5.1 software, and were calculated from the intensities of red (R), green (G), and blue (B) light (grey level= 0.299R+0.587G+0.114B) (Peli 1992; Muangsong *et al.* 2011). In order to be compared with the $\delta^{13}C$ profile easily, the raw grey level data were calculated with a 37-point moving average. Additionally, to calculate the correlation coefficient between grey level and $\delta^{13}C$, we averaged the grey level data around each point where the powder subsample was taken for $\delta^{13}C$ analysis. Values of grey level range from 0 to 255, and indicate the optical density of reflected light of scanned images. The higher the grey level, the brighter the surface colour. One big thin section was cut to observe the character of the stalagmite laminae under a BX60 Olympus microscope equipped with UV-excitation. Moreover, two subsamples were drilled from the milk-white coloured and dark-transparent intervals, respec-

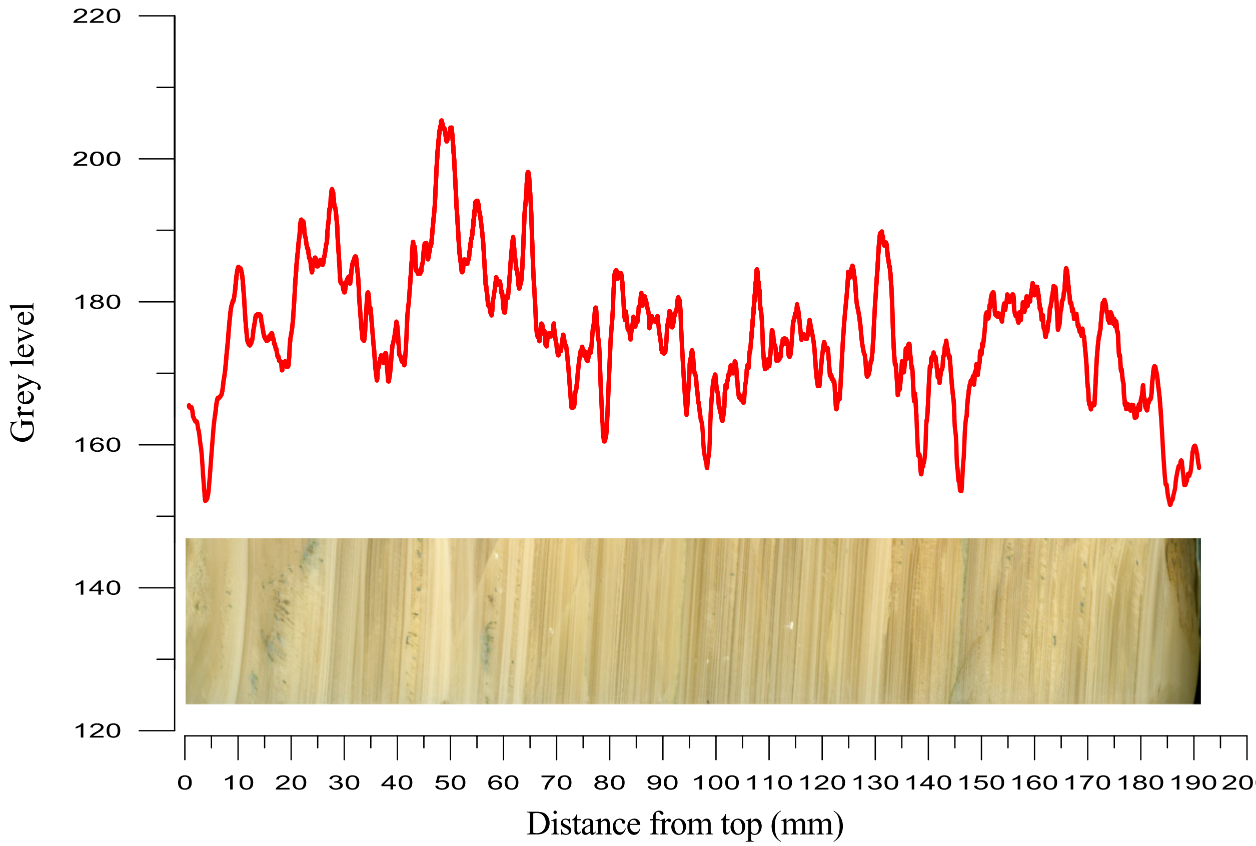

Fig. 2. Plot of grey level of stalagmite BW-1 versus distance from the top. The stalagmite section is shown at the bottom for comparison. The high grey level values match the milk-white coloured intervals, whereas the low grey level values match the transparent-brown calcite locations. At the very top and bottom of the stalagmite profile, the grey level may be lower than the true values (see main text for details). This figure is available in colour at http://www.boreas.dk.

