



1 Lacustrine methane release on the Tibetan Plateau as an
2 important driver of Early Miocene global warming

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4 Cheng Yang^{1,2,3}, Xiugen Fu^{1,2,3,4*}, Jinxian Deng^{1,2,3*}, Hengye Wei^{1,2,3}, Yue Wen^{1,2,3},
5 Tianxiang Wen^{1,2,3}, Shengqiang Zeng^{5*}

6 ¹ School of Geoscience and Technology, Southwest Petroleum University, Chengdu
7 610500, China

8 ² State Key Laboratory of Oil and Gas Reservoir Geology and Exploitation, Southwest
9 Petroleum University, Chengdu
10 610500, China

11 ³ Qiangtang institute of Sedimentary Basin, Southwest Petroleum University, Chengdu
12 610500, China

13 ⁴ Qinghai Provincial Key Laboratory of Plateau Saline-Lacustrine Basinal Oil & Gas
14 Geology, Dunhuang
15 736202, China

16 ⁵ Chengdu Center, China Geological Survey (Geosciences Innovation Center of
17 Southwest China), Chengdu 610218, China

18 *Correspondence to: Xiugen Fu (fuxiugen@126.com), Jinxian Deng (448920726@qq.com),
19 Shengqiang Zeng (zengsq@126.com)

20 Address: School of Geoscience and Technology, Southwest Petroleum University, Chengdu,
21 610500, China

22



23 **Abstract.** The Early Miocene was a key period of significant global warming. While
24 previous studies often attributed this warming to the India-Asia collision and associated
25 volcanism, an alternative mechanism may involve large-scale methane release from
26 organic-rich lake sediments. To test the hypothesis that methane emissions from Tibetan
27 Plateau lakes contributed to Early Miocene warming, we analyzed organic carbon,
28 stable isotopes, and elemental concentrations in samples from the organic-rich
29 Dingqinghu Formation in the Lunpola Basin, central Tibetan Plateau. Our results
30 identify an exceptionally strong positive carbonate carbon isotope excursion ($\delta^{13}\text{C}_{\text{carb}}$
31 up to +13.79‰) within the lacustrine deposits. The large carbon isotope difference
32 between carbonate and organic matter ($\Delta^{13}\text{C} > 32\text{‰}$) indicates that methanogenesis,
33 specifically via acetate fermentation, was the dominant microbial process. Extremely
34 low sulfur contents likely suppressed sulfate-driven anaerobic oxidation of methane,
35 facilitating direct methane release to the atmosphere. Furthermore, volcanic activity
36 during this interval was limited, suggesting a negligible role in carbon cycle
37 perturbations. The close temporal correspondence between Early Miocene warming,
38 rising atmospheric CO_2 , and methane emissions documented on the Tibetan Plateau
39 indicates that methane release from these plateau lakes may have played an important
40 role in driving global warming and increasing contemporary CO_2 levels.

41



1 Introduction

The Cenozoic era witnessed several global warming episodes, such the Paleocene-Eocene Thermal Maximum and the Middle Miocene Climatic Optimum, with temperatures and atmospheric CO₂ concentrations significantly exceeding pre-industrial levels (Methner et al., 2020; Cenozoic CO₂ Proxy Integration Project (CenCO2PIP) Consortium, 2023; Ivany et al., 2025). This period was also defined by continental collision between India and Eurasia and the subsequent uplift of the Tibetan Plateau, a tectonic event that profoundly reshaped Asian climate and potentially influenced global patterns (Wang et al., 2008; Yang et al., 2022; Zhang et al., 2024; Zhang et al., 2025). The temporal coincidence of these events has led to the hypothesis that the India-Asia collision and its associated volcanism may have been a primary driver of Cenozoic warming (Gutjahr et al., 2017; Kender et al., 2021; Tremblin et al., 2022).

