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RESEARCH LETTER

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The Climate Control of Soil Organic Carbon Dynamics Inferred From Speleothem Radiocarbon Ages

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Key Points:

- Using ^{14}C ages of dissolved organic carbon in speleothem as an alternative approach to date the speleothem is questionable
- The deviations of speleothem $^{14}C_{DOC}$ from the U-Th ages can be used to infer the turnover of the soil organic carbon (SOC)
- Climate dominates the speleothem $^{14}C_{DOC}$ ages and SOC turnover on spatio-temporal scales

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract The complexity of processes affecting soil organic carbon (SOC) turnover on spatio-temporal scales often hinders the extrapolation of results from specific sites to larger scales. This study presents Holocene speleothem U-Th ages paired with ^{14}C ages of carbonate and dissolved organic carbon (DOC) through three caves located on a north-south transect through China. The deviations of speleothem $^{14}C_{DOC}$ ages from the U-Th ages show clearly spatial variability, and they are positively correlated with mean ages of modern SOC and soil turnover time, suggesting that deviations can be used to infer the SOC turnover. We further demonstrate that slow SOC turnover (large deviation) was associated with weak monsoon (low temperature/less precipitation) on temporal scales. Our findings reveal that climate dominates the speleothem $^{14}C_{DOC}$ ages and SOC turnover. As global warming likely will intensify, the accelerated turnover of SOC, particularly at higher latitude areas, may partially offset the existing soil carbon stock.

Plain Language Summary Understanding the controls of the soil organic carbon (SOC) turnover on spatio-temporal scales is of great significance to predict the soil carbon stock. The current knowledge of SOC turnover on the longer temporal scale is scarce due to a lack of accurate chronology. Here, we present Holocene speleothem U-Th ages paired with ^{14}C ages of carbonate and dissolved organic carbon (DOC) through three caves located on a north-south transect through China. We found the deviations of speleothem $^{14}C_{DOC}$ ages from the baseline U-Th ages positively and negatively correlated with soil turnover time and climate variables (i.e., temperature and precipitation), respectively. Concretely, long turnover time as indicated by large deviations was associated with weaker monsoon (low temperature and less precipitation), and vice versa. It suggests that climate dominates SOC turnover and the speleothem $^{14}C_{DOC}$ ages. Our study not only provides a compelling approach to diagnose the past cycling of SOC under climate shifts exceeding present-day variability and but also projects a promising perspective for studying the interactions between SOC turnover rates and climate changes on broad spatio-temporal scales. Meanwhile, it creates an opportunity to evaluate the changes in SOC turnover rates for the future.

1. Introduction

The terrestrial carbon cycle is crucial to the evolution of atmospheric greenhouse gases (Eglinton et al., 2021; Hein et al., 2020). The amount of organic carbon stored in soils (SOC), in particular, plays a key role, not only because soils store the largest amount of organic carbon in the terrestrial system, but also because the SOC cycle regulates ecosystems (Wang et al., 2021). The turnover rate of SOC is essential to understand and quantify the soil carbon storage capacity (Friedlingstein et al., 2006; Shi et al., 2020) and fluxes to and from the atmosphere (Carvalhais et al., 2014; Eglinton et al., 2021). Climatic factors, including precipitation and temperature, have been proposed to dominate soil turnover rates (Carvalhais et al., 2014; Schimel et al., 1994; S. E. Trumbore et al., 1996) as both affect the decomposition of SOC via microbial respiration (Carvalhais et al., 2014; Eglinton et al., 2021; Hein et al., 2020; Knorr et al., 2005).

Previous studies have shown significant spatial and geographical heterogeneity in SOC storage and turnover rates (Carvalhais et al., 2014; Eglinton et al., 2021), while the driving forces and the extent of turnover rates are

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debated, and sometimes are even contradictory in different studies (Conant et al., 2011; Wang et al., 2018). This is because different field experiments were conducted under specific environmental conditions and some results attained from modern incubation experiments with conditions different from real field environment (Conant et al., 2011; Eglinton et al., 2021; Hein et al., 2020; Wang et al., 2018). This limits our ability to extrapolate such studies to large spatial and temporal scales. In particular, on the temporal scales, the carbon cycling study with precisely dated chronology is still scarce (Hein et al., 2020). Resolving these issues is necessary to reliably assess soil carbon storage and its role in the terrestrial carbon cycle under future warming climate.

Cave carbonate deposits (speleothems), especially stalagmites, are a remarkable paleoenvironmental archive. On the basis of highly precise U-series ages (Bajo et al., 2020; Cheng et al., 2016), past information about climatic and environmental changes can be reconstructed. Organic matter (OM), as an important climate indicator in paleoclimatic studies, is also preserved in trace amounts in speleothems (Blyth et al., 2016). Amongst the various organic components, dissolved organic carbon (DOC) largely carried by percolating water from surface soil (Baker & Genty, 1999; Baker et al., 1998; Ban et al., 2008; ; Liao et al., 2018; van Beynen et al., 2002) accounts for the largest proportion of stalagmite organic matter (OM) (Blyth et al., 2016). Its composition and age will likely mirror the composition and age of soil OM, as soil DOC mainly originates from decomposition of stable soil OM, and is in isotopic and chemical equilibrium with bulk soil OM (Gmach et al., 2020; Kaiser & Kalbitz, 2012; Sanderman et al., 2008). SOC ^{14}C ages increase proportionally with SOC turnover times (Eglinton et al., 2021) and can be used to deduce SOC turnover rates and evaluate the carbon sequestration potential of a soil (Eglinton et al., 2021; Hein et al., 2020; Shi et al., 2020). As ^{14}C ages of speleothem DOC ($^{14}C_{DOC}$) may reflect the age of soil DOC, they could provide a powerful means to reveal the past cycling of SOC under climate shifts exceeding present-day variability.

Here, we present new ^{14}C ages of both carbonate ($^{14}C_{carb}$) and DOC from five stalagmites from three caves situated along a north-south transect in China, that is, Longfeng Cave (LF19, LF27, LF32), Jiuxian Cave (JX) (C996-1) and Shennong Cave (SN-27) (Figure 1 and Figure S1 in Supporting Information S1). The ^{14}C ages are paired to previously (Cai et al., 2010; Wei et al., 2020) and newly (Table S1 in Supporting Information S1) obtained, high precision U-Th ages, allowing us a comparison between ^{230}Th and ^{14}C ages on spatial and temporal scales. Using the ^{230}Th dates as baseline ages, the deviation of $^{14}C_{DOC}$ ages is used to evaluate whether speleothem $^{14}C_{DOC}$ ages can be used as an alternative dating method to U-Th (Akers et al., 2019; Blyth et al., 2017; Borsato et al., 2000; Genty et al., 2011; Lechleitner et al., 2019), and to investigate potential controls on speleothem $^{14}C_{DOC}$ ages related to soil and karst SOC cycling (Lechleitner et al., 2019).

2. Materials and Methods

The study samples were collected from Longfeng Cave (LF19, LF27, and LF32), JX (C996-1) and Shennong Cave (SN-27), these caves are located in the different positions with varied climate and environment conditions (Figure 1 and Text S1 in Supporting Information S1). Longfeng Cave (113.55°E, 38.68°N, ∼1,456 m above sea level, asl, Wei et al., 2020) is located at the foot of Wutai Mountain in Xinzhou city, Shanxi Province, northern China. JX (33°34′N, 109°6′E, elevation = 1,495 m asl) is located on the south slope of the Qinling Mountains in central China (Cai et al., 2010). Shennong cave (28°42′N, 117°15′E, elevation = 383 m asl) is located in the northeast of Jiangxi Province, southern China (Zhang et al., 2015). Notably, both annual mean precipitation and temperature increase from higher to lower latitude cave sites, and the detailed information about these caves and local climates can refer to the mentioned literature.