tively, for TOC concentration analysis. The details of the method are as follows. Firstly, the powder of the subsample was weighed and put on the 0.526 cm^2 circular quartz filter (0.4 µm pore size, Whatman), which was prefired for at least 3 h at 850°C to remove adsorbed organic vapours. Secondly, one drip of distilled water was placed on the surface of the subsample and then the filter was put into an oven at 50°C for 24 h to dry the filter and to make the sample powder stick firmly onto the filter. Finally, the dried filter was placed in a quartz boat and put into the oven of A DRI Model 2001Thermal/Optical Carbon Analyzer. While the oven temperature was stepwise heated to 120, 250, and 450°C in a pure He environment, three OC fractions were produced: OC1, OC2, and OC3, respectively. TOC, short for TOC concentration, was calculated as OC1+OC2+OC3 divided by the weight of the subsamples.

The above analytical work was carried out in the Key Laboratory of Cenozoic Geology Environment, Institute of Geology and Geophysics, Chinese Academy of Science, China.

Results

Chronology

Following the ^{230}Th dating results and the age model performed in the previous study (Ma *et al*. 2012), the BW-1 record covers a time period from 13.97 to 10.39 ka BP with a hiatus from 12.99 to 13.21 ka BP. Accordingly, the average growth rate is ~76 µm a^{-1}, and the average sampling interval for the stable isotope analyses and grey level is ~14 years and ~1 month, respectively.

Grey level profile and TOC concentrations

The observed grey level values of stalagmite BW-1 varied between 138 (darker) and 214 (brighter), averaging 180, which reflect visible changes in the material constitution on the polished stalagmite surface profile. The high grey level values match the milk-white coloured intervals on the polished stalagmite surface (Fig. 2), where the concentration of opaque impurities