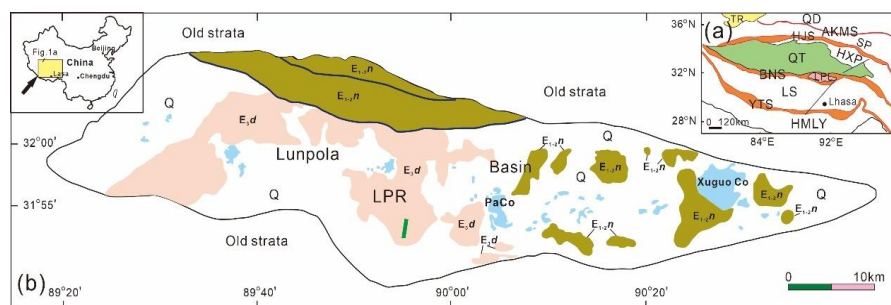


Figure 1: (a) Map of the Tibetan plateau showing major terranes. (b) Simplified geological map of the Lunpola Basin, showing location of sampling section. AKMS, Anyimaqen–Kunlun–Muztagh Suture; BNS, Bangong Lake–Nuijiang River Suture; HJS, Hoh Xil–Jinsha River Suture; HMLY, Himalayas; HXP, Hoh Xili piedmont zone; LPL, Lunpola Basin; LS, Lhasa terrane; QD, Qaidam; QT, Qiagtang Basin; SP, Songpan–Ganzi flysch complex; TR, Tarim Basin; YTS, Yarlung Tsangpo Suture (modified from Fu et al., 2015).

However, Cenozoic volcanism on the Tibetan Plateau, while episodic, was relatively limited in scale (Xie et al., 2024). For instance, during the third volcanic phase (ca. 28–18 Ma), activity was largely confined to restricted areas like the Lunpola Basin, with



64 significantly reduced spatial distribution and magma volume compared to earlier stages
65 (Wang et al., 2024). Consequently, although regional magmatic events reflect the
66 tectonic-magmatic evolution of the Plateau, their associated volatile fluxes were likely
67 negligible on a global scale compared to large igneous provinces (Zhang et al., 2021).
68 This suggests that the direct contribution of Tibetan volcanism to the rise in atmospheric
69 CO₂ and global warming was probably limited.

70 An alternative mechanism for driving warming may involve large-scale methane
71 release from organic-rich lake sediments. During diagenesis and burial, microbial
72 methanogenesis in such sediments can generate substantial methane (CH₄) (Martens
73 and Berner, 1974; Whiticar, 1999; Bastviken et al., 2004; Dean et al., 2018). Under
74 stratified or anoxic lacustrine conditions, this methane can accumulate and be released
75 to atmosphere via ebullition and diffusion (Bastviken et al., 2004; Encinas Fernández
76 et al., 2014; DelSontro et al., 2016). Lakes are a significant natural source of methane
77 emissions, and during specific geological intervals, widespread organic-rich lacustrine
78 systems could have release methane at a scale capable of influencing the global climate
79 system by enhancing the greenhouse effect and perturbing the carbon cycle (Dean et al.,
80 2018; Sun et al., 2022; Zhuang et al., 2023). Consequently, organic-enriched lake
81 sediments act not only a crucial long-term carbon sink but also, under certain conditions,
82 as a potent methane source, with substantial implications for both paleoclimate and
83 contemporary climate systems.

84 During the Miocene, the Tibetan Plateau hosted extensive lacustrine systems. The
85 Lunpola and Nima basins, for example, preserve thick successions of organic-rich
86 deposits and oil shales. In the Dingqinghu Formation, these successions reach
87 thicknesses exceeding 148 m and exhibit total organic carbon (TOC) contents of up to
88 17.60% (Fu et al., 2012; Fang et al., 2020; Fu et al., 2020; Lu, 2023; Zeng et al., 2024).
89 These deposits not only record high organic carbon accumulation but also represent a
90 potential source of large methane emissions during deposition and early diagenesis,
91 which could have contributed to regional or even global warming (Zhou et al., 2024;



92 Nie et al., 2023).

93 To test the hypothesis that methane release from Tibetan Plateau lakes contributed to
94 Early Miocene warming, we conducted organic carbon, stable isotope, and element
95 analyses on samples from the Dingqinghu Formation in the Lunpola Basin (Fig. 1a).
96 Integrated with a robust chronological framework (Mao et al., 2019), this multi-proxy
97 dataset allows us to correlate regional geological events with global climate records and
98 evaluate the potential mechanisms driving Early Miocene climate warming.