The accelerator mass spectrometry (AMS) ^{14}C dating subsamples for DOC were cut from near the growth axis of the stalagmites. The subsamples are solid cuboids with the weight of ∼2.5 g. To completely remove inorganic carbon and any external contamination, essential pre-combusting the glassware and pre-cleaning the surface of subsamples (Lechleitner et al., 2019) were implemented. Then, subsamples were digested by using 3M ortho-phosphoric acid solution. After that, the digestion solutions were added 3 mL of supersaturated potassium persulfate oxidizing solution (100 ml super cleaned water+4 g $K_2S_2O_8$ (Sigma-Aldrich, 99.99%) + 20 μL H_3PO_4) (Lang et al., 2012) to oxidize the DOC to CO_2 at 99°C for 1–2 hr. The CO_2 was then purified and cryogenically transported to a storage vessel for graphitization. See Text S2 in Supporting Information S1 for detailed information.

For the $^{14}C_{carb}$, the entire speleothem samples were pre-cleaned by ultrasonic treatment in Milli-Q water. Approximately 12 mg of carbonate powder was drilled from the middle of the cubic subsamples which were prepared

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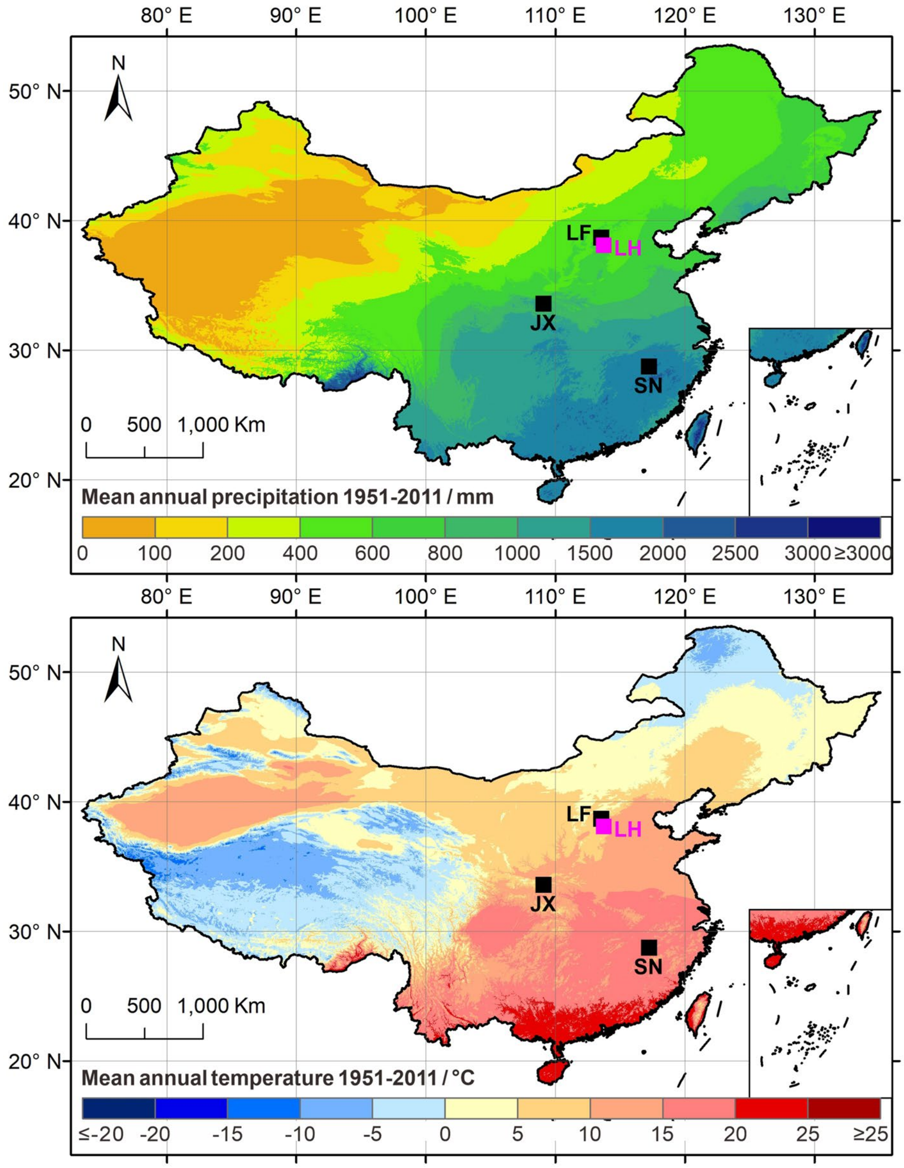

Figure 1. Locations of the study sites and climatological variables. The black squares show the locations of study sites: LF (Longfeng Cave) in Northern China, JX (Jiuxian Cave) in Central China and SN (Shennong Cave) in Southern China. The purple square shows the location of LH (Lianhua Cave) site (Dong et al., 2015). The upper and lower panels show precipitation and temperature, respectively. Both of them are plotted using the average annual precipitation and temperature data of 1951–2011 from Zhao et al. (2019).

for $^{14}C_{DOC}$ measurement. The carbonate powders were converted to CO_2 using 85% H_3PO_4 under vacuum at 70°C to expedite carbonate digestion, and then were purified.

The resulting CO_2 was reduced to graphite using Zn/Fe catalytic reduction by an automated procedure (Jull, 2013; Slota et al., 1987), and measured at the Xi'an AMS Center (Zhou et al., 2006). Results from the blank assessment and reliability of the method are described in Text S2 and shown in Figure S2 in Supporting Information S1.

3. Results

Twelve subsamples of both speleothem carbonate and DOC, paired to U-Th ages and spread throughout the Holocene, were ^{14}C-dated by AMS. ^{14}C ages were calibrated using CALIB Rev. 8.1.0 by Stuiver and Reimer (2020), based on the IntCal20 Northern Hemisphere radiocarbon age calibration curve (Reimer et al., 2020). The

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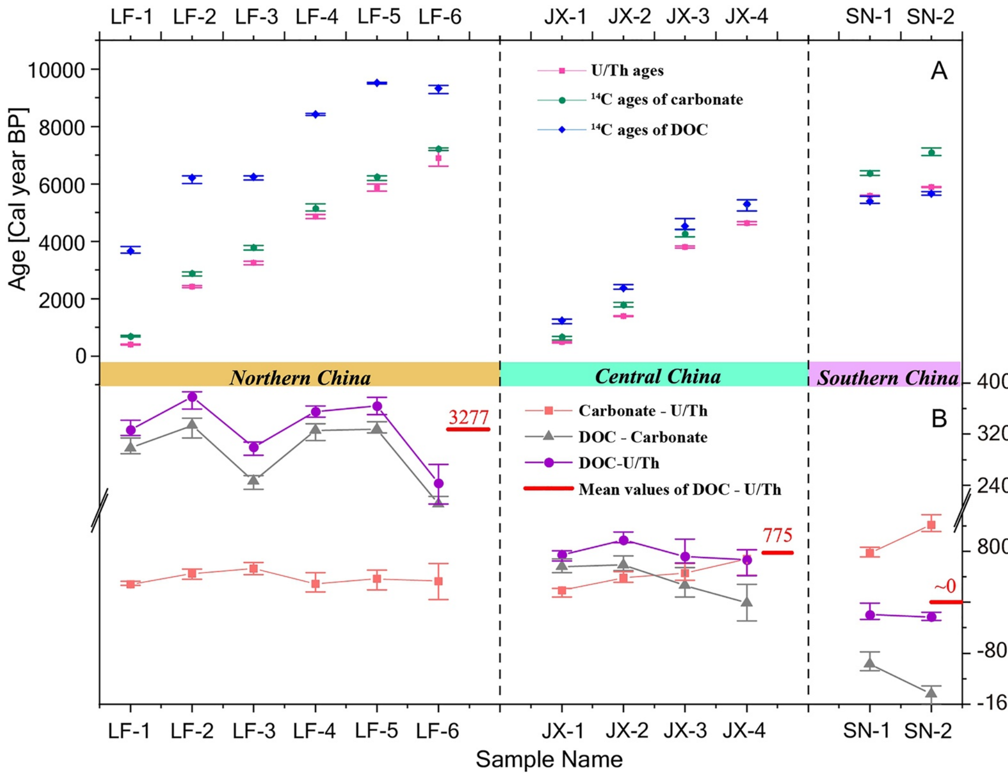

