



Lake geochemistry reveals marked environmental change in Southwest China during the Mid Miocene Climatic Optimum

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Abstract The Mid-Miocene Climatic Optimum (MMCO; ~15–17 Ma) was one of the short-term climatic warm events that punctuated the Cenozoic long-term cooling trend. Because there are very few terrestrial records of this event, most of our understanding comes from marine cores. In this report, we first present new palaeomagnetic data that revises the dating of our 400 m-thick lacustrine section in Wenshan (Yunnan), previously thought to be Late Miocene. These new data suggest an older age, ca. 15.2–16.5 Ma, coinciding with the MMCO. We measured $\delta^{13}\text{C}$ on bulk organic matter ($\delta^{13}\text{C}_{\text{org}}$), total organic carbon (TOC), total nitrogen (TN) and C/N ratios at a high sample resolution to: (1) reconstruct the palaeoenvironmental changes in the lake catchment area, and (2) infer

mechanisms responsible for these changes. Our results show that all four geochemical parameters demonstrate that a strong environmental change occurred around the middle of the section, shortly after the C5Cn/C5Br geomagnetic reversal and the Early/Middle Miocene boundary at 15.97 Ma. We propose that the environmental shift may be due to a combination of a change in climate, which became cooler, together with a change in organic matter cycling within the lake. This study provides a new insight into the MMCO and demonstrates that although the MMCO was generally a warm event, it was also a time of climatic instability and abrupt environmental changes.

Keywords Yunnan · Mid-Miocene Climatic Optimum · Bulk organic carbon isotopes ($\delta^{13}\text{C}_{\text{org}}$) · C/N ratio · TOC

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1 Introduction

Marine isotopic data have revealed that the long-term cooling trend of the Cenozoic was punctuated by short-term climatic events [1]. The Miocene climate exhibited marked changes including the Mid-Miocene Climatic Optimum (MMCO, ~15–17.0 Ma [2]) during which temperatures were inferred to be ~3–8 °C higher than pre-industrial level [3, 4]. The mechanisms leading to the MMCO are still debated [2], but evidence points to an increase in the concentration of atmospheric CO₂ [4].

Until now, the MMCO has been mainly investigated using marine isotopic data [1, 2, 5, 6], since terrestrial outcrops are relatively scarce. In Antarctica, a palynological study and leaf wax geochemistry revealed a warmer and wetter climate than today, and demonstrated that Antarctica could support a tundra vegetation [7, 8]. In China, a lithological study conducted in the northeast Tibetan Plateau and a palynological investigation in Tian Shan (northwest China) show evidence of a warm and wet period during the MMCO [9, 10], demonstrating that the lower latitude continental areas were also affected by the MMCO. However, it is still unclear how subtropical environments responded to the MMCO due to the scarcity of suitably dated outcrops.

In Yunnan (southwest China), the evolution of the Miocene climate has been extensively investigated using different floral proxies, such as pollen [11, 12] and plant mega-fossils [13–17], indicating that Yunnan was already under a monsoonal climate in the Miocene [11, 14]. We have previously reported palynological results from a 400 m-thick sedimentary succession in Wenshan, southeast Yunnan, to reveal how the vegetation dynamics responded to a monsoonal climate [11]. Lakes are environmentally sensitive and so are ideal to study past climatic fluctuations [18]. For example, lake sediment geochemistry has been widely used to explore environmental changes on the Tibetan Plateau during the Miocene [19, 20] and in Yunnan during the Quaternary [21–23]. However, there is very limited research using terrestrial geochemistry to investigate environmental changes in Yunnan during the Miocene, and the MMCO in particular.

In the present study, we present new magnetostratigraphic data to date the sedimentary succession. Then, we employ lake sediment organic geochemistry with high sampling density to: (1) further investigate palaeoenvironmental changes in the lake catchment area in Wenshan using geochemical proxies ($\delta^{13}\text{C}$ on bulk organic matter, total organic carbon, total nitrogen, and C/N ratio); and (2) discuss the factors responsible for these palaeoenvironmental changes.

2 Materials and methods

2.1 Geological setting

Our study section of the Wenshan palaeo-lake is located on the South China Block, close to the Ailao Shan-Red River Fault, which underwent a mid-Tertiary (~35–17 Ma ago) sinistral shear displacement before being reactivated as a right lateral system during the Pliocene [24]. The time when this fault was active is still debated, but evidence shows that there was movement during the Miocene and this may have influenced the topography of Wenshan at the time of sediment accumulation [25, 26]. A recent study showed that Xiaolongtan Basin, located about 110 km east of the Wenshan Basin, had already attained its near modern elevation by ~13 Ma [27].

Today, Wenshan experiences a monsoonal climate with a mean annual temperature (MAT) of 18.2 °C and a mean annual precipitation (MAP) of 1059 mm, with most precipitation occurring during the summer [28]. On limestone, the vegetation is composed of a semi-humid broadleaved evergreen forest, while on acidic soil the vegetation is mainly classified as subtropical monsoon broadleaved evergreen forest [29].

The sampling site (23°24' N; 104°12' E, 1270 m a.s.l.) of the Wenshan palaeo-lake is situated near Dayigu village in the Wenshan Basin, southeast Yunnan, China (Fig. 1). The site is surrounded by mountains that are mainly composed of limestone and mudstone, with ages ranging from the Cambrian to the Triassic [30] (Fig. 1). The palaeo-lake sediments are about 400 m thick and unconformably overlie Paleogene breccia. In the lower third of the succession, Quaternary conglomerates cover approximately 100 m of the section, dividing the succession into two parts: the lower part (~90 m thick) and the upper part (~300 m thick) (Fig. 2a). There is, however, no obvious sedimentological difference between the two parts. The Miocene sediments are composed of thin to medium (<30 cm) cycles of sandstone, siltstone, and calcareous mudstone. The transition between each cycle is always gradual, suggesting nearly continuous deposition. The sediments are often unconsolidated, ranging in colour from dark grey to creamy yellow, and contain numerous fragments of organic matter (Fig. 2c). The sediments were deposited in thin to very thin parallel beds and thick laminae (Fig. 2b, e). The depositional environment is interpreted to be lacustrine [31] and the presence of a thick succession of laminae suggests that the lake was relatively deep, while the absence of bioturbation reveals that the bottom of the lake might have been suboxic to anoxic, promoting organic matter preservation (Fig. 2c). We