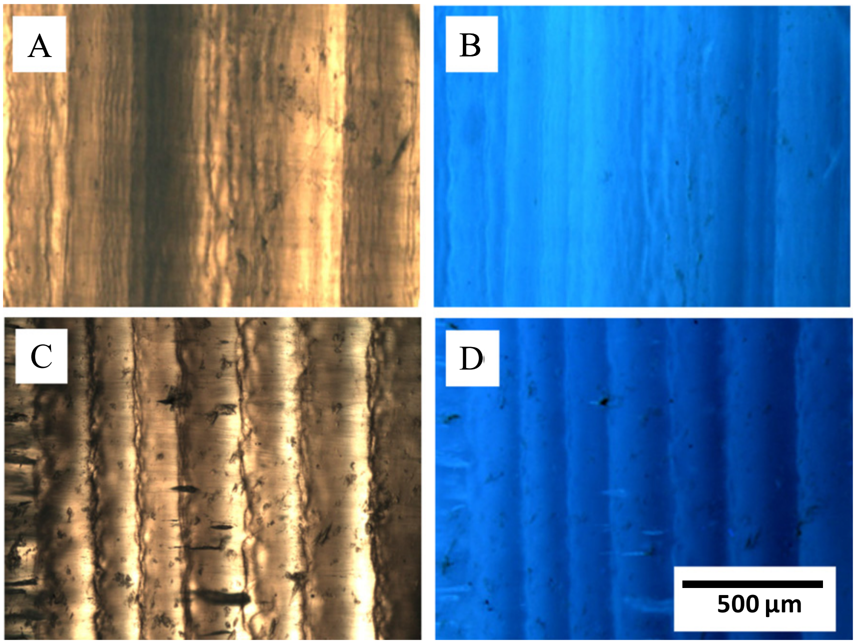

Fig. 3. Micrographs from stalagmite BW-1. A and B show micrographs of the high grey level laminae in stalagmite BW-1 under a microscope with transmitted and fluorescent light, respectively; C and D show micrographs of the low grey level laminae under a microscope with transmitted and fluorescent light, respectively.

is high (thin section observed under transmitted light microscope, Fig. 3A), whereas the low grey level values are associated with transparent-brown calcite locations (Fig. 2), where the opaque impurities are few (Fig. 3C). The grey level values of the YD period, between 12.85 and 11.56 ka BP, averaging 170, are lower than the average of the whole profile. At the very top and bottom locations of the stalagmite surface profile, the observed grey level values may be lower than the true values. This is because the stalagmite surface is not absolutely smooth, which reduces the reflected light intensities at those locations. Moreover, the TOC concentration of the milk-white coloured interval is 2.5 µg mg^{-1}, which is much higher than the transparent-brown interval (0.6µg mg^{-1}).

δ^{13}*C profile*

The $\delta^{13}C$ record varies from –7.19 to –11.01‰, with an overall mean of –9.5‰. The complete $\delta^{13}C$ time series, plotted in Fig. 4, displays several prominent centennial- to decadal-scale oscillations. Between 13.97 to 12.85 ka BP (Pre-YD), the $\delta^{13}C$ record averages –9.78‰, almost equivalent to the average value of the whole record. During the YD period, between 12.85 and 11.56 ka BP, the $\delta^{13}C$ values are much heavier, with an average of –8.85‰ and a series of fluctuations. Prominent positive excursions are centred at ~12.24, ~11.90, and ~11.60 ka BP and slightly negative excursions at ~12.45 and ~11.80 ka BP. After YD until the termination of this record, from 11.56 to 10.39 ka BP, the $\delta^{13}C$ value averages –9.91‰, slightly lower than the average value of the whole record.

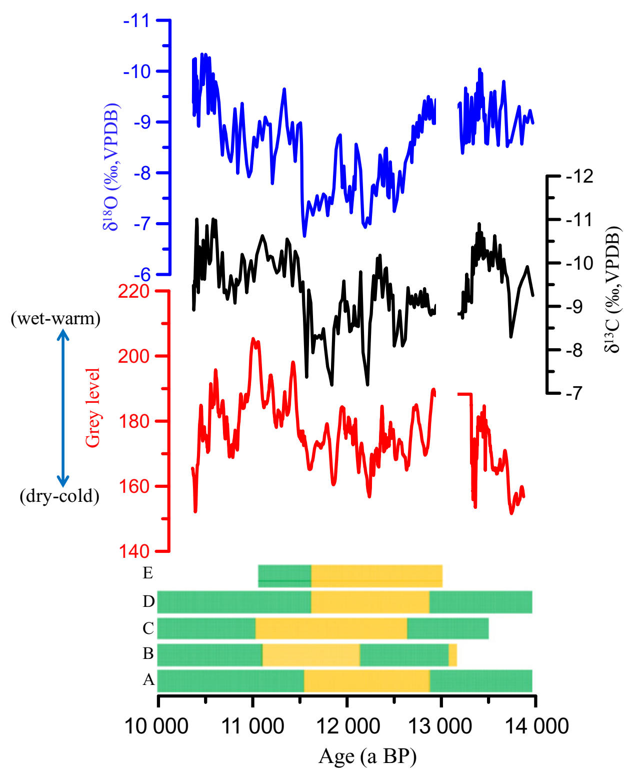

Fig. 4. Comparison between multiproxy records of stalagmite BW-1 and other regional climatic records. A. Jalai Nur (Wang *et al.* 1994). B. Donggan Lake (Wei *et al.* 1997). C. Ningjin Lake (Guo *et al.* 2000). D. Waqie profile (Zhou *et al.* 2001a). E. Changjiang and Huanghe sediments (Yi & Saito 2004). Green and orange bars represent wet and dry periods, respectively. This figure is available in colour at http://www.boreas.dk.