Figure 2. Comparison between ages and age differences between dating methods. (a) Comparison of speleothem $^{14}C_{DOC}$ ages (blue), U-Th ages (pink) and $^{14}C_{carb}$ ages (green). ^{14}C ages were calibrated by use of CALIB Rev. 8.1.0 by Stuiver and Reimer (2020) based on the IntCal20 (Reimer et al., 2020). (b) Comparison of the differences between $^{14}C_{carb}$ ages and U-Th ages (light pink), the differences between $^{14}C_{DOC}$ ages and U-Th ages (purple), and between $^{14}C_{DOC}$ ages and $^{14}C_{carb}$ ages (gray), the red solid lines and numbers are representative for the mean values of the differences between $^{14}C_{DOC}$ ages and U-Th ages. The errors of the age differences are calculated using propagated uncertainties, see Text S4 in Supporting Information S1.

uncertainties of calibrated ages are no more than 150 years (2σ), and most of them are just a few decades (Figure 2a and Table S2 in Supporting Information S1). The detailed ages and uncertainties are shown in Figure 2; Figure S1 and Table S2 in Supporting Information S1.

Most of the U-Th ages have dating errors of less than 50 years (2σ), and only one age has dating uncertainties larger than 250 years, that is, subsample of LF27 has an error of 279 years at U-Th age of 6886 yr. BP. Comparing the calibrated speleothem $^{14}C_{DOC}$ age with the corresponding U-Th age from the same deposition layer, the $^{14}C_{DOC}$ ages are between 2427 and 3784 years older for samples from Longfeng Cave (LF19, LF27 and LF32) and between 665 and 973 years older for samples from JX (C996-1), but nearly equivalent to or even younger (~180 years) for samples from Shennong Cave (SN27) (Figure 2b). The younger $^{14}C_{DOC}$ ages of SN-27 were likely caused by modern carbon contamination during the pre-treatment process. The blank assessment shows that the amounts of modern carbon are less than 4.5 μg and the maximum contamination may not exceed 2% for those subsamples of SN-27. Such contamination may cause the speleothem $^{14}C_{DOC}$ ages to be younger, but cannot exceed 200 years for the subsamples collected from stalagmite SN-27 (Text S2 in Supporting Information S1) which has the lowest amount of DOC in SN-27 in all our studied stalagmites (Figure S3A in Supporting Information S1). Therefore, we think these uncertainties have negligible impacts on the following discussions.

The $^{14}C_{carb}$ ages are also older than the corresponding U-Th ages, between 284 and 531 years for LF, 184–677 years for JX, and 776–1212 years for SN (Figure 2b and Table S2 in Supporting Information S1). Speleothem $^{14}C_{carb}$ ages are generally constrained by the "dead carbon proportion" (DCP) which can originate from bedrock dissolution and aged soil OM decomposition (Cheng et al., 2018; Griffiths et al., 2012; Lechleitner et al., 2016; Noronha et al., 2014). Our results show a variable contribution of DCP to the speleothem carbonate ^{14}C ages, ranging from 2.8% to 5.4% for LF, from 4.0% to 5.4% for JX and from 9.4% to 13.3% for SN (Table S3 in Supporting Information S1).

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4. Discussion

4.1. The Comparison of ^{14}C Ages of Speleothem Carbonate and DOC and ^{230}Th Ages

The results from Longfeng and Jiuxian Caves indicate that $^{14}C_{DOC}$ ages from these caves are not reflecting the true age of the speleothem and are therefore not suitable as an alternative to U-Th dating. Similar findings have been reported in several studies (Blyth et al., 2017; Genty et al., 2011; Lechleitner et al., 2019). It is likely that the process of flushing of soil OM from soil to groundwater causes a mix of soil OM of different ages, resulting in a dampened speleothem $^{14}C_{DOC}$ signal, and the presence of "old" groundwater may further prolong the lag time (McGarry & Baker, 2000). Previous studies tested using speleothem OM ^{14}C ages where U-Th dating fails, but robust comparison with reliable U-Th ages was not possible (Akers et al., 2019; Borsato et al., 2000). Our results suggest that, due to the complicated origin of speleothem OM, speleothem $^{14}C_{DOC}$ ages may introduce a considerable temporal lag into the extracted proxy information (e.g., oxygen isotopes) and its application as a dating tool needs to be carefully considered.

Older speleothem $^{14}C_{DOC}$ than U-Th ages indicates the presence of pre-aged DOC in these karst systems, potentially contributing to the DCP in speleothem carbonate when decomposed (Cai et al., 2005; Genty et al., 2001; Noronha et al., 2015). However, there is no consistent relationship between the age differences of $^{14}C_{DOC}$ and ^{230}Th, and the differences of $^{14}C_{carb}$ and ^{230}Th (Figure 2b), implying the decomposition of DOC does not dominate the carbonate DCP in our study sites. It is likely that the DCP in our stalagmites is dominated by the contribution from bedrock dissolution, and pre-aged DOC is only adding to that effect. However, the relative importance of each contribution to DCP is likely to be site specific and dependent on soil type/host rock, and regional climatic and environmental conditions, particularly precipitation and temperature (Griffiths et al., 2012; Lechleitner et al., 2016; Noronha et al., 2014; Rudzka et al., 2011).

4.2. Sources and Transport of Speleothem DOC

The OM in speleothems can originate from the surface soil and biosphere, the karst overlying the cave system, and from the in-cave environment (Blyth et al., 2016). Soil-derived OM can be transported in different forms (e.g., in dissolved, colloidal and particulate forms) into caves (Ban et al., 2008; Blyth et al., 2016; Hartland et al., 2012; Liao et al., 2018, 2021). Relative to colloidal and particulate organic substances, dissolved organic matter (DOM) is the component which is most likely to be leached into caves and entrapped in speleothems, especially its hydrophilic fractions (Blyth et al., 2016). The sources of speleothem DOM to date are mainly attributed to the overlying soil and karst system, based on the positive relationship between drip water DOC concentrations after rain events and the timing of speleothem fluorescence intensity (Baker & Genty, 1999; Baker et al., 1998; Ban et al., 2008; Liao et al., 2018; van Beynen et al., 2002).