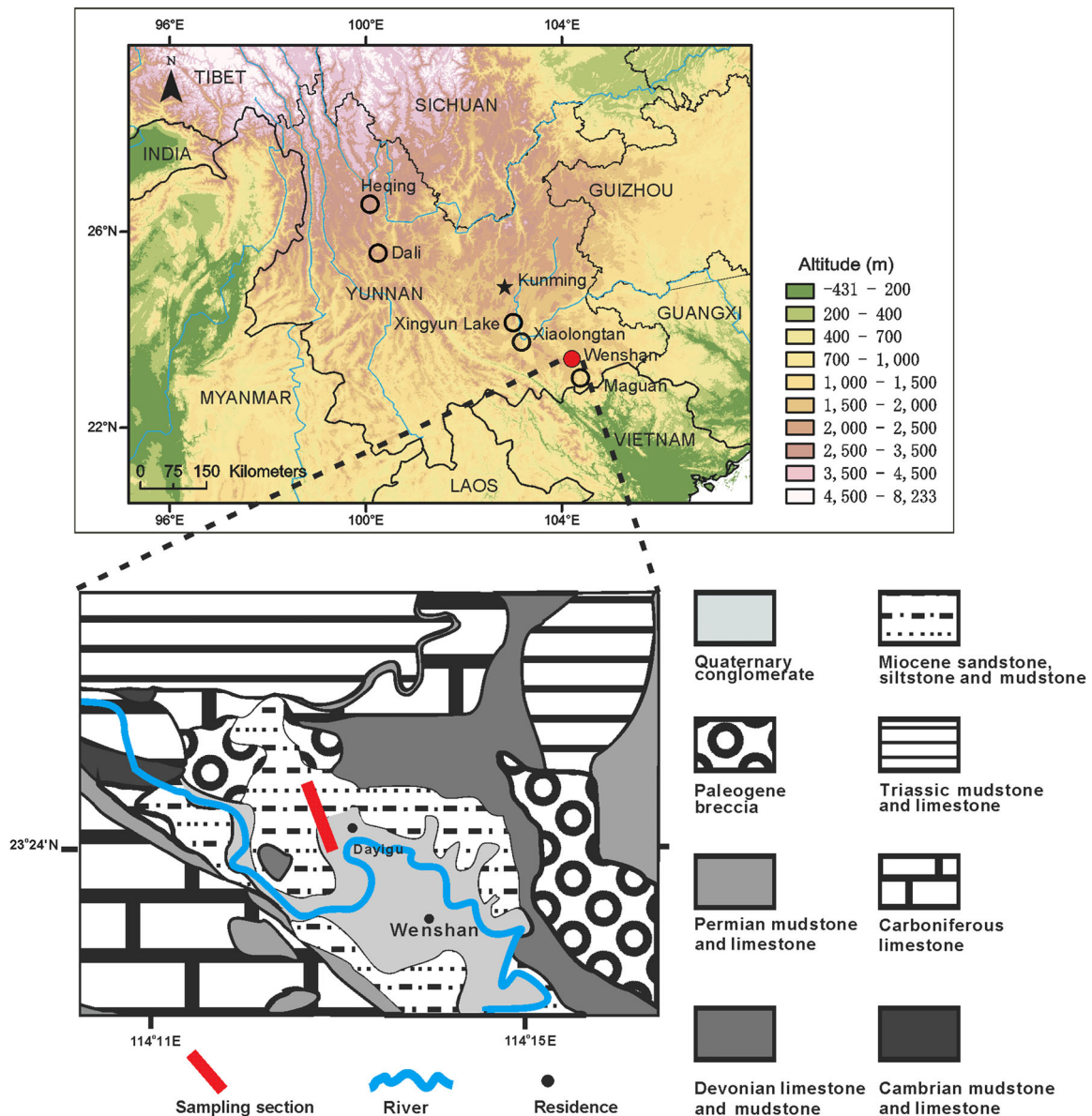


Fig. 1 Location of Wenshan (filled circle) and other places in Yunnan discussed in this study (open circle), and geological map of the study area, adapted from Li et al. [11]

observed six thin beds (~ 10 cm) of rounded clast-supported conglomerates and matrix-supported conglomerates which could represent brief episodes of fluvial activity or distant landslides (Fig. 2a).

2.2 Magnetostratigraphic samples and method

To accurately constrain the age of the Wenshan sediments, we conducted a high-resolution magnetostratigraphy study on a 270 m section in the Wenshan Basin. However, a ~ 50 m interval in the middle part is covered due to house building. A total of 236 of palaeomagnetic samples were

collected using a gasoline-powered drill at intervals of ~ 1 m.

Systematic rock magnetic experiments were conducted on representative samples in order to determine the remanence carrier in the sediments (see supporting online material and Figs. S1, S2 and S3 for detail). Remanence was measured with a 2G Enterprises Model 760-R cryogenic magnetometer situated in a magnetically shielded room (<300 nT). Samples were subjected to progressive alternating field (AF) demagnetization in a maximum field of 60–70 mT with 5–10 mT intervals. All the palaeomagnetic measurements were carried out at the

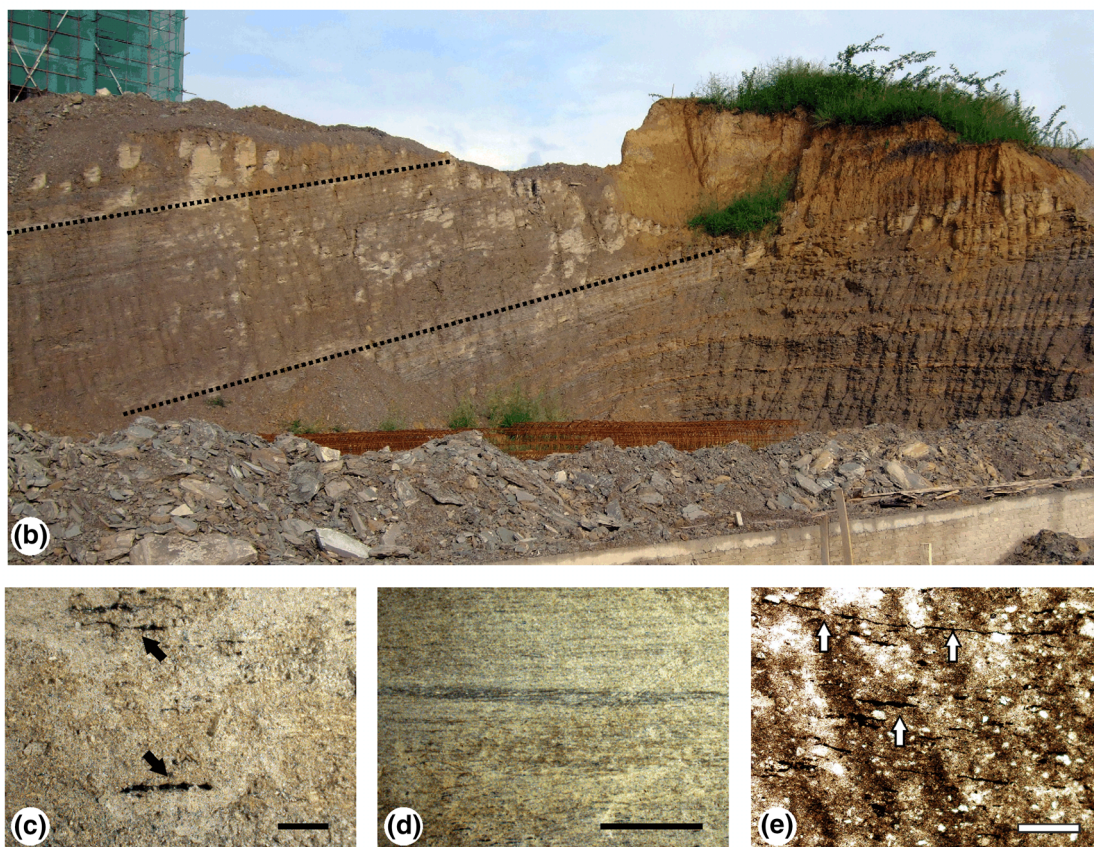
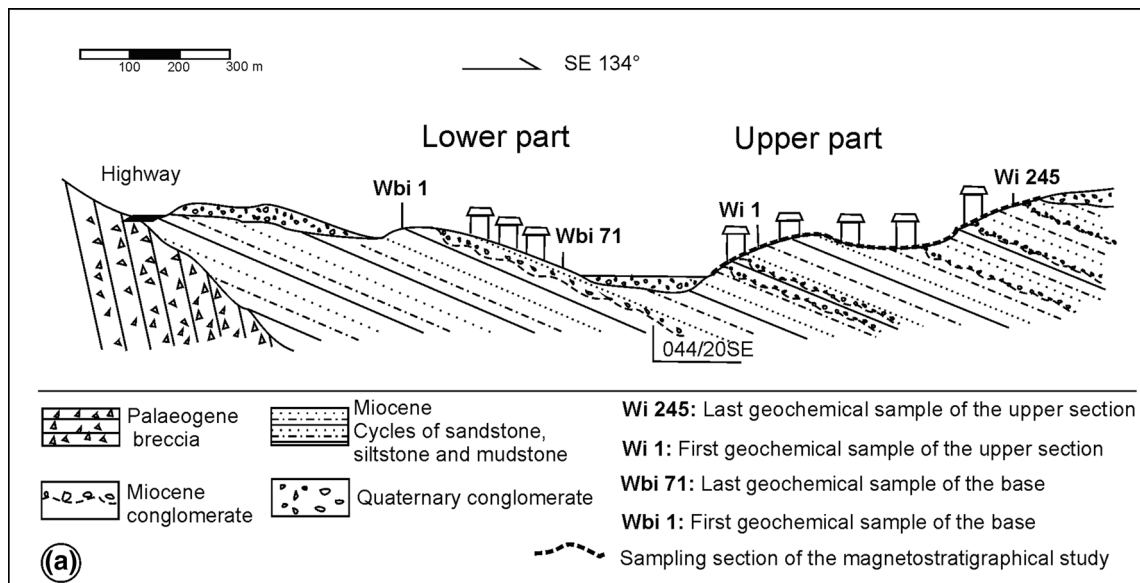