Discussion

The palaeoenvironmental significance of $\delta^{13}C$ of stalagmite BW-1 inferred from the grey level profile

The significance of the carbon isotope composition of the stalagmite is related to the sources of dissolved carbon in the drip-water, including soil CO_2 and carbonate bedrock (Hendy 1971; Genty *et al.* 2001).

Soil CO_2 is affected by the rate of biogenic CO_2 supply from root transpiration, the rate of organic matter decomposition (Linge *et al.* 2001; Frappier *et al.* 2002), and possibly the type of vegetation cover (Dorale *et al.* 1998; Denniston *et al.* 1999). The vegetation above Kulishu cave is now mainly dominated by C3 plants, secondary-growth deciduous broadleaf trees and shrubs (Ma *et al.* 2012). In addition, the $\delta^{13}C$ record of stalagmite BW-1, ranging from –7.19 to –11.01‰, falls just within the field of speleothem $\delta^{13}C$ expected at a site overlain by C3 vegetation (–14 to –6‰) (McDermott 2004). In regions where vegetation type is predominantly C3, the influence of vegetation on speleothem $\delta^{13}C$ primarily reflects changes in the density of vegetative cover and biomass (Baker *et al.* 1997; Baldini *et al.* 2005; Cosford *et al.* 2009). Therefore, Kulishu stalagmite $\delta^{13}C$ may be controlled primarily by the vegetative productivity rather than the vegetation C3/C4 ratio.

Climatic conditions, in particular temperature and precipitation, affect the vegetation density, biological productivity, and the $\delta^{13}C$ of plants above a cave, particularly in semiarid climates. During periods of greater precipitation and higher temperature, on the one hand, the $\delta^{13}C$ of plants decreases significantly (Ren & Yu 2011); on the other hand, plant cover and biological activity increase, which raises soil CO_2 production leading to a greater proportion of soil CO_2 dissolved in seepage waters. As kinetic fractionation in biological processes favours ^{12}C, organically derived CO_2 released to the soil through root respiration and microbial decomposition of organic matter is relatively depleted in ^{13}C. Accordingly, inheriting from the CO_2 dissolved in soil water and dripwater, the stalagmite $\delta^{13}C$ will be lower and vice versa (Bar-Matthews *et al.* 2003; Drysdale *et al.* 2004; Cosford *et al.* 2009). Additionally, inorganic processes that respond to climatic conditions also contribute to the $\delta^{13}C$ values of stalagmites. Under wetter conditions, increased drip rates result in lower stalagmite $\delta^{13}C$ values owing to less time for $CaCO_3$ precipitation on both the unsaturated zone of karstic aquifer (Baker *et al.* 1997) and the stalagmite surface (Bar-Matthews *et al.* 1996; Mickler *et al.* 2004, 2006; Cosford *et al.* 2009; Scholz *et al.* 2009; Dreybrodt & Scholz 2011; Deininger *et al.* 2012). As the biological activities and inorganic processes drive stalagmite $\delta^{13}C$ in the same 'direction', lower $\delta^{13}C$ values of stalagmites reflect relatively