Speleothem DOC is likely sourced from the drip water feeding it (Blyth et al., 2016; Hartland et al., 2012), and the concentration of DOC in drip water is strongly controlled by precipitation (Baker & Genty, 1999; Baker et al., 1998; Ban et al., 2008; Liao et al., 2018; Toth, 1998). This is because increased frequency and intensity of precipitation heightens the water head pressure, exceeding field capacity, resulting in soil DOC being leached downward into the unsaturated zone, and eventually transported into cave through drip water (Ban et al., 2008; Cruz Jr et al., 2005; Liao et al., 2018). In addition, the rewetting effect also can affect the concentration of soil DOC, in that rainfall after long periods of drought may increase the concentration of DOC in soil solution because the reduced decomposition rates in dry soils lead to the accumulation of microbial products (Kalbitz et al., 2000). Consequently, in monsoon regions, enhanced precipitation resuming after the non-monsoon season would increase the DOC concentration of exported percolating water in the karst system as revealed by karst monitoring studies (Liao et al., 2018). Although the response of drip water DOC concentrations to hydrological dynamics is sometimes lagged, this lag is typically of less than 1 year (Liao et al., 2018). Dampening effects in the downward percolation of soil DOC may principally be related to adsorption of DOC to mineral surfaces and subsequent accumulation in mineral soil horizons (Hemingway et al., 2019; Jardine et al., 1990; McDowell & Wood, 1984). Nevertheless, enhanced precipitation can also increase the velocity of pore water movement and decrease the contact time between soil solution and the solid matrix, leading to suppressed DOC adsorption in mineral soil horizons (Kalbitz et al., 2000).

Soil DOC generally originates from recent plant residues/litter and decomposition of stable soil OM (Guo et al., 2020; Michalzik et al., 2003). The latter gradually is treated as the main source for soil DOC (Gmach

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et al., 2020; Sanderman et al., 2008), because both laboratory and field experiments revealed that DOC in the soil is mainly derived from humified SOC (Currie et al., 1996). In particular, both $^{13}C/^{12}C$ and $^{14}C/^{12}C$ ratios of SOC and DOC at the same soil depths showed that fresh plant residues comprise only a small fraction of field-collected DOC due to the rapid decomposition of fresh DOC by microorganisms (Fröberg et al., 2006; Gregorich et al., 2000; Karltun et al., 2005; Sanderman et al., 2008). At the same time, the carbon isotopic compositions (both $^{13}C/^{12}C$ and $^{14}C/^{12}C$) of water-extractable OM in soils were closest to those of the average SOC rather than fresh materials (Gregorich et al., 2000; Sanderman et al., 2008). Thus, decomposition of SOC is treated as the most important DOC source, and it contributes more DOC than recently added litter does (Fröberg et al., 2003; Zsolnay, 1996). As mentioned above, speleothem DOC appears to be primarily derived from soil DOC, and therefore can be used as an indicator for SOC cycling.

4.3. The Spatio-Temporal Distribution of Speleothem $^{14}C_{DOC}$ Ages

Given that ^{14}C compositions of soil DOC represent those of average SOC, used to infer soil carbon turnover rates (Eglinton et al., 2021; Kaiser & Kalbitz, 2012; Shi et al., 2020; S. Trumbore, 2009), and considering the link between speleothem Organic carbon (OC) and DOC, variations in speleothem $^{14}C_{DOC}$ ages can be used to infer the changes in past soil turnover rates.

The systematic deviation of calibrated speleothem $^{14}C_{DOC}$ from the baseline U-Th ages at three cave sites in this study show latitudinal spatial variability. Mean deviations of speleothem $^{14}C_{DOC}$ ages, that is, 3277 (LF, northern China), 773 (JX, central China), and close to 0 years (SN, southern China) (Figure 2b and Table S4 in Supporting Information S1), are generally consistent with the present spatial distribution patterns of modern SOC ^{14}C ages in China and associated soil turnover times (Text S3 and Figure S4 in Supporting Information S1) (Carvalhais et al., 2014; Eglinton et al., 2021; Shi et al., 2020). Older (younger) speleothem $^{14}C_{DOC}$ ages correspond to aged (younger) ^{14}C ages of SOC and longer (shorter) turnover times, indicating that speleothem $^{14}C_{DOC}$ ages are closely related to the ^{14}C age of SOC and soil turnover times.

Mean deviations of speleothem $^{14}C_{DOC}$ ages also match local SOC ^{14}C ages in absolute terms (Shi et al., 2020) (Figure S4 in Supporting Information S1). Considering the aforementioned links between speleothem DOC and SOC, this implies that speleothem $^{14}C_{DOC}$ ages reflect SOC radiocarbon ages, and can be used to indicate the turnover time of SOC (Carvalhais et al., 2014). We then further inspect the correlation between the mean deviations in speleothem $^{14}C_{DOC}$ ages with climate parameters, since soil turnover time is primarily controlled by climate and environmental conditions (Carvalhais et al., 2014; Eglinton et al., 2021; Fan et al., 2020). The results show a negative relationship between speleothem $^{14}C_{DOC}$ ages and climate parameters (temperature and precipitation) on spatial scales (Table S4 in Supporting Information S1), consistent with the spatial pattern of soil carbon turnover time estimations based on the ratio of soil carbon stock to net primary productivity (NPP)/heterotrophic respiration flux and model estimate (Carvalhais et al., 2014; Eglinton et al., 2021; Varney et al., 2020).

On the temporal scale, the deviations of speleothem $^{14}C_{DOC}$ ages at LF varied between 2427 and 3784 years with a gradual increase over the long term. A similar long-term trend is apparent in the sample from JX, with deviations increasing from 649 years at ~4500 yr BP to 973 years at ~500 yr BP (Figure 3). Changes in age deviations at both cave sites roughly agree with the long-term trend in East Asian summer monsoon (EASM) intensity proxies over the Holocene (Cai et al., 2010; Dong et al., 2015) (Figure 3), further providing evidence on the linkage between speleothem $^{14}C_{DOC}$ ages and climate. Notably, a decoupling between the $^{14}C_{DOC}$ record and the climate reconstructions is apparent at ca. 3242 and 5868 yr BP at LF (Figure 3a), which may be related to factors other than climate. Environmental protection mechanisms affecting the retention of DOC are also thought to play a role in terrestrial DOC cycling (Gmach et al., 2020; Kalbitz et al., 2000). Hence, the effects of mechanisms such as the amount and type of soil colloids, soil depth, soil particle size, soil pH and ionic strength, cave bedrock thickness, and groundwater pathways (Jardine et al., 1990) on DOC in the vadose zone also need to be considered (Guo et al., 2020; Kalbitz et al., 2000), and may affect our data set. Meanwhile, the effects of other factors (e.g., upstream rainout, water recycling, moisture source change) on speleothem $\delta^{18}O$ also should be considered (Cai et al., 2021), as they might also lead to such mismatch.

Overall, speleothem $^{14}C_{DOC}$ ages at the three sites studied here replicate ^{14}C ages of SOC and appear to be controlled by soil carbon turnover which is primarily driven by climatic change (Carvalhais et al., 2014; Eglinton et al., 2021; Fan et al., 2020; Shi et al., 2020). It is clear that warm temperature and high precipitation led to higher