Fig. 2 Sediments in Wenshan **a** Cross section of the study area, **b** Base of the section displaying parallel beds outlined by the *dashed lines*. The width of view is about 15 m. **c** Binocular microscope (Leica SAPO) view of the sample number 55. *Arrows* point to organic matter. **d** Binocular microscope (Leica SAPO) view of sample number 42 exhibiting horizontal laminae. **e** Microscope (Olympus Bx51) view of thin section of sample number 41 under plane-polarized light revealing organic matter deposited in sub-parallel laminae (*arrows*). Scale bars = 1 mm in (c, d), and 250 μ m in (e)

Palaeomagnetism and Geochronology Laboratory of the Institute of Geology and Geophysics, Chinese Academy of Sciences in Beijing. Demagnetization results were

evaluated by orthogonal diagrams [32] and the principal components direction was computed using a “least-squares fitting” technique [33].

2.3 Geochemical samples and methods

Three hundred and seventeen geochemical samples were collected from shale and mudstone on freshly exposed outcrop with an interval of 50 cm between most samples. Sandstone was avoided as much as possible because it usually contains very little organic matter.

We analysed bulk organic carbon isotopes ($\delta^{13}\text{C}_{\text{org}}$), total organic carbon (TOC), total nitrogen (TN), and C/N (atomic) ratio to explore environmental changes within the palaeo-lake. Before measuring the $\delta^{13}\text{C}_{\text{org}}$ and TOC values, we followed the procedure described in Leng et al. [34] to remove carbonates from the samples by immersing them in 10 % HCl until complete degassing of CO_2 . The values of $\delta^{13}\text{C}_{\text{org}}$ and TOC were measured in the Isotope Laboratory of GeoZentrum Nordbayern, Friedrich Alexander Universität Erlangen Nürnberg, using a CE 1110 Elemental analyser connected online to a ThermoFisher Delta V plus mass spectrometer. Isotope values are reported using the conventional δ -notation in per mil (‰) relative to V-PDB. TOC is the percentage of the weight of total organic carbon over the weight of total dry sediments after acid digestion. The analytical precision based on replicate analyses of lab standards, calibrated against the international standards USGS 24 and USGS 40, was better than ± 0.07 ‰ for $\delta^{13}\text{C}_{\text{org}}$ and 0.2 % for TOC (1 σ).

The TN values were quantified in the Central Laboratory of the Tropical Forest Ecology of Xishuangbanna Tropical Botanical Garden using a Vario MAX CN and a Vario PYRO cube from Elementar Analysensystem GmbH (Germany). The C/N (atomic) ratio is calculated using the method described in Meyers and Teranes [35]:

$$\text{C/N} = \frac{\text{TOC}(\%) / 12.01}{\text{TN}(\%) / 14.01},$$

where 12.01 is the atomic weight of carbon and 14.01 is the atomic weight of nitrogen. When calculating the C/N ratio both TOC and TN are expressed in percentages, but in the rest of the study the TN values are reported in per mil (‰) because the TN values are low. We used total nitrogen in bulk sediments, because in most inorganic sediments the concentration of inorganic nitrogen is small compared to organic nitrogen due to the high TOC values [36].

The palaeomagnetic samples and geochemical samples were correlated to the same sedimentary layers.

3 Results

3.1 Chronology construction

The AF demagnetization method used in this study was capable of isolating the characteristic remanent

magnetization (ChRM) after removal of soft secondary components of magnetization. Representative demagnetization diagrams are shown in Fig. 3. For most samples, the high-stability ChRM component was separated between 30 and 70 mT. The behaviours indicate that magnetite dominates the ChRM carriers in the Wenshan lacustrine sediments. After stepwise AF demagnetization, a total of 217 specimens (92 %) gave reliable characteristic remanence directions.

Two magnetostratigraphic zones were identified from the Wenshan palaeo-lake sediments: a reverse polarity from the top to 145 m, and a normal polarity from 145 m to the bottom of the section (Fig. 4f). We correlated the polarity sequence to chrons C5Br–C5Cn of the Astronomically Tuned Neogene Time Scale (ATNTS2012) [37], thus constraining the Wenshan section to a time interval ranging from ~ 16.5 to 15.2 Ma based on the following reasons.

First, Qi described a *Gigantamynodon* fossil [38] in the nearby Maguan County [30], which is about 50 km southeast of Wenshan. The sediments of Wenshan and Maguan have been inferred to be of the same age [30] although we cannot discount correlation difficulties due to undiscovered faulting in this region. The morphology of the *Gigantamynodon* is similar to the *Plesiaceratherium* discovered in the upper Dingqing Formation of the Lunpola Basin found in the Tibetan Plateau and Shanwang Basin, eastern China [39]. The latter has been well dated between 18 and 16 Ma by $^{40}\text{Ar}/^{39}\text{Ar}$ dating [40]. Moreover, a bentonite layer intercalated between the middle to lower Dingqing Formation has been dated by SIMS U–Pb zircon to 23.5 ± 0.2 Ma [41]. Therefore, the age of the Wenshan sediments is around the early Miocene.