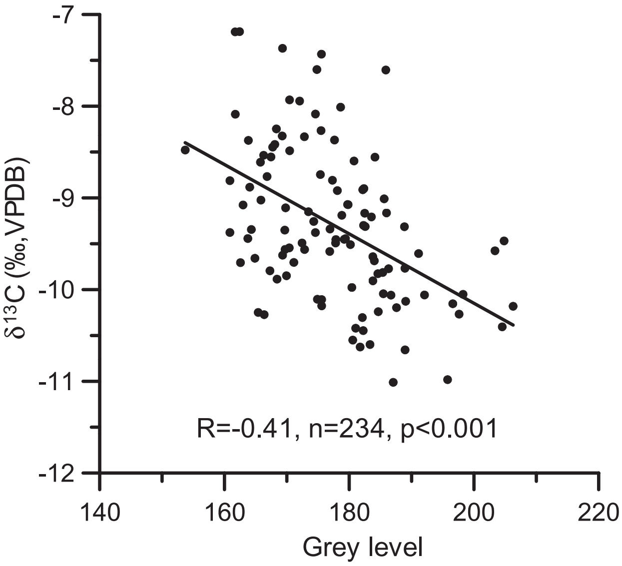

Fig. 5. The correlation between $\delta^{13}C$ (‰, VPDB) and the grey level of stalagmite BW-1.

increased precipitation and higher temperature, and vice versa (Cosford *et al.* 2009).

The grey level, to some extent, supports the idea that the $\delta^{13}C$ record of stalagmite BW-1 can be interpreted as the variations of precipitation and temperature. There is a significantly negative correlation between $\delta^{13}C$ and grey level (R=–0.41, n=234, p<0.001) (Fig. 5), which suggests that an identical physical process has controlled the variations in the two proxies over most of the time since formation. The low $\delta^{13}C$ and high grey level match the milk-white coloured intervals on the polished stalagmite surface (Fig. 2), where the concentrations of both opaque impurities (thin section observed under transmitted light microscope, Fig. 3A) and TOC are higher than the transparent-brown locations (Fig. 3C). Additionally, similarly to Shihua cave, the opaque impurities are often, but not always, coincident with enhanced luminescent bands under reflected-light microscopy with UV-excitation (Fig. 3B, D; Tan & Liu 2003; Cai *et al.* 2010). Some studies have proved that the fluorescence of speleothems is derived from organic acids that have been carried by groundwater from the overlying soil, and co-precipitated with the speleothem calcite (McGarry & Baker 2000; Orland *et al.* 2012). Therefore, we infer that the impurities may be flushed onto the stalagmite surface with DOC (the luminescent material) by heavy summer precipitation in most cases (Ban *et al.* 2008; Orland *et al.* 2012). As for the high $\delta^{13}C$ and low grey level, they match the transparent-brown locations of stalagmite surface (Fig. 2), where the concentrations of both opaque impurities (Fig. 3C) and TOC are very low, inferring that the frequency of high intensity of summer precipitation is low. In brief, the $\delta^{13}C$ record of stalagmite

BW-1 reflects vegetative productivity and inorganic processes, which depend upon on the variations of precipitation and temperature. The lower the $\delta^{13}C$ value, the higher the temperature and the precipitation, and vice versa.

Palaeoenvironment changes across the YD in Beijing implied by the $\delta^{13}C$ and grey level records of stalagmite BW-1

Three distinct time intervals are defined by the palaeoenvironmental conditions expressed in the $\delta^{13}C$ and grey level records of stalagmite BW-1: (i) a warm-humid stage (Pre-YD, 13.97 to 12.85 ka BP, including a hiatus from 12.99 to 13.21 ka BP reported before); (ii) a cool-arid stage (YD, 12.85 to 11.56 ka BP); and (iii) a warm-humid stage (Post-YD, 11.56 to 10.39 ka BP).

During the first stage (Pre-YD, 13.97 to 12.85 ka), including a hiatus correlated to the intra-Allerød cold period (Ma *et al.* 2012), as discussed above, the grey level may be lower than the real value and cannot be used to explore the signal of the $\delta^{13}C$ in this section. Nevertheless, the relatively low $\delta^{13}C$ value resulting from a lower degree of mixing between atmospheric and biological CO_2 may partly infer increased vegetation and robust biological productivity supported by warm and humid conditions. Allowing for the chronology uncertainties, this inference is in good agreement with previous studies, such as lake sediment profiles (Wang *et al.* 1994; Wei *et al.* 1997; Guo *et al.* 2000) and loess paleosol sequences (Zhou *et al.* 2001b), all of which infer the relatively warm and wet climatic conditions in Beijing and the adjacent area before the YD (Fig. 4).