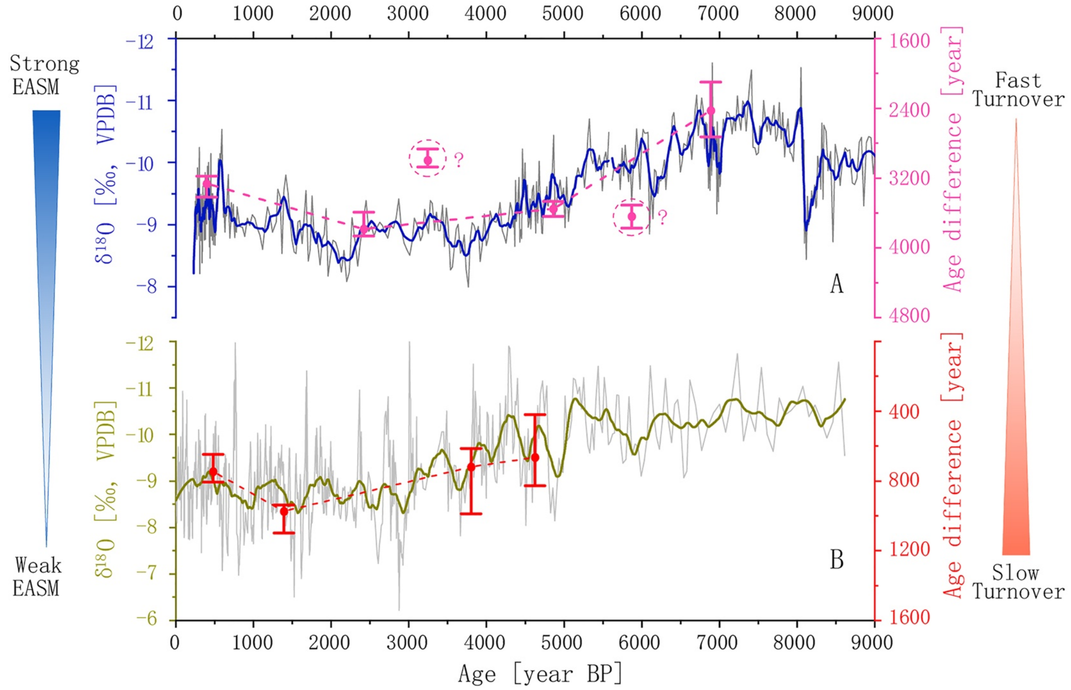

Figure 3. Comparison of age differences between U-Th and $^{14}C_{DOC}$ dating with (a) A speleothem $\delta^{18}O$ record from Lianhua cave (Dong et al., 2015) nearby Longfeng Cave (Wei et al., 2020). The thin gray curve is the original record, overlain by an 11-points smoothed average (blue). (b) A speleothem $\delta^{18}O$ record of C996-1 of Jiuxian cave, the gray curve is the original data and brown-yellow curve is the 500-years smoothed curve (Cai et al., 2010). The errors of age differences are calculated using propagated uncertainties, see Text S4 in Supporting Information S1.

NPP (i.e., SOC influx) and hence more fresh OM input to soil (Feng et al., 2007), and that warm and humid climate conditions also facilitate the outflux of SOC including pre-aged carbon via microbial respiration and vertical/lateral export (Carvalhais et al., 2014; Eglinton et al., 2021; Hein et al., 2020; Knorr et al., 2005). Jointly, these two processes lead to accelerated SOC turnover rates, resulting in younger SOC and speleothem $^{14}C_{DOC}$ ages (Figure S5 in Supporting Information S1). We expect the opposite to hold true for more arid and colder climate conditions, as indicated by the coherence between speleothem $^{14}C_{DOC}$ data and paleo-EASM records from the region (Figure 3). Although the dominant control variables on soil turnover times are diverse and vary spatially, precipitation and temperature appear to be dominating at our study sites, in agreement with previous studies (Carvalhais et al., 2014; Eglinton et al., 2021; Hein et al., 2020). Notably, the direct link between in-cave speleothem OC and the overlying soil still needs to be investigated and carefully considered, and more cave monitoring data (such as $^{14}C_{DOC}$ of drip water) and speleothem $^{14}C_{DOC}$ data are needed from different regions and climatic conditions to test the global nature of this relationship.

4.4. Implications for Understanding Soil Carbon Cycling in the EASM Region

Older speleothem $^{14}C_{DOC}$ ages compared to the corresponding U-Th ages have been attributed to the contribution and recycling of pre-aged organic carbon with long turnover times from the soil and karst system (Blyth et al., 2017). However, the extent to which pre-aged organic carbon affects speleothem $^{14}C_{DOC}$ and how this effect evolves remains poorly understood. Our results on the spatial scale present a significant latitudinal gradient, that is, longer SOC turnover times at higher latitudes (less precipitation and lower temperature), and vice versa, indicating that SOC turnover rates vary in different climatic zones (Figure 2 and Table S4 in Supporting Information S1). NPP and the heterotrophic respiration flux are important parameters that constrain soil carbon turnover time (Carvalhais et al., 2014; Eglinton et al., 2021). Satellite and model data show that both NPP and heterotrophic respiration flux are generally higher at lower latitudes in the Asian monsoon region, due to the overall more favorable climate conditions (Feng et al., 2007; Seino & Uchijima, 1992). Additionally, enhanced decomposition and export of soil carbon caused by higher temperature and precipitation could further facilitate

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the turnover of SOC, and our data shows a tendency for higher vertical export of DOC during periods of more rapid soil carbon turnover rates (Figure S6 in Supporting Information S1), likely accelerating the depletion of soil carbon stocks (Hein et al., 2020).

Over longer timescales, the similar trend between both mean deviations of speleothem $^{14}C_{DOC}$ ages and speleothem DOC concentrations (Figure S6 and Text S5 in Supporting Information S1), and EASM variations (Figure 3) indicate that changes in EASM strength may modulate SOC turnover times.

With the intensification of global warming, temperatures are rapidly increasing in regions with low mean annual temperatures (García-Palacios et al., 2021) at higher latitude areas where SOC turnover times are long. As non-labile SOC is more sensitive to temperature than labile SOC (Knorr et al., 2005), SOC with longer turnover times will become more vulnerable to decomposition, and turnover rates of SOC with lower decay rates will increase proportionally more than that of SOC with faster decay rates (Eglinton et al., 2021). As warmer and wetter climate conditions cause faster SOC turnover leading to enhanced release of CO_2 from soils (García-Palacios et al., 2021) through microbial respiration (Eglinton et al., 2021; Knorr et al., 2005) and export-induced oxidation of SOC (Hein et al., 2020), a positive feedback can be established, which may partially offset the existing soil carbon stock. Meanwhile, considering the consistency of the distribution of soil DOC concentrations between modern (Guo et al., 2020) and past (as reconstructed from speleothems in this study, Figure S3 in Supporting Information S1) on the spatial scale, it is likely that SOC distribution patterns did not shift substantially during the Holocene, providing the potential to extrapolate and validate model simulations of past changes in the terrestrial carbon cycle. Speleothems offer an intriguing archive to promote studies on the interactions between SOC turnover rates and climate changes on broad spatio-temporal scales. Given the novelty of our approach, further investigations in different regions and time periods are needed to better understand threshold values and boundary conditions for SOC cycling and the mechanisms governing the transfer of SOC to speleothems.

5. Conclusions

We present paired Holocene speleothem U-Th and ^{14}C ages of carbonate and DOC through three caves located on a north-south transect through China. The comparison of these ages suggests speleothem $^{14}C_{DOC}$ ages as a dating tool needs to be questioned. It is likely that the DCP in our stalagmites was dominated by the contribution from bedrock dissolution, the pre-aged DOC came second to that effect. Importantly, our findings implicate that climate dominates the speleothem $^{14}C_{DOC}$ ages and SOC turnover on spatio-temporal scales. As global warming likely will intensify, the accelerated turnover of SOC, particularly at higher latitude areas, may partially offset the existing soil carbon stock.

Data Availability Statement

All the additional information and data sets supporting the conclusions are available online and can be accessed from: http://paleodata.ieecas.cn/FrmDataInfo_EN.aspx?id=01f4bf72-a77b-43d7-a6e3-bd962c37c9c2.