Second, our $\delta^{13}\text{C}_{\text{org}}$ values do not show any significant contribution to the vegetation biomass from C_4 plants, which started to be important components of the flora only since the start of the Late Miocene [42–44], suggesting that the age of the Wenshan sediments predates the Late Miocene.

Third, previous magnetostratigraphic investigations on similar lithological facies in the nearby Dali and Xiaolongtan Basins suggest a sediment accumulation rate of 9–23 cm/ka [45, 46]. Considering that the Dali Basin, Xiaolongtan Basin and Wenshan Basin share similar geological settings and lithologies, the sediment accumulation rate in the Wenshan Basin might be similar to that of Dali and Xiaolongtan Basins. Thus, the 270 m section in the Wenshan Basin should span at least 1 Ma. Moreover, the lithology from the bottom to the top of the studied section is homogeneously mudstone, suggesting that there should be no large variation of sediment accumulation rate during deposition. Therefore, the 128 m normal polarity and the 145 m reverse polarity would represent similar time intervals.

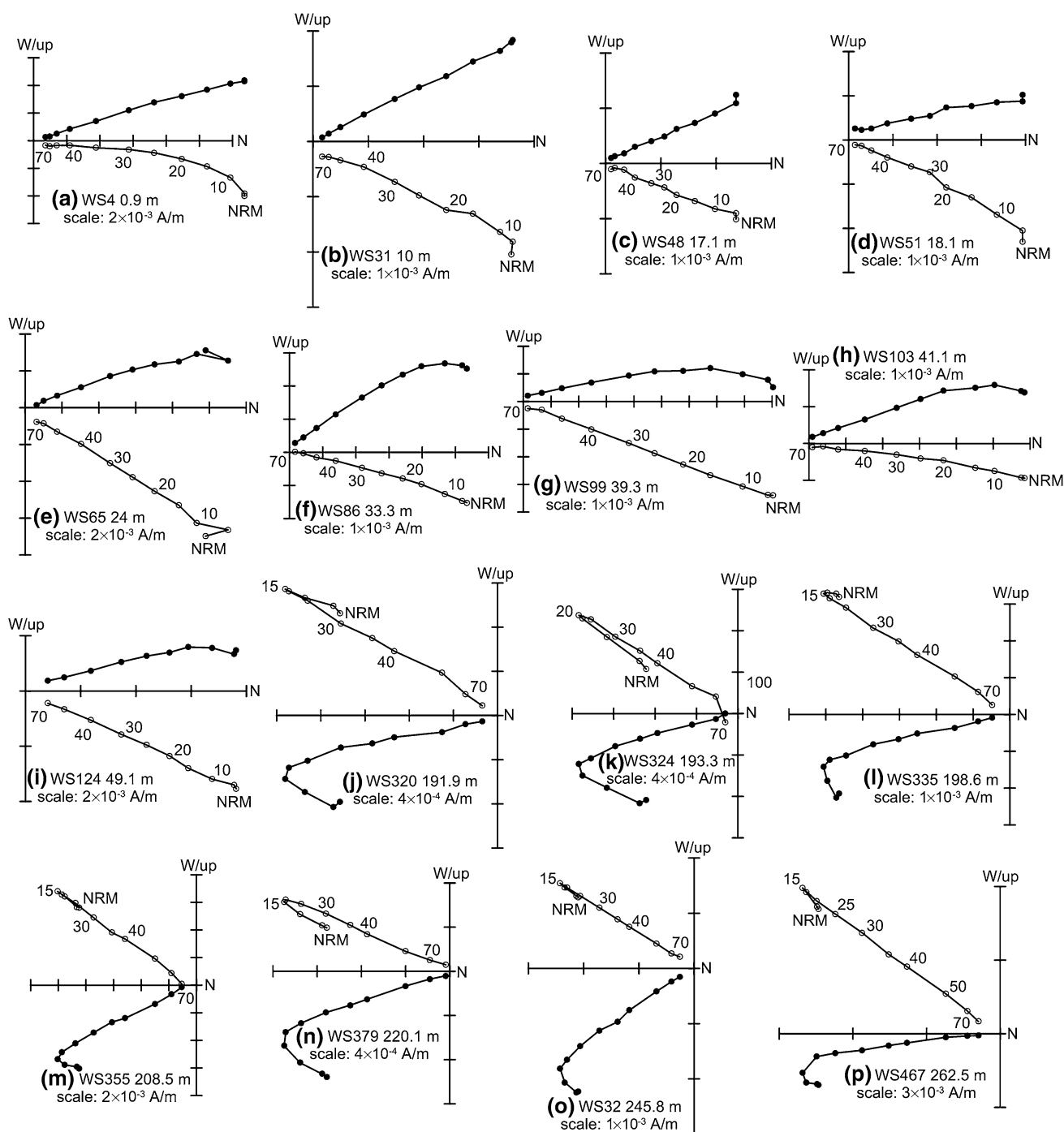


Fig. 3 Orthogonal projections of progressive alternating field demagnetization for representative samples. Solid (open) circles represent the horizontal (vertical) planes. The numbers refer to alternating fields in mT

Combining these three arguments and the character of ATNTS2012 in the late Early-Middle Miocene, we suggest that the most reasonable correlation is that the Wenshan sediments range from ~ 16.5 to 15.2 Ma, within the Mid-Miocene Climatic Optimum. The average sampling resolution is therefore 2–5 ka for the geochemical samples.

3.2 Geochemical data

The $\delta^{13}\text{C}_{\text{org}}$ values vary widely (Fig. 5), spanning from -25.5 ‰ to -32.4 ‰ and have a mean value of -28.9 ‰ (Table 1). TOC and TN values range from 0.04 % to 7.6 % (mean value is 2.3 %) and from 0.04 % to 2.5 % (mean