During the second stage (12.85 to 11.51 ka BP), corresponding to the YD event, the relative high $\delta^{13}C$ and low grey level reflect a cold-dry episode, with a series of climatic fluctuations inferred by the large variability in $\delta^{13}C$ and grey level. Extreme climatic instability, in particular severe drought intervals, occurred at *c.* 12.24, *c.* 11.90, and *c.* 11.60 ka BP. The high $\delta^{13}C$ values may result from the lower drip rates, consistent with decreased precipitation, and more time for $CaCO_3$ precipitation in the epikarst. Besides, two slightly wetter intervals are centred at *c.* 12.45 and *c.* 11.80 ka BP, resulting in relatively robust biological productivity and increased drip rates.

Other palaeoclimate records from Beijing and the adjacent area also demonstrated a general cold-dry period during the YD event, such as the rapid decline in pollen and charcoal concentrations in Beijing (Zhang *et al.* 1996), an increase in herbaceous pollen in northern China (Sun & Chen 1991; Guo *et al.* 2000; Yi & Saito 2004), and lowered lake levels in both the Beijing area (Wei *et al.* 1997) and adjacent Inner Mongolia (Wang *et al.* 1994; Peng *et al.* 2005). Moreover, the YD sequences from Loess Plateau records (Zhou *et al.* 2001a) reflect an initial cold, dry phase, *c.* 12 900 to *c.* 12 400 cal. a BP, followed by a relative humid phase, from *c.* 12 420 to 11 960 cal. a BP, then an extremely cold and dry phase, from *c.* 11 960 to 11 500 cal. a BP, which is in good agreement with our inferences (Fig. 4).

The final stage began at 11.51 ka BP, the termination of the YD, when the $\delta^{13}C$/grey level jump abruptly to much lighter/higher values within about 38 years, based on the laminae counts on the reflected image of the stalagmite profile (Ma *et al.* 2012). This suggests that the climate changed from the cold-dry (YD) to warm-wet conditions (normal) very sharply, accompanied by the abruptly increased biological productivity and drip rates. From then to the termination (10.39 ka BP) of our record, the relatively low $\delta^{13}C$ and high grey level infer in general warm and wet conditions. The vegetation of the Zhaitang area of Beijing changes from temperate grassland to temperate meadow steppe following the YD (Xia *et al.* 2012). In addition, in the record of Ningjin Lake, in Hebei Province, the pollen species increased abruptly and some hydrophyte pollens were found after the YD event, inferring a warm and wet period at this time (Guo *et al.* 2000) (Fig. 4).

Conclusions

Evaluation of $\delta^{13}C$ together with the grey level of the stalagmite BW-1 from Kulishu cave, Beijing, China, suggests that the $\delta^{13}C$ reveals shifts of both vegetative productivity and inorganic processes, which are in turn affected by temperature and precipitation. Thus, the $\delta^{13}C$ and the grey level can provide insights on the history of vegetation and climatic conditions. Higher temperature and precipitation support robust vegetation and biological activity and increased drip rates, which will result in lower $\delta^{13}C$ and higher grey level values of stalagmite. By contrast, cooler and drier conditions result in diminished vegetative cover, increased mixing of atmospheric CO_2, and lowered drip rates, which favour higher $\delta^{13}C$ and lower grey level values of stalagmite. Accordingly, palaeoenvironmental conditions expressed in the $\delta^{13}C$ and grey level records of stalagmite BW-1 define three distinct intervals: (i) a warm-humid stage (Pre-YD, 13.97 to 12.85 ka BP, including a hiatus from 12.99 to 13.21 ka BP reported before); (ii) a cool-arid stage (YD, 12.85 to 11.56 ka); and (iii) a warm-humid stage (Post-YD, 11.56 to 10.39 ka) that is consistent with regional vegetation and climatic variability expressed in other palaeoclimatic records.

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