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References

Akers, P. D., Brook, G. A., Railsback, L. B., Cherkinksy, A., Liang, F., Ebert, C. E., et al. (2019). Integrating U-Th, ^{14}C, and ^{210}Pb methods to produce a chronologically reliable isotope record for the Belize River Valley Maya from a low-uranium stalagmite. *The Holocene*, *297*(7), 1234–1248. https://doi.org/10.1177/0959683619838047

Bajo, P., Drysdale, R. N., Woodhead, J. D., Hellstrom, J. C., Hodell, D., Ferretti, P., et al. (2020). Persistent influence of obliquity on ice age terminations since the middle Pleistocene transition. *Science*, *367*(6483), 1235–1239. https://doi.org/10.1126/science.aaw1114

Baker, A., & Genty, D. (1999). Fluorescence wavelength and intensity variations of cave waters. *Journal of Hydrology*, *217*(1–2), 19–34. https://doi.org/10.1016/S0022-1694(99)00010-4

Baker, A., Genty, D., & Smart, P. L. (1998). High-resolution records of soil humification and paleoclimate change from variations in speleothem luminescence excitation and emission wavelengths. *Geology*, *26*(10), 903–906. https://doi.org/10.1130/0091-7613(1998)026<0903:HRROSH>2.3.CO;2

Ban, F., Pan, G., Zhu, J., Cai, B., & Tan, M. (2008). Temporal and spatial variations in the discharge and dissolved organic carbon of drip waters in Beijing Shihua Cave, China. *Hydrological Processes*, *22*(18), 3749–3758. https://doi.org/10.1002/hyp.6979

Blyth, A. J., Hartland, A., & Baker, A. (2016). Organic proxies in speleothems—New developments, advantages and limitations. *Quaternary Science Reviews*, *149*, 1–17. https://doi.org/10.1016/j.quascirev.2016.07.001

Blyth, A. J., Hua, Q., Smith, A., Frisia, S., Borsato, A., & Hellstrom, J. (2017). Exploring the dating of "dirty" speleothems and cave sinters using radiocarbon dating of preserved organic matter. *Quaternary Geochronology*, *39*, 92–98. https://doi.org/10.1016/j.quageo.2017.02.002

Borsato, A., Frisia, S., Jones, B., & Van Der Borg, K. (2000). Calcite moonmilk: Crystal morphology and environment of formation in caves in the Italian Alps. *Journal of Sedimentary Research*, *70*(5), 1171–1182. https://doi.org/10.1306/032300701171

Cai, Y., Cheng, X., Ma, L., Mao, R., Breitenbach, S. F., Zhang, H., et al. (2021). Holocene variability of East Asian summer monsoon as viewed from the speleothem $\delta^{18}O$ records in central China. *Earth and Planetary Science Letters*, *558*, 116758. https://doi.org/10.1016/j.epsl.2021.116758

Cai, Y., Tan, L., Cheng, H., An, Z., Edwards, R. L., Kelly, M. J., et al. (2010). The variation of summer monsoon precipitation in central China since the last deglaciation. *Earth and Planetary Science Letters*, *291*(1–4), 21–31. https://doi.org/10.1016/j.epsl.2009.12.039

Cai, Y., Warren, B., Peng, Z., & Zhang, Z. (2005). Effect of dead carbon on the ^{14}C dating of the speleothem. *Chinese Science Bulletin*, *50*(8), 817–821. https://doi.org/10.1360/04wd0082

Carvalhais, N., Forkel, M., Khomik, M., Bellarby, J., Jung, M., Migliavacca, M., et al. (2014). Global covariation of carbon turnover times with climate in terrestrial ecosystems. *Nature*, *514*(7521), 213–217. https://doi.org/10.1038/nature13731

Cheng, H., Edwards, R. L., Sinha, A., Spötl, C., Yi, L., Chen, S., et al. (2016). The Asian monsoon over the past 640, 000 years and ice age terminations. *Nature*, *534*(7609), 640–646. https://doi.org/10.1038/nature18591

Cheng, H., Edwards, R. L., Southon, J., Matsumoto, K., Feinberg, J. M., Sinha, A., et al. (2018). Atmospheric $^{14}C/^{12}C$ changes during the last glacial period from Hulu Cave. *Science*, *362*(6420), 1293–1297. https://doi.org/10.1126/science.aau0747

Conant, R. T., Ryan, M. G., Ågren, G. I., Birge, H. E., Davidson, E. A., Eliasson, P. E., et al. (2011). Temperature and soil organic matter decomposition rates—Synthesis of current knowledge and a way forward. *Global Change Biology*, *17*(11), 3392–3404. https://doi.org/10.1111/j.1365-2486.2011.02496.x

Cruz, F. W., Jr., Karmann, I., Magdaleno, G. B., Coichev, N., & Viana, O., Jr. (2005). Influence of hydrological and climatic parameters on spatial-temporal variability of fluorescence intensity and DOC of karst percolation waters in the Santana Cave System, Southeastern Brazil. *Journal of Hydrology*, *302*(1–4), 1–12. https://doi.org/10.1016/j.jhydrol.2004.06.012

Currie, W. S., Aber, J. D., McDowell, W. H., Boone, R. D., & Magill, A. H. (1996). Vertical transport of dissolved organic C and N under long-term N amendments in pine and hardwood forests. *Biogeochemistry*, *35*(3), 471–505. https://doi.org/10.1007/bf02183037

Dong, J., Shen, C. C., Kong, X., Wang, H. C., & Jiang, X. (2015). Reconciliation of hydroclimate sequences from the Chinese Loess Plateau and low-latitude East Asian Summer Monsoon regions over the past 14, 500 years. *Palaeogeography, Palaeoclimatology, Palaeoecology*, *435*, 127–135. https://doi.org/10.1016/j.palaeo.2015.06.013

Eglinton, T. I., Galy, V. V., Hemingway, J. D., Feng, X., Bao, H., Blattmann, T. M., et al. (2021). Climate control on terrestrial biospheric carbon turnover. *Proceedings of the National Academy of Sciences of the United States of America*, *118*(8), e2011585118. https://doi.org/10.1073/pnas.2011585118

Fan, N., Koirala, S., Reichstein, M., Thurner, M., Avitabile, V., Santoro, M., et al. (2020). Apparent ecosystem carbon turnover time: Uncertainties and robust features. *Earth System Science Data Discussions*, *2020*, 1–25. https://doi.org/10.5194/essd-2019-235

Feng, X., Liu, G., Chen, J. M., Chen, M., Liu, J., Ju, W. M., et al. (2007). Net primary productivity of China's terrestrial ecosystems from a process model driven by remote sensing. *Journal of Environmental Management*, *85*(3), 563–573. https://doi.org/10.1016/j.jenvman.2006.09.021

Friedlingstein, P., Cox, P. P., Betts, R., Bopp, L., von Bloh, W., Brovkin, V., et al. (2006). Climate-carbon cycle feedback analysis: Results from the (CMIP)-M-4 model intercomparison. *Journal of Climate*, *19*(14), 3337–3353. https://doi.org/10.1175/JCLI3800.1

Fröberg, M., Berggren, D., Bergkvist, B., Bryant, C., & Knicker, H. (2003). Contributions of Oi, Oe and Oa horizons to dissolved organic matter in forest floor leachates. *Geoderma*, *113*(3–4), 311–322. https://doi.org/10.1016/S0016-7061(02)00367-1

Fröberg, M., Berggren, D., Bergkvist, B., Bryant, C., & Mulder, J. (2006). Concentration and fluxes of dissolved organic carbon (DOC) in three Norway spruce stands along a climatic gradient in Sweden. *Biogeochemistry*, *77*, 1–23. https://doi.org/10.1007/s10533-004-0564-5