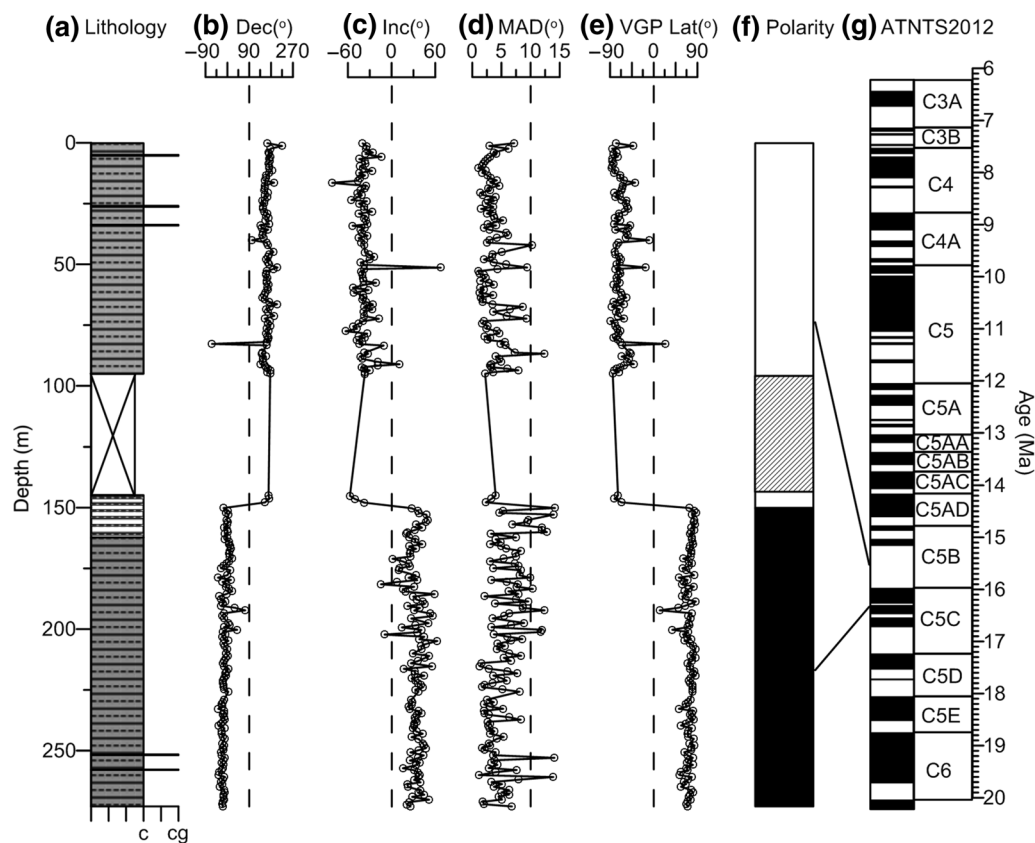


Fig. 4 **a** Lithology, and **b–f** magnetostratigraphy results of the Wenshan section, and **g** its correlation with the ATNTS2012 [37]. *Dec*: Declination; *Inc*: Inclination; *MAD*: maximum angular deviation; *VGP Lat*: latitude of virtual geomagnetic pole; *c*: clay; *cg*: conglomerate

1.4 ‰), respectively. The lowest TOC values are found in the sandstone samples, which is consistent with the fact that finer sediments usually yield higher TOC values [36]. The C/N ratios fluctuate from 1.5 to 54 with a mean value of 18.7.

Employing the Constrained Incremental Sum of Squares (CONISS) algorithm (calculated in the software R using the package “rioja”), we distinguish two zones (Zones 1 and 2) based on the values of the geochemical parameters (see Fig S4, online). Zone 1 is ~290 m thick and according to our new chronology, the boundary between Zone 1 and Zone 2 is situated ~30 m above the C5Cn/C5Br boundary dated at 15.97 Ma. Despite short-term variability within our measurements, there are visible long-term trends on a larger scale. Specifically, all four geochemical parameters mostly exhibit values above the overall mean in Zone 1 and below the overall mean in Zone 2 and their medians are statistically significantly different (Table 1; Fig. 5). The highest value for each geochemical parameter is attained in Zone 1, while the lowest value is found in Zone 2. This suggests that a significant environmental change occurred between Zone 1 and Zone 2.

4 Discussion

4.1 Environmental significance of geochemical proxies

During sediment transport, a significant proportion of organic matter is usually lost through cycling [47, 48], but the original signatures of C/N and $\delta^{13}\text{C}_{\text{org}}$ of bulk organic matter are not markedly affected in aqueous environments [47–49]. Small variations are observed in the isotopic composition of soil organic matter during litter decay [48], but this is likely to have little impact in Wenshan compared to the large range in $\delta^{13}\text{C}_{\text{org}}$ values. After deposition at the bottom of the lake, degradation of organic matter continues, but it is still possible that the original signature of bulk organic matter is retained despite long residence times in sediments, potentially allowing palaeoenvironmental interpretations [50].

C/N ratios provide insights into the source of organic matter. Algae usually have a C/N ratio ranging from 4 to 10 because algae are rich in proteins (N-compounds) and devoid of cellulose (C-compounds) [35, 49]. Conversely, C_3 and C_4 land plants are rich in C-compounds (cellulose and lignin), and thus have C/N ratios ranging from 18 to

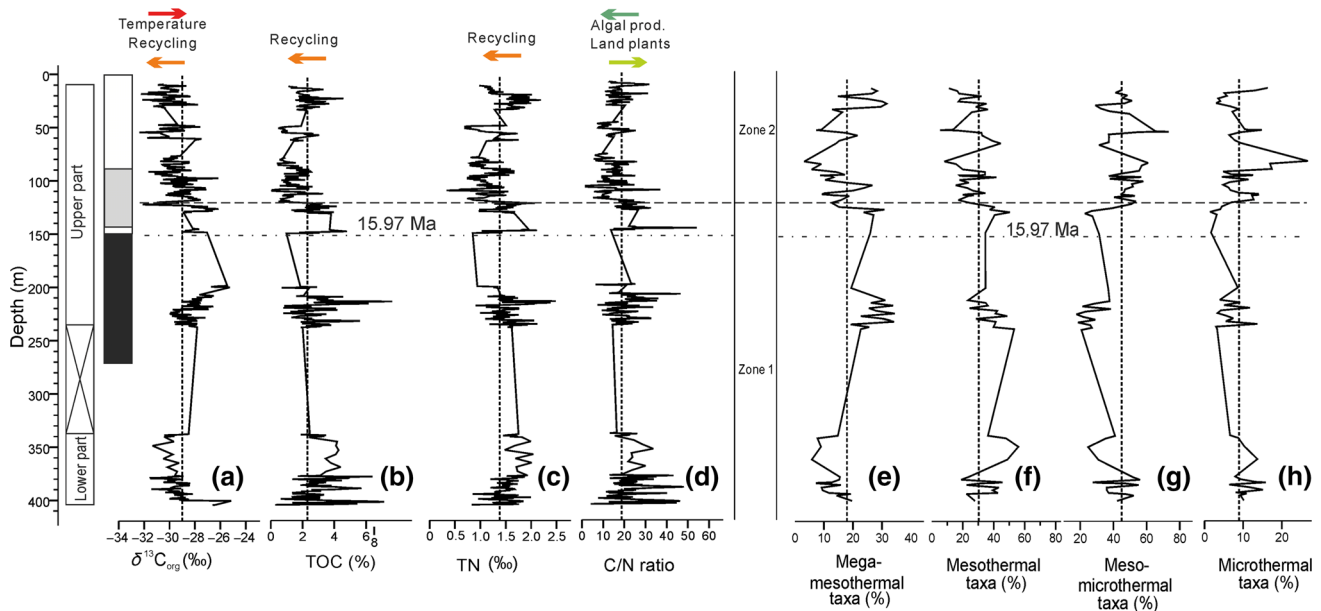


Fig. 5 (Color online) Magnetostratigraphic, geochemical indices (a, b, c and d) and pollen elements [11] (e, f, g and h) of the palaeo-lake in Wenshan. For the full list of pollen taxa, see Table S1, online. The two zones, separated by the horizontal dashed line, were determined by the CONISS algorithm based on the values of geochemical parameters (see Fig. S4, online). Note that the TN values are expressed in permil while the TOC values are expressed as percentages, however, TOC and TN are expressed in the same units when calculating the C/N ratios. The horizontal dashed and dotted line represent the C5Cn/C5Br geomagnetic reversal dated at 15.97 Ma [37]. The vertical dashed line is the mean value of each parameter for the complete sedimentary succession