99 **2 Geological setting**

100 The Lunpola Basin is a Cenozoic fault-depression basin located on the central Tibetan
101 Plateau, within the central segment of the Bangong-Nujiang Suture Zone (BNSZ), a
102 major tectonic boundary separating the Lhasa and Qiangtang terranes. The basin
103 extends approximately 200 km from east to west and 10–30 km from north to south,
104 and is recognized as the highest-altitude petroleum-bearing basin in the world (Deng et
105 al., 2012; Rowley & Currie, 2006; Fu et al., 2012; Wang et al., 2025).

106 The Tibetan Plateau comprises a series of east-striking tectonic units bounded by
107 major suture zones. From north to south, they are the Kunlun-Qaidam Terrane, the
108 Songpan-Ganzi flysch complex, the Qiangtang Terrane, and the Lhasa Terrane,
109 separated by the Anyimaqen-Kunlun-Muztagh, Hoh Xil-Jinsha River, and Bangong
110 Lake-Nujiang River (BNS) suture zones, respectively (Fig. 1a). The BNS originated
111 from the diachronous, west-to-east closure of the Meso-Tethyan ocean basin,
112 culminating in the Late Jurassic–Early Cretaceous Lhasa-Qiangtang collision (Kapp
113 and DeCelles, 2019). It was later reactivated during contractional events in the latest
114 Cretaceous and Oligocene–early Miocene, which led to the development of
115 intermontane basins in central Tibet, including the Lunpola, Nima, and Gerze basins
116 (e.g., Wei et al., 2017; Han et al., 2019).

117 Structurally, the basin is influenced by strike-slip and thrust faulting, forming a pod-
118 like framework characterized by “north–south structural zoning and east–west fault
119 blocking.” In the north–south direction, the basin exhibits high flanks and a central



120 depression. The southern margin is controlled by thrust belts and uplifts, while the
121 northern part shows localized highs despite significant subsidence and thick
122 sedimentary infill. The cumulative thickness of the Niubao and Dingqinghu formations
123 exceeds 3,000 m. East–west segmentation by faults results in a pattern of peripheral
124 uplift and central subsidence (Lei et al., 1997).

125 The tectonic evolution of the Lunpola Basin involved two main stages. The Eocene
126 rifting, during which the Niubao Formation (E_{1-2n}) was deposited, consisting mainly of
127 alluvial fan to fan-delta coarse clastics. The Oligocene depression, marked by
128 accumulation of the Dingqinghu Formation (E_{3d}), dominated by deep lacustrine fine-
129 grained mudstones and oil shales (Deng et al., 2012; Lu, 2023). The Dingqinghu
130 Formation is distributed primarily in the central and western parts of the basin, with
131 thickness varying from 300 to 1,100 m due to differential basement subsidence. It
132 comprises greenish-gray shales, mudstones, and oil shales interbedded with thin
133 sandstones, interpreted as having been deposited in a low-energy, semi-deep to deep
134 lacustrine environment under stable and strongly reducing conditions (Fu et al., 2015).

135 This study focuses on the middle member of the Dingqinghu Formation, which is
136 Early Miocene in age (ca. 20.6 ± 0.1 Ma; Mao et al., 2019) and consists of organic-rich
137 mudstones, shales, and oil shales. These oil shale samples are dark brown to black in
138 color and contain high organic matter content.

139 **3 Method**

140 The study area and section location are presented in Fig. 1b. The vertical distribution
141 of samples within the section is presented in Figs. S1. A total of 63 samples were
142 collected from the Lunpola oil shale section. The average vertical sampling interval is
143 60 cm.

144 *3.1 TOC analysis*

145 First, grind the 10 mg rock sample into a powder with a particle size smaller than 200
146 mesh. Then, depending on the lithology and colour of the sample, 0.095–0.105 g of the



147 powdered rock is weighed into a crucible and placed into the container. Slowly add 10%
148 HCl along the inner wall of the container (ensuring it does not overflow the crucible
149 opening), then allow it to decarburise for over 24 hours until the reaction is complete.
150 Remove the crucible, rinse the sample with high-purity water to wash away residual
151 acid, and continue washing until the rinse water is neutral. After washing, the samples
152 were dried in an electric thermostatic drying oven at 60–80°C and then analysed for
153 TOC content using an LDH CS analyser (Model: TK851-6K) at the Qiangtang Institute
154 of Sedimentary Basin, Southwest Petroleum University. For every 10 samples, a
155 parallel sample was included for quality control. Results were recorded in wt%, with
156 an analytical accuracy better than $\pm 0.1\%$.