García-Palacios, P., Crowther, T. W., Dacal, M., Hartley, I. P., Reinsch, S., Rinnan, R., et al. (2021). Evidence for large microbial-mediated losses of soil carbon under anthropogenic warming. *Nature Reviews Earth & Environment*, *2*(7), 507–517. https://doi.org/10.1038/s43017-021-00178-4

Genty, D., Baker, A., Massault, M., Proctor, C., Gilmour, M., Pons-Branchu, E., & Hamelin, B. (2001). Dead carbon in stalagmites: Carbonate bedrock paleodissolution vs. ageing of soil organic matter. Implications for ^{13}C variations in speleothems. *Geochimica et Cosmochimica Acta*, *65*(20), 3443–3457. https://doi.org/10.1016/S0016-7037(01)00697-4

Genty, D., Genty, D., Konik, S., Valladas, H., Blamart, D., Hellstrom, J., et al. (2011). Dating the Lascaux cave gour formation. *Radiocarbon*, *53*(3), 479–500. https://doi.org/10.1017/S0033822200034603

Gmach, M. R., Cherubin, M. R., Kaiser, K., & Cerri, C. E. P. (2020). Processes that influence dissolved organic matter in the soil: A review. *Scientia Agricola*, *77*(3), e20180164. https://doi.org/10.1590/1678-992X-2018-0164

Gregorich, E. G., Liang, B. C., Drury, C. F., Mackenzie, A. F., & McGill, W. B. (2000). Elucidation of the source and turnover of water soluble and microbial biomass carbon in agricultural soils. *Soil Biology and Biochemistry*, *32*(5), 581–587. https://doi.org/10.1016/S0038-0717(99)00146-7

Griffiths, M. L., Fohlmeister, J., Drysdale, R. N., Hua, Q., Johnson, K. R., Hellstrom, J. C., et al. (2012). Hydrological control of the dead carbon fraction in a Holocene tropical speleothem. *Quaternary Geochronology*, *14*, 81–93. https://doi.org/10.1016/j.quageo.2012.04.001

Guo, Z., Wang, Y., Wan, Z., Zuo, Y., He, L., Li, D., et al. (2020). Soil dissolved organic carbon in terrestrial ecosystems: Global budget, spatial distribution and controls. *Global Ecology and Biogeography*, *29*(12), 2159–2175. https://doi.org/10.1111/geb.13186

Hartland, A., Fairchild, I. J., Lead, J. R., Borsato, A., Baker, A., Frisia, S., & Baalousha, M. (2012). From soil to cave: Transport of trace metals by natural organic matter in karst dripwaters. *Chemical Geology*, *304*, 68–82. https://doi.org/10.1016/j.chemgeo.2012.01.032

Hein, C. J., Usman, M., Eglinton, T. I., Haghipour, N., & Galy, V. V. (2020). Millennial-scale hydroclimate control of tropical soil carbon storage. *Nature*, *581*(7806), 63–66. https://doi.org/10.1038/s41586-020-2233-9

Hemingway, J. D., Rothman, D. H., Grant, K. E., Rosengard, S. Z., Eglinton, T. I., Derry, L. A., & Galy, V. V. (2019). Mineral protection regulates long-term global preservation of natural organic carbon. *Nature*, *570*(7760), 228–231. https://doi.org/10.1038/s41586-019-1280-6

Jardine, P. M., Wilson, G. V., McCarthy, J. F., Luxmoore, R. J., Taylor, D. L., & Zelany, L. W. (1990). Hydrogeochemical processes controlling the transport of dissolved organic carbon through a forested hill-slope. *Journal of Contaminant Hydrology*, *6*(1), 3–19. https://doi.org/10.1016/0169-7722(90)90008-5

Jull, A. J. T. (2013). AMS radiocarbon dating. In S. A. Elias (Ed.), *Encyclopedia of quaternary science* (2nd ed., pp. 316–323). Elsevier.

Kaiser, K., & Kalbitz, K. (2012). Cycling downwards—Dissolved organic matter in soils. *Soil Biology and Biochemistry*, *52*, 29–32. https://doi.org/10.1016/j.soilbio.2012.04.002

Kalbitz, K., Solinger, S., Park, J. H., Michalzik, B., & Matzner, E. (2000). Controls on the dynamics of dissolved organic matter in soils: A review. *Soil Science*, *165*(4), 277–304. https://doi.org/10.1097/00010694-200004000-00001

Karltun, E., Harrison, A. F., Alriksson, A., Bryant, C., Garnett, M. H., & Olsson, M. T. (2005). Old organic carbon in soil solution DOC after afforestation—Evidence from C-14 analysis. *Geoderma*, *127*(3–4), 188–195. https://doi.org/10.1016/j.geoderma.2004.12.008

Knorr, W., Prentice, I. C., House, J. I., & Holland, E. A. (2005). Long-term sensitivity of soil carbon turnover to warming. *Nature*, *433*(7023), 298–301. https://doi.org/10.1038/nature03226

19448007, 2023, 3, Downloaded from https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2022GL101875 by Universität Bern, Wiley Online Library on [17/02/2023]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

Lang, S. Q., Bernasconi, S. M., & Früh-Green, G. L. (2012). Stable isotope analysis of organic carbon in small (μg C) samples and dissolved organic matter using a GasBench preparation device. *Rapid Communications in Mass Spectrometry*, *26*(1), 9–16. https://doi.org/10.1002/rcm.5287

Lechleitner, F. A., Baldini, J. U. L., Breitenbach, S. F. M., Fohlmeister, J., McIntyre, C., Goswami, B., et al. (2016). Hydrological and climatological controls on radiocarbon concentrations in a tropical stalagmite. *Geochimica et Cosmochimica Acta*, *194*, 233–252. https://doi.org/10.1016/j.gca.2016.08.039

Lechleitner, F. A., Lang, S., Haghipour, N., McIntyre, C., Baldini, J., Prufer, K., & Eglinton, T. I. (2019). Towards organic carbon isotope records from stalagmites: Coupled $\delta^{13}C$ and ^{14}C analysis using wet chemical oxidation. *Radiocarbon*, *61*(03), 749–764. https://doi.org/10.1017/RDC.2019.35

Liao, J., Hu, C., Li, X., & Ruan, J. (2021). Drying increases organic colloidal mobilization in the karst vadose zone: Evidence from a 15-year cave-monitoring study. *Hydrological Processes*, *35*(4), e14163. https://doi.org/10.1002/hyp.14163

Liao, J., Hu, C., Wang, M., Li, X., Ruan, J., Zhu, Y., et al. (2018). Assessing acid rain and climate effects on the temporal variation of dissolved organic matter in the unsaturated zone of a karstic system from southern China. *Journal of Hydrology*, *556*, 475–487. https://doi.org/10.1016/j.jhydrol.2017.11.043

McDowell, W. H., & Wood, T. (1984). Podzolization: Soil processes control dissolved organic carbon concentrations in stream water. *Soil Science*, *137*(1), 23–32. https://doi.org/10.1097/00010694-198401000-00004

McGarry, S. F., & Baker, A. (2000). Organic acid fluorescence: Applications to speleothem palaeoclimate reconstruction. *Quaternary Science Reviews*, *19*(11), 1087–1101. https://doi.org/10.1016/S0277-3791(99)00087-6

Michalzik, B., Tipping, E., Mulder, J., Gallardo Lancho, J. F., Matzner, E., Bryant, C., et al. (2003). Modelling the production and transport of dissolved organic carbon in forest soils. *Biogeochemistry*, *66*(3), 241–264. https://doi.org/10.1023/B:BIOG.0000005329.68861.27