more than 100 [35, 49]. Ratios between 10 and 18 reveal that the source of organic matter was a combination of algae and vascular land-plants. Therefore, the C/N ratios reflect the proportion of autochthonous (within the lake) and allochthonous (outside the lake) organic matter preserved in the sediments. In Wenshan, the C/N ratios in the whole sedimentary succession may seem unusually high, but similar high values have been found in Xingyun Lake (Yunnan) between 12 and 1.2 ka [23]. The C/N ratios in Zone 1 (average 22.5) indicate higher relative contributions of terrestrial organic matter, while the lower values in Zone 2 (average 14.7) imply that the relative algal contribution increased. The C/N values of a few samples are very low (<4), probably because when TOC values are lower than 0.3 %, the proportion of inorganic TN can be a large fraction of the residual nitrogen and could biased C/N values to artificially low values [36]. Therefore, C/N with very low TOC should be taken cautiously, but when TOC values are above 1 % the C/N ratio remains a reliable indicator of organic matter source [36].

The $\delta^{13}\text{C}_{\text{org}}$ values can be used to distinguish between photosynthetic pathways: C_3 plants fractionate the source of carbon by about -20‰ while in C_4 plants it is only about -7‰ . As a result, C_3 plants have values averaging -27‰ , while for C_4 plants it is -14‰ [51]. Terrestrial plants using the CAM photosynthetic pathway (crassulacean acid metabolism) have $\delta^{13}\text{C}_{\text{org}}$ values ranging from

-10‰ to -20‰ . However, CAM plants, which are mainly desert plants and succulents [51], are unlikely to contribute significantly to the $\delta^{13}\text{C}_{\text{org}}$ signal in a monsoonal climate such as Wenshan [11]. In Wenshan, the average of $\delta^{13}\text{C}_{\text{org}}$ values is -28.9‰ (Table 1) and implies that C_4 plants did not contribute significantly to the biomass preserved in the lake (Fig. 6). This agrees with current evidence that, in Asia, C_4 plants only began to be noticeable components of terrestrial biomass from the Late Miocene [42, 44].

The $\delta^{13}\text{C}_{\text{org}}$ signature can also be influenced by environmental factors. The values of $\delta^{13}\text{C}_{\text{org}}$ are primarily controlled by the isotopic composition of the carbon source during photosynthesis [34, 35, 52]; an increase in the isotopic composition of the source of carbon (such as atmospheric or aqueous CO_2) produces an increase in $\delta^{13}\text{C}_{\text{org}}$ [34, 35]. When primary productivity is high, $\delta^{13}\text{C}_{\text{org}}$ values increase because C_3 plants preferentially absorb ^{12}C during photosynthesis, leaving the remaining carbon enriched in ^{13}C if the carbon source is not replenished [35]. Furthermore, the isotopic composition of carbon from a recycled or respired source is lower than that of atmospheric CO_2 , and consequently decreases the values of $\delta^{13}\text{C}_{\text{org}}$ [52]. Therefore, high $\delta^{13}\text{C}_{\text{org}}$ values might reflect high primary productivity or low carbon cycling, or a combination of both. Finally, the $\delta^{13}\text{C}_{\text{org}}$ values can reflect changes in the concentration of atmospheric CO_2 [35] and climatic

Table 1 Mean values of the geochemical parameters

Zone	1	2	Zone 1/Zone 2 ^a	Complete section
$\delta^{13}\text{C}_{\text{org}}$ (‰)	-28.7 ± 1.27	-29.6 ± 1.13	$U = 6922; P < 0.001$	-28.9 ± 1.75
C/N (atomic)	22.5 ± 8.5	14.7 ± 5.9	$U = 4480; P < 0.001$	18.7 ± 8.6
TOC (%)	2.99 ± 1.29	1.60 ± 0.83	$U = 6443; P < 0.001$	2.31 ± 1.32
TN (%)	1.53 ± 0.29	1.23 ± 0.35	$U = 5361; P < 0.01$	1.38 ± 0.38

The median of each geochemical parameter in Zone 1 and Zone 2 are significantly different

^a To test the significance of the difference between the mean values of Zone 1 and Zone 2, we used the Mann–Whitney test instead of the *t* test because the data were not normally distributed

conditions, such as temperature [52, 53] and precipitation [54, 55]. The controlling climatic factor depends on which one limits plant growth [54]. For terrestrial plants, $\delta^{13}\text{C}_{\text{org}}$ values are often negatively correlated with precipitation [54, 55]. In Wenshan, however, because organic matter originated from terrestrial as well as aquatic plants, $\delta^{13}\text{C}_{\text{org}}$ is more likely to be related to temperature than precipitation. To better understand the climatic factors influencing $\delta^{13}\text{C}_{\text{org}}$, we compared our geochemical results with previously published palynological results from the same section [11]. We assessed possible correlations between the geochemical parameters and the palynological taxa grouped by ecological preferences (see online Table S1 for the full list of woody pollen taxa). The weak but positive correlation between $\delta^{13}\text{C}_{\text{org}}$ and mega-mesothermal floral elements ($r = 0.264$, $P < 0.05$; Fig. 7a; and online Table S2) corroborates the possible relationship between $\delta^{13}\text{C}_{\text{org}}$ and temperature.

The values of TOC reflect the balance between primary productivity and preservation of organic matter within the lake. Thus, high TOC values in Zone 1 may indicate a high primary productivity or high preservation of organic

matter. Preservation of organic matter is enhanced when the lake bottom is poorly oxygenated, promoting anaerobic decay, which is less efficient than aerobic cycling, consequently increasing TOC values [49]. In Wenshan, TOC positively correlates with TN ($r = 0.738$, $P < 0.001$; Fig. 8a and online Table S3 for the full results), suggesting that high values of TOC may be linked to high rates of organic matter preservation. Additionally, TOC positively correlates with the C/N ratio ($r = 0.916$, $P < 0.001$; Fig. 8b), implying that high values of TOC may be associated with a higher input of terrestrial plants. An et al. [22] studied a lacustrine section in Heqing Basin (Yunnan) and used TOC values that positively correlate with the C/N ratio as a proxy for monsoon precipitation. An et al. [22] argue that when monsoon precipitation increases, it leads to more abundant terrestrial vegetation biomass and greater input of land-plant matter in the lake, and therefore to both high TOC and C/N values. The interpretation of An et al. [22] is confirmed by the negative coupling between TOC and the Rb/Sr ratio. However, using TOC and the C/N ratio as a proxy for precipitation should be regarded with caution in the context of Wenshan. For example, increased TOC values can also be due to higher preservation of organic matter rather than an increase in primary productivity, whilst higher C/N values can reflect a decrease in lacustrine primary productivity, even though the terrestrial contribution remains unchanged. In Wenshan, the values of TOC, TN and C/N positively correlate with mesothermal elements, but negatively correlate with meso-microthermal elements (Fig. 7), suggesting that TOC, TN and C/N values are influenced by climate-related factors. It is, however, difficult to disentangle the effect of temperature and precipitation as they may co-vary.