157 3.2 Inorganic carbon and oxygen isotope analysis

158 Carbon isotope samples were analysed in the laboratory of the Qiangtang Institute of
159 Sedimentary Basin. First, an appropriate amount of carbonate sample was ground with
160 an agate mortar to less than 200 mesh, then dried in an oven at 60°C for approximately
161 4 hours to remove adsorbed water. Then, approximately 1 mg of the sample was placed
162 into the sample tube of the GasBench Plus sample preparation system. The sample was
163 dried at 70°C, the tube was sealed, and the air in the sample tube was purged with high-
164 purity helium (He). Using an acid pump with an acid needle, excess 100% phosphoric
165 acid (H_3PO_4) was added to the sample tube. The phosphoric acid was allowed to react
166 with the carbonate sample to produce CO_2 gas. The generated CO_2 was then carried by
167 high-purity helium into the Delta Q (Thermo Fisher) system, where the carbon and
168 oxygen isotopic compositions were measured. A standard reference material was
169 analysed after every 12 samples for quality control. Results were reported in $\delta^{13}\text{C}$ V-
170 PDB and $\delta^{18}\text{O}$ V-PDB values relative to the PDB standard. The GB04416 carbonate
171 standard yielded $\delta^{13}\text{C} = 1.67 \pm 0.05\%$, and measurement accuracy was $\pm 0.05\%$.
172 For carbonate samples, analytical reproducibility of replicate standards was better than
173 $\pm 0.07\%$.



174 3.3 *Organic carbon isotope analysis*

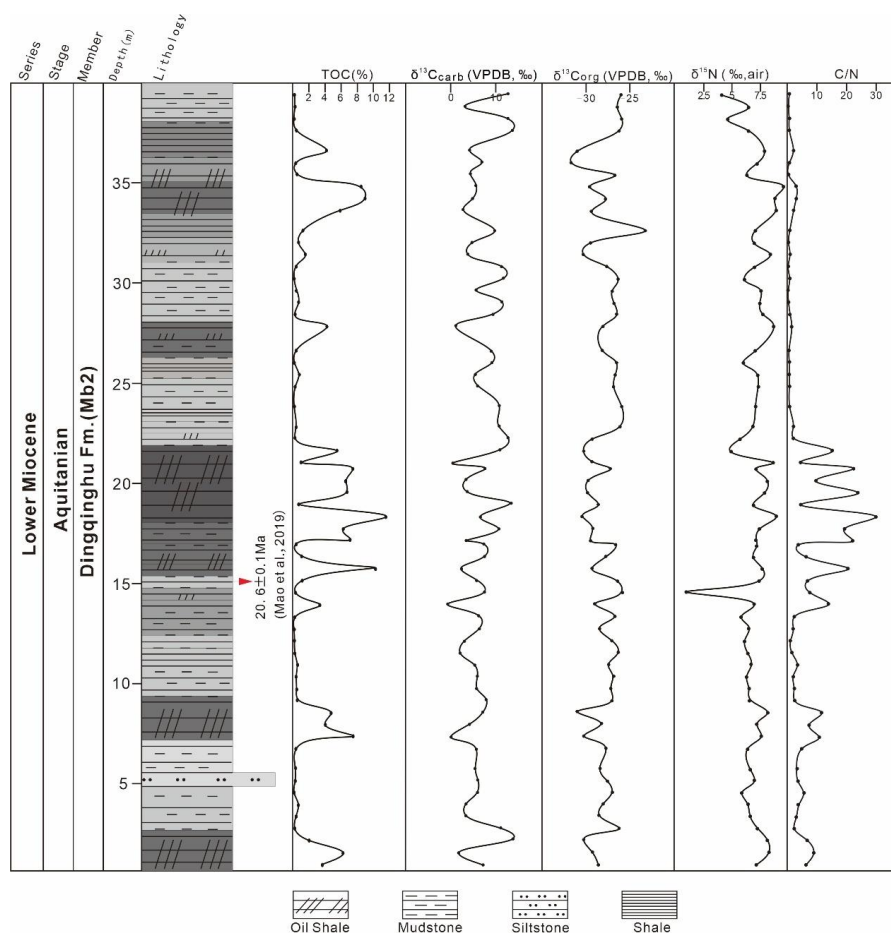
175 The whole rock powder sample was decarbonised with 10% HCl for 48 hours to
176 eliminate all carbonate components, then washed with ultra-pure water until neutral,
177 and the residual sample was dried, ground to less than 200 mesh, weighed 0.2 to 5 mg
178 of residual powder sample into a tin cup, and tightly wrapped into cubes. Under the He
179 atmosphere, the gas is sent into the high-temperature oxidation tube through the EA
180 auto-sampler and instantly oxidised at 980°C in an oxygen atmosphere to generate a
181 mixture of various gas components such as NO, N₂O, N₂O₂, N₂, CO, CO₂, H₂O, SO₂
182 and halogen. In the reaction tube, CO is oxidised to CO₂ by chromium oxide and silver-
183 coated cobalt oxide, and SO₂ and halogen gases are removed. Subsequently, in a
184 reduction tube at 650°C, copper wire reduces nitrogen oxides to N₂ and absorbs excess
185 O₂. The resulting helium and carbon dioxide gases pass through a magnesium
186 perchlorate chemical trap to remove moisture, and then are separated by a
187 chromatographic column into N₂ and CO₂, which enter a thermal conductivity detector
188 (TCD) to determine the carbon content. A small amount of CO₂ is then introduced into
189 a connected Delta Q mass spectrometer via a continuous flow ConFIV system to
190 measure the isotope ratio. All carbon isotope values are reported in the conventional δ
191 -notation in per mil relative to V-PDB (Vienna-PDB). The accuracy and reproducibility
192 of the analysis were checked through repeated tests of the international standard sample
193 USGS40, and the accuracy (1 σ) was less than 0.09‰. The repeatability test accuracy
194 of the sample is less than 0.13‰.

195 3.4 *Total nitrogen (TN) and nitrogen isotope ($\delta^{15}\text{N}$)*

196 The total nitrogen (TN) and nitrogen isotope ($\delta^{15}\text{N}$) contents of samples were measured
197 at Qiangtang Institute of Sedimentary Basin, Southwest Petroleum University, China,
198 using a Vario Macro Cube elemental analyser (Elementar, Hanau, Germany).
199 Approximately 200 mg of the powder samples were weighed, and then add 50 mg of
200 WO₃ oxidant. The mixture was tightly enclosed with a 35 × 35 mm tin capsule and
201 prepared for analysis. Standard deviations for carbon, nitrogen and sulfur contents are



202 <0.05 wt% (1 σ) based on replicate analyses of multiple samples. Thus, the analytical
 203 precision of TN measurements was always better than 0.05 wt.%.



204
 205 Figure 2: TOC, $\delta^{13}\text{C}_{\text{carb}}$ (the Carbonate carbon isotope), $\delta^{13}\text{C}_{\text{org}}$ (the Organic Carbon Isotope), $\delta^{15}\text{N}$
 206 (the Organic Nitrogen isotope) and C/N ratio data in Dingqinghu Formation of lunpola basin.

207 3.5 The major elements and trace elements

208 Elemental analyses were conducted at the Qiangtang Basin Research Institute,
 209 Southwest Petroleum University. Powdered samples (<200 mesh) were packed into
 210 Chemplex cups, compacted, and sealed with polypropylene film. Major and selected
 211 trace element concentrations (including Ca, Mg, Ba, Cu, Ni, V and Sr) were measured
 212 using a Bruker S1 TITAN 800 X-ray fluorescence (XRF) spectrometer manufactured



213 by Bruker, Germany. The standard deviation for major elements was better than $\pm$
 214 0.05%, and that for trace elements was better than $\pm 20 \mu\text{g/g}$.

215 **4 Result**

216 *4.1 TOC content*

217 The total organic carbon content of mudstone and oil shale samples from the Second
 218 Member of the Dingqinghu formation is illustrated in Fig. 2. The TOC values exhibit a
 219 wide variation range (0.12%-11.54%), with an average of 2.35%, categorizing these
 220 strata as high-quality source rocks. Notably, the oil shale intervals demonstrate
 221 exceptional organic enrichment, characterized by TOC values spanning 0.56%-11.54%,
 222 with an average of 5.37%, which exceeds the threshold for effective hydrocarbon
 223 generation in lacustrine systems. In contrast, the interbedded mudstones and shales
 224 display comparatively lower TOC concentrations, ranging from 0.12% to 7.36%, with
 225 an average of 0.91%.