Noronha, A. L., Johnson, K. R., Hu, C., Ruan, J., Southon, J. R., & Ferguson, J. E. (2014). Assessing influences on speleothem dead carbon variability over the Holocene: Implications for speleothem-based radiocarbon calibration. *Earth and Planetary Science Letters*, *394*, 20–29. https://doi.org/10.1016/j.epsl.2014.03.015

Noronha, A. L., Johnson, K. R., Southon, J. R., Hu, C., Ruan, J., & McCabe-Glynn, S. (2015). Radiocarbon evidence for decomposition of aged organic matter in the vadose zone as the main source of speleothem carbon. *Quaternary Science Reviews*, *127*, 37–47. https://doi.org/10.1016/j.quascirev.2015.05.021

Reimer, P. J., Austin, W. E. N., Bard, E., Bayliss, A., Blackwell, P. G., Bronk Ramsey, C., et al. (2020). The IntCal20 Northern Hemisphere radiocarbon age calibration curve (0-55 cal kBP). *Radiocarbon*, *62*(4), 725–757. https://doi.org/10.1017/rdc.2020.41

Rudzka, D., McDermott, F., Baldini, L. M., Fleitmann, D., Moreno, A., & Stoll, H. (2011). The coupled $\delta^{13}C$-radiocarbon systematics of three Late Glacial/early Holocene speleothems; insights into soil and cave processes at climatic transitions. *Geochimica et Cosmochimica Acta*, *75*(15), 4321–4339. https://doi.org/10.1016/j.gca.2011.05.022

Sanderman, J., Baldock, J. A., & Amundson, R. (2008). Dissolved organic carbon chemistry and dynamics in contrasting forest and grassland soils. *Biogeochemistry*, *89*(2), 181–198. https://doi.org/10.1007/s10533-008-9211-x

Schimel, D. S., Braswell, R., Holland, E. A., McKeown, R., Ojima, D. S., Painter, T. H., et al. (1994). Climatic, edaphic, and biotic controls over storage and turnover of carbon in soils. *Global Biogeochemistry Cycles*, *8*(3), 279–293. https://doi.org/10.1029/94GB00993

Seino, H., & Uchijima, Z. (1992). Global distribution of net primary productivity of terrestrial vegetation. *Journal of Agricultural Meteorology*, *48*(1), 39–48. https://doi.org/10.2480/agrmet.48.39

Shi, Z., Allison, S. D., He, Y., Levine, P. A., Hoyt, A. M., Beem-Miller, J., et al. (2020). The age distribution of global soil carbon inferred from radiocarbon measurements. *Nature Geoscience*, *13*(8), 555–559. https://doi.org/10.1038/s41561-020-0596-z

Slota, P. J. J., Jull, A. J. T., Linick, T. W., & Toolin, L. J. (1987). Preparation of small samples for ^{14}C accelerator targets by catalytic reduction of CO. *Radiocarbon*, *29*(2), 303–306. https://doi.org/10.1017/S0033822200056988

Stuiver, M., & Reimer, P. (2020). CALIB Rev. 8.1.0. Retrieved from http://calib.org/calib/

Toth, V. A. (1998). Spatial and temporal variations in the dissolved organic carbon concentrations in the vadose karst waters of Marengo Cave, Indiana. *Journal of Cave and Karst Studies*, *60*, 167–171.

Trumbore, S. (2009). Radiocarbon and soil carbon dynamics. *Annual Review of Earth and Planetary Sciences*, *37*(1), 47–66. https://doi.org/10.1146/annurev.earth.36.031207.124300

Trumbore, S. E., Chadwick, O. A., & Amundson, R. (1996). Rapid exchange between soil carbon and atmospheric carbon dioxide driven by temperature change. *Science*, *272*(5260), 393–396. https://doi.org/10.1126/science.272.5260.393

van Beynen, P. E., Schwarcz, H. P., Ford, D. C., & Timmins, G. T. (2002). Organic substances in cave drip waters: Studies from Marengo cave, Indiana. *Canadian Journal of Earth Science*, *39*, 279–284. https://doi.org/10.1139/e01-072

Varney, R. M., Chadburn, S. E., Friedlingstein, P., Burke, E. J., Koven, C. D., Hugelius, G., & Cox, P. M. (2020). A spatial emergent constraint on the sensitivity of soil carbon turnover to global warming. *Nature Communications*, *11*(1), 1–8. https://doi.org/10.1038/s41467-020-19208-8

Wang, C., Houlton, B. Z., Liu, D., Hou, J., Cheng, W., & Bai, E. (2018). Stable isotopic constraints on global soil organic carbon turnover. *Biogeosciences*, *15*(4), 987–995. https://doi.org/10.5194/bg-15-987-2018

Wang, C., Qu, L., Yang, L., Liu, D., Morrissey, E., Miao, R., et al. (2021). Large-scale importance of microbial carbon use efficiency and necromass to soil organic carbon. *Global Change Biology*, *27*(10), 2039–2048. https://doi.org/10.1111/gcb.15550

Wei, Y., Cai, Y., Cheng, X., Xue, G., Lu, Y., Ma, L., et al. (2020). Holocene and deglaciation hydroclimate changes in northern China as inferred from stalagmite growth frequency. *Global and Planetary Change*, *195*, 103360. https://doi.org/10.1016/j.gloplacha.2020.103360

Zhang, H., Cai, Y., Tan, L., Cheng, H., Qin, S., An, Z., et al. (2015). Large variations of $\delta^{13}C$ values in stalagmites from southeastern China during historical times: Implications for anthropogenic deforestation. *Boreas*, *44*(3), 511–525. https://doi.org/10.1111/bor.12112

Zhao, H., Huang, W., Xie, T., Wu, X., Xie, Y., Feng, S., & Chen, F. (2019). Optimization and evaluation of a monthly air temperature and precipitation gridded dataset with a 0.025 spatial resolution in China during 1951–2011. *Theoretical and Applied Climatology*, *138*(1), 491–507. https://doi.org/10.1007/s00704-019-02830-y

Zhou, W. J., Zhao, X., Lu, X., Liu, L., Wu, Z., Cheng, P., et al. (2006). The 3MV multi-element AMS in Xian, China: Unique features and preliminary tests. *Radiocarbon*, *48*(2), 285–293. https://doi.org/10.1017/S0033822200066492

Zsolnay, A. (1996). In A. Piccolo (Ed.) *Dissolved humus in soil waters in Humic substances in terrestrial ecosystems*. (Vol. 171–223). Elsevier.

19448007, 2023, 3, Downloaded from https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2022GL101875 by Universität Bern, Wiley Online Library on [17/02/2023]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

10.1029/2022GL101875

References From the Supporting Information

Lechleitner, F. (2021). *flechleitner/DCF_calculator: (v1.0)*. Zenodo. [code]. https://doi.org/10.5281/zenodo.5503025

Wang, Y., Zhou, W., & Cheng, P. (2020). Sources analysis of dissolved organic carbon in water using carbon isotope method. *Journal of Earth Environment*, *11*, 435–446. (Chinese edition with English abstract).

Zhang, H., Zhang, X., Cai, Y., Sinha, A., Spötl, C., Baker, J., et al. (2021). A data-model comparison pinpoints Holocene spatiotemporal pattern of East Asian summer monsoon. *Quaternary Science Reviews*, *261*, 106911. https://doi.org/10.1016/j.quascirev.2021.106911

Zhao, X., Ma, C., & Xiao, L. (2014). The vegetation history of Qinling Mountains, China. *Quaternary International*, *325*, 55–62. https://doi.org/10.1016/j.quaint.2013.10.054

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