4.2 Environmental interpretation

In Zone 1, C/N values are often above 18, implying that preserved organic matter mainly comes from land-plants. Thus, the $\delta^{13}\text{C}_{\text{org}}$ values reflect conditions in the catchment area. High $\delta^{13}\text{C}_{\text{org}}$, TOC and TN values suggest a high primary productivity in the catchment area, or high

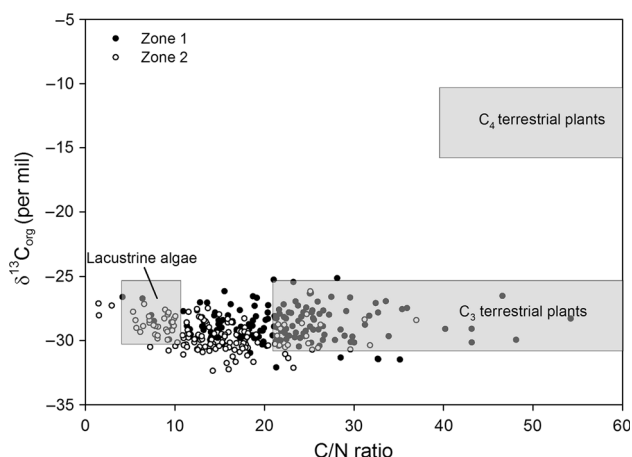


Fig. 6 C/N ratios and $\delta^{13}\text{C}_{\text{org}}$ values plotted against modern global fields (grey boxes) for lacustrine algae, C_3 and C_4 land plants, after Meyers and Teranes [35]

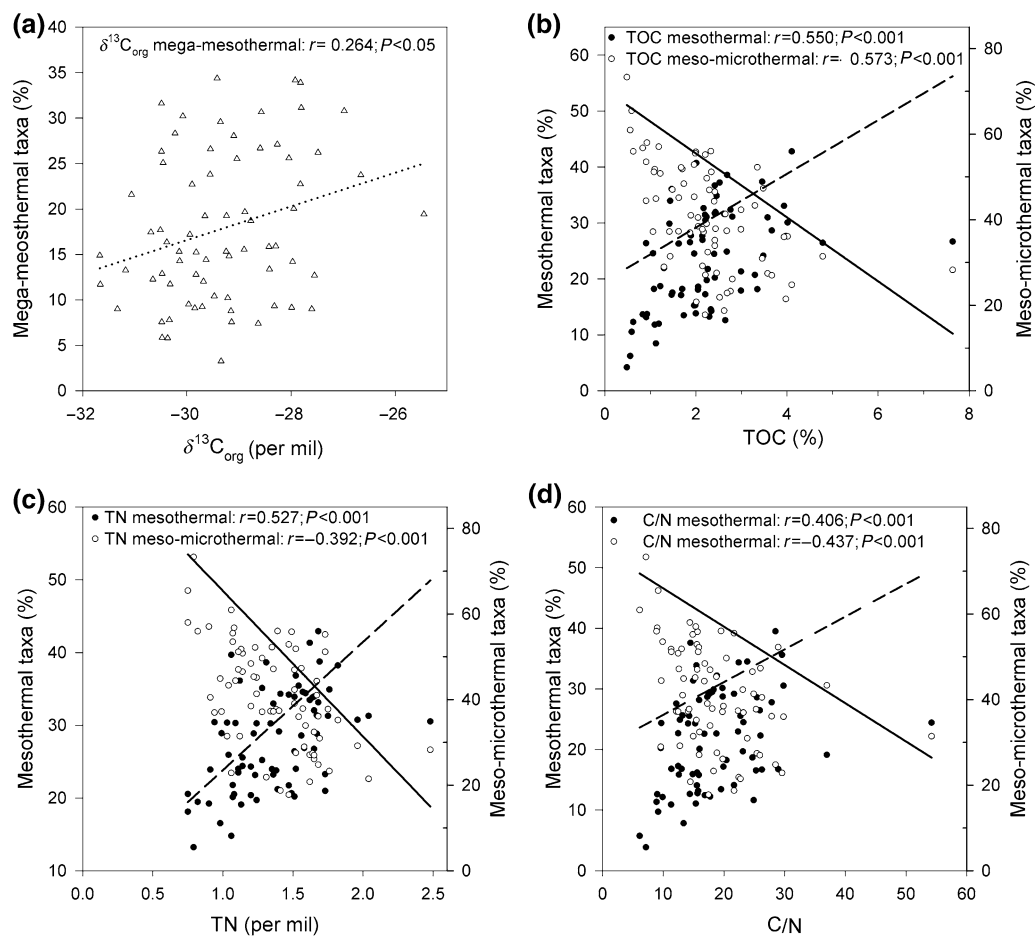


Fig. 7 Correlation between the geochemical and palynological parameters. Note that for this analysis, we used only the samples for which geochemical and palynological data were available (72 samples)

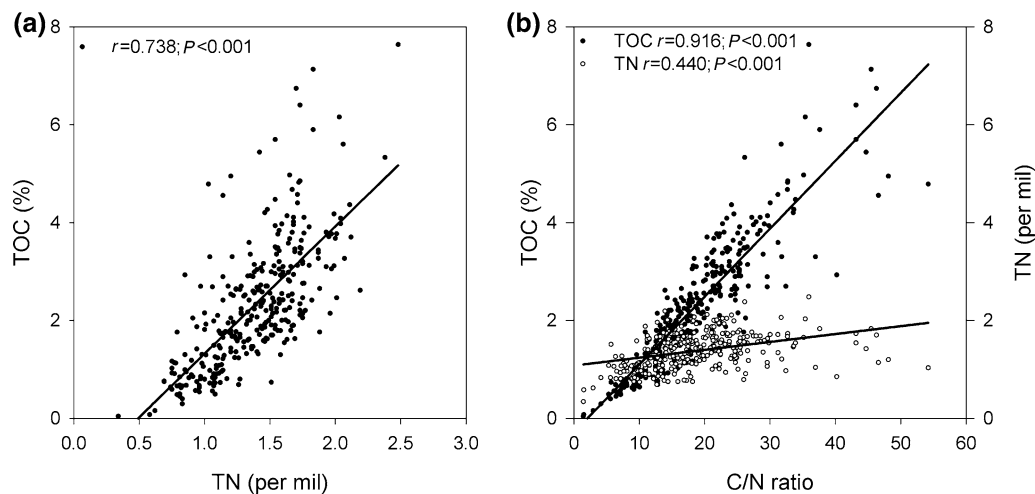


Fig. 8 TOC positively correlates with TN and C/N

preservation of organic matter within the lake, or a combination of both. If high TOC values reflected high precipitation, as in An et al. [22], we would expect high

nutrient influx brought about by inwash events [56]. Because algal production is low, it suggests that the nutrient supply is also low, possibly because of poor water

circulation, which may be due to stratification caused by high evaporation under high temperatures. This is corroborated by the high values of megathermal pollen taxa and $\delta^{13}\text{C}_{\text{org}}$ in Zone 1 (Fig. 5). We propose therefore that high $\delta^{13}\text{C}_{\text{org}}$, TOC and TN values may primarily reflect high organic matter preservation.