226 *4.2 Stable Isotope geochemistry*

227 As shown in Fig. 2, the variations of $\delta^{13}\text{C}_{\text{carb}}$ values of the second member of the
 228 Dingqinghu formation exhibit an extreme positive excursion, ranging from -0.84‰ to
 229 13.79‰, with an average of 6.59‰. The variation curve of $\delta^{13}\text{C}_{\text{carb}}$ values reveals that
 230 mudstone intervals display the most pronounced positive excursion (1.23‰–13.67‰,
 231 averaging 7.39‰), while oil shale intervals show relatively lower values (-0.84‰–
 232 13.79‰, averaging 4.91‰). The $\delta^{13}\text{C}_{\text{org}}$ values of the Second Member of the
 233 Dingqinghu Formation range from -31.64‰ to -23.31‰, with an average value of -
 234 27.97‰. The $\delta^{13}\text{C}_{\text{org}}$ curve demonstrates a negative excursion in the oil shale layers,
 235 where values vary between -31.03‰ and -26.76‰ (average: -29.34‰). In contrast,
 236 the mudstone intervals exhibit a positive excursion, with a broader $\delta^{13}\text{C}_{\text{org}}$ range of -
 237 31.64‰ to -23.31‰ and an average of -27.33‰. The curves of $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{org}}$
 238 exhibit high coupling, which reflect the synergistic response mechanism of lake carbon
 239 cycle and climate environment. The organic nitrogen isotope values ($\delta^{15}\text{N}(\text{‰,air})$) of



240 the Second Member of the Dingqinghu Formation range from 1.06‰ to 9.74‰, with
 241 an average value of 7.11‰.

242 **5 Discussion**

243 *5.1 Influence of modifying factors*

244 Since both carbonates and organic matter can preserve diagenetic information rather
 245 than primary depositional signals after sedimentation, the influence of diagenesis must
 246 be evaluated prior to interpreting the isotopic compositions. Following precipitation in
 247 lake water bodies, carbonate minerals undergo a series of diagenetic alteration
 248 processes during burial, influenced by temperature and pore fluids. These processes,
 249 including dissolution and recrystallization, can lead to isotopic re-equilibration between
 250 the minerals and fluids (Boever et al., 2017; Hillaire et al., 2021; Huntington and
 251 Petersen, 2023). The extent of diagenetic alteration in lacustrine sediments can be
 252 assessed using carbon and oxygen isotopes of carbonate minerals. During diagenesis,
 253 carbon in pore fluids is typically scarce relative to the reservoir within carbonates,
 254 resulting in a rock-buffered system for carbon. Conversely, oxygen is predominantly
 255 sourced from pore fluids, creating a fluid-buffered system for oxygen. Consequently,
 256 oxygen isotopic compositions are highly susceptible to alteration during diagenesis,
 257 while carbon isotopic compositions generally remain more resistant to change (Wang,
 258 2008; De Boever et al., 2017; Horacek et al., 2007; Ritter et al., 2017; Huntington and
 259 Petersen, 2023). The correlation between carbonate $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{18}\text{O}$ values can
 260 therefore be utilized to assess the degree of diagenetic influence on carbon isotopes. In
 261 the present study, the $\delta^{13}\text{C}_{\text{carb}}$ vs. $\delta^{18}\text{O}$ plot (Figs. S2a) exhibits a weak correlation
 262 ($R^2=0.0132$), indicating that the $\delta^{13}\text{C}_{\text{carb}}$ values retain primary depositional signals.
 263 Furthermore, the Mn/Sr ratio, a sensitive indicator of post-depositional alteration (Hu
 264 et al., 2023), show no significant correlation with either $\delta^{13}\text{C}_{\text{carb}}$ ($R^2=0.0012$, Figs. S2b)
 265 or $\delta^{13}\text{C}_{\text{org}}$ ($R^2=0.017$, Figs. S2c). These results collectively suggest a negligible
 266 influence of diagenesis on the measured carbon isotopic signatures.

267 Diagenetic and metamorphic processes can significantly alter the $\delta^{15}\text{N}$ values of lake



268 sediments. During diagenesis, microbial decomposition releases ^{14}N -enriched NH_4^+
 269 into the pore water, resulting in ^{15}N enrichment of residual organic matter and more
 270 positive ^{15}N values (Macko et al., 1987; Altabet, 1988; Lourey et al., 2003; Papineau et
 271 al., 2009). Metamorphic processes similarly affect ^{15}N through thermal degradation of
 272 nitrogen-rich compounds, which preferentially releases ^{14}N -enriched NH_3 or N_2 ,
 273 leaving residual organic matter with elevated ^{15}N values (Freudenthal et al., 2001;
 274 Lehmann et al., 2002; Altabet, 2006; Robinson et al., 2012; Stüeken et al., 2016; Xia et
 275 al., 2022). Correlative relationships among TOC, TN, $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ serve as key
 276 indicators for assessing post-depositional alteration. In the present study, we observe
 277 no significant correlations between $\delta^{15}\text{N}$ and TOC (Figs. S2d), TN (Figs. S2e), C/N
 278 (Figs. S2f), or $\delta^{13}\text{C}_{\text{org}}$ (Figs. S2g). This demonstrates the $\delta^{15}\text{N}$ values retain primary
 279 depositional signals without diagenetic or metamorphic overprinting (Kipp et al., 2018;
 280 Mettam et al., 2019).

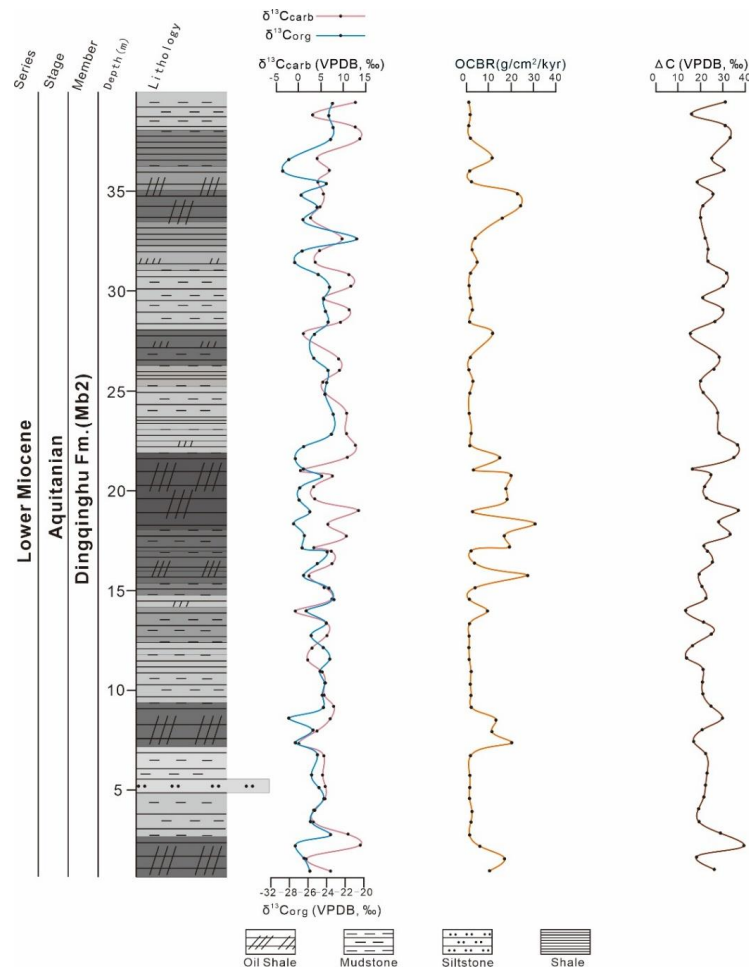
281 Regarding terrigenous influences, lacustrine systems with substantial terrigenous
 282 inputs typically exhibit $\delta^{15}\text{N}$ values significantly lower than autochthonous algal
 283 organic matter (+5‰ to +10‰), as terrigenous organic materials (e.g., plant debris, soil
 284 humus) generally show lower $\delta^{15}\text{N}$ values (-2‰ to +5‰) (Talbot, 2002; Shen et al.,
 285 2006). While a $\delta^{15}\text{N}_{\text{org}}$ -C/N correlation typically indicates terrestrial influence, its
 286 absence in our dataset (Figs. S2f) confirms the $\delta^{15}\text{N}_{\text{org}}$ values are unaffected by
 287 terrigenous input.

288 5.2 Early Miocene positive carbon isotope excursion and methane release

289 The pronounced positive carbonate carbon isotope excursion recorded in the
 290 Dingqinghu Formation is fundamentally controlled by the fractionation mechanisms of
 291 lacustrine carbonate carbon isotopes. Consequently, investigating these fractionation
 292 processes not only elucidates the drivers of extreme $\delta^{13}\text{C}_{\text{carb}}$ excursions but also provide
 293 critical insights into the paleolake's carbon cycle dynamics and ecosystem function
 294 during this interval. Positive $\delta^{13}\text{C}_{\text{carb}}$ excursions in lacustrine systems primarily arise
 295 from isotope fractionation during three key processes: 1) Enhanced organic matter



296 production and burial, 2) Methane generation and release, and 3) Evaporative CO₂,
297 degassing in saline lakes (Michener and Lajtha, 2008; Thottathil et al., 2022). This
298 study will systematically evaluate these mechanisms to determine their relative
299 contributions to the observed $\delta^{13}\text{C}_{\text{carb}}$ excursions.



300
301 Figure 3: $\delta^{13}\text{C}_{\text{carb}}$ (Carbonate carbon isotope), $\delta^{13}\text{C}_{\text{org}}$ (Organic Carbon Isotope), OCBR (Organic
302 carbon burial rate) and $\Delta^{13}\text{C}$ value curves of Dingqinghu Formation.

303 5.2.1 Enhanced organic carbon burial

304 In high-productivity lacustrine systems, variations in carbonate $\delta^{13}\text{C}$ values are
305 predominantly controlled by lacustrine productivity (Teranes and McKenzie,

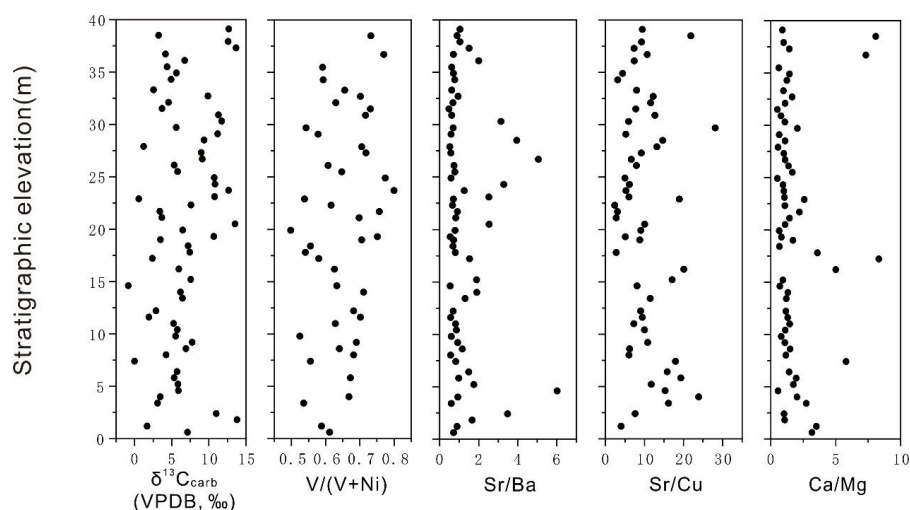


1999; Teranes and Bernasconi, 2005; Zhu, 2013). This relationship arises from photosynthetic fractionation because aquatic plants preferentially assimilate ^{12}C when utilizing CO_2 and bicarbonate from dissolved inorganic carbon (DIC) pools. This process enriches residual DIC ^{13}C , leading to subsequent ^{13}C enrichment in precipitated carbonates (Schelske and Hodell, 1991; Neumann et al., 2002). Under high productivity where aquatic plants preferentially consume dissolved CO_2 , the substantial $\delta^{13}\text{C}$ difference between bicarbonate (relatively ^{13}C -enriched) and aqueous CO_2 (relatively ^{13}C -depleted) further elevates $\delta^{13}\text{C}$ values in both DIC and organic matter (Meyers, 1997; Hodell and Schelske, 1998; Leng and Marshall, 2004; Xu et al., 2006; Zhu et al., 2011). During lake eutrophication, algal blooms generate substantial organic matter while preferentially incorporating ^{12}C . Resultant water column anoxia preserves algal