In Zone 2, C/N values are as low as 6, indicating a substantial increase in the proportion of autochthonous vs allochthonous input in the preserved organic matter (Fig. 5). Thus, geochemical parameters reflect the conditions of the lake and its catchment area. Zone 2 also witnessed a decrease in megathermal pollen taxa and $\delta^{13}\text{C}_{\text{org}}$ values, suggesting lower temperatures. The decrease in the TOC, TN and $\delta^{13}\text{C}_{\text{org}}$ values can be due to a combination of a decrease in primary productivity and a decrease in preservation of organic matter. However, the decrease in TOC, TN and $\delta^{13}\text{C}_{\text{org}}$ values is unlikely to have been caused by a decrease in primary productivity, as the C/N ratios signal the intensification of algal productivity. We therefore propose that $\delta^{13}\text{C}_{\text{org}}$, TOC and TN values may reflect a relative decrease in preservation of organic matter, possibly due to an increase in organic matter cycling. High levels of organic matter cycling would increase the availability of nutrients and consequently increase lacustrine primary productivity, as evidenced by the lower C/N ratios.

4.3 Possible mechanisms for palaeoenvironmental change

Our magnetostratigraphic results indicate that the sediments were deposited during the MMCO. This is the first time that the MMCO has been reported in Yunnan. We have seen that there was a significant environmental shift between Zone 1 and Zone 2 as evidenced by the relative increase of the autochthonous vs allochthonous contribution in the organic matter preserved in the sediments. Lakes are very sensitive to local perturbation, so we first examine the factors acting locally before considering factors acting globally.

The relative increase of the autochthonous versus allochthonous contribution may be related to the following factors: (1) decreasing the contribution of terrestrial organic matter; (2) increasing the contribution of lacustrine primary productivity; or (3) affecting both the terrestrial and lacustrine input.

The decrease in the contribution of land-plant organic matter may be due to a shift in the terrestrial vegetation dynamics, such as a retreat in forest cover in Zone 2. However, if the lower relative terrestrial input was due to an opening in vegetation cover, we should see an increase in the abundance of pioneer taxa, such as *Alnus* and *Cupressacea/Taxodiaceae* [21, 56]. We did not observe such a pattern, implying that there is no evidence to support

a retreat of the forest cover in Wenshan. It is possible, however, that the sampling resolution of the present study is too low to capture the successional shift.

A change in pattern of organic matter cycling might be responsible for the increase in algal primary production in Zone 2. More favourable conditions for lacustrine algal productivity may be due to improved cycling of organic matter and nutrients [35], explaining the lower TOC and TN values in Zone 2. Furthermore, if cycling is enhanced, it would also decrease the $\delta^{13}\text{C}_{\text{org}}$ values because the isotopic composition of the recycled carbon source is lower than that of CO_2 during photosynthesis. Possible reasons for improved recycling of organic matter could be due to intensified lake circulation, because of either a change in the lake basin configuration, or a change in climate, or both. The lake-basin configuration could have evolved from a closed lake in Zone 1 to an open, through-flow lake in Zone 2. Indeed, the Ailao Shan-Red River Fault was active during the Miocene [25, 26] and could have diverted freshwater toward the lake catchment and contributed to a change in the lake-basin configuration. A change in climate may also have improved the lake water mixing, because when temperatures are lower, evaporation decreases, subsequently increasing the precipitation/evaporation ratio and possibly increasing the lake level [57].

The decrease in C/N ratio between zones 1 and 2 is concomitant with an increase in the percentages of meso-microthermal and microthermal floral elements, and a decrease in mega-mesothermal and mesothermal floral elements (Fig. 5). This suggests that the environmental transition might be climate-related and that Zone 2 may have experienced a combination of cooler temperatures and lower precipitation. Lower temperatures and precipitation may have decreased primary productivity in the lake catchment during Zone 2, providing an explanation for lower TOC and $\delta^{13}\text{C}_{\text{org}}$ values, but this would not explain the increase in relative algal primary productivity. However, lower precipitation could have led to lower frequency of inwash events bringing land-plant matter, increasing the relative representation of lacustrine primary production.

Climatic change could be related to local or global climatic perturbation, particularly because the Wenshan sedimentary succession has been interpreted to date from the time of the MMCO. Within the MMCO however, punctuated climatic events have been recognized. The shift between Zones 1 and Zone 2 could reflect the cooling event Mi-2, although this event is dated around 16.0 Ma [58]. Alternatively, a “peak warmth” centred around 15.6 Ma has been described from the Pacific Ocean and Antarctica [2, 6, 8]. It is therefore possible that the boundary between Zones 1 and 2 is related to the end of the “peak warmth”. It is difficult to correlate short-term punctuated climatic events from the oceanic and terrestrial record because

climatic changes are expressed differently in oceanic and terrestrial realms. The response times of the two realms are markedly different. Whatever the case, the rapid environment change between Zones 1 and 2 exemplifies the climatic instability during the MMCO.

Finally, Wan et al. [58] show that during the MMCO, higher temperature and precipitation significantly increased the sediment accumulation rate in the South China Sea, due to stronger chemical weathering and physical erosion caused by warmer temperatures and stronger precipitation. However, the increase in the sediment accumulation rate was not linear and was punctuated by trend reversals [58] mirroring the rapid environmental fluctuations such as that experienced in Wenshan.

However expressed, our record suggests rapid environmental change in Yunnan during the MMCO.

5 Conclusions

We studied a 400 m-thick lacustrine section in Wenshan, southeast Yunnan, to explore environmental change in the lake catchment area. First, we revised the geological age of the section, previously thought to be the Late Miocene. Magnetostratigraphic dating suggests an older age of ca. 15.2–16.5 Ma, and that deposition was during the MMCO. Then, we measured four geochemical indices ($\delta^{13}\text{C}_{\text{org}}$, TOC, C/N and TN) in a sedimentary section of the palaeo-lake to better understand environmental changes during the MMCO. Our results indicate a reversal of the long-term trends of all four geochemical indices between Zones 1 and 2. The reversal occurred near the middle of the sedimentary section, shortly after the C5Cn/C5Br geomagnetic reversal or the Early/Middle Miocene boundary at 15.97 Ma. In order to refine our interpretation of the geochemical parameters, we also assessed possible correlations with pollen elements from the same section. Our measurements demonstrate that the lake catchment area underwent marked changes that may reflect climatic instability during the MMCO.

Specifically, our results point to an increase in the contribution of algal organic matter relative to that of terrestrial C_3 plants preserved in the sediments, which reveals a shift in the lake environment between Zone 1 and Zone 2. The shift in the origin of organic matter found in the sediments may be due to a joint change in climate and organic matter cycling, itself likely influenced by secular climate change as an indirect factor, or tectonic events which may have re-routed water toward the lake basin.

The palynological study supports the climate-change hypothesis, because the lake environment shift was synchronous with a decrease in mega-mesothermal and mesothermal element percentages and an increase in the

meso-microthermal and microthermal element percentages, suggesting that the climate became cooler and possibly drier.

This study establishes that a subtropical lake catchment area such as Wenshan underwent strong environmental changes during the MMCO, demonstrating that although the MMCO was generally a warm event, local climate experienced marked short-term fluctuations.

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Conflict of interest The authors declare that they have no conflict of interest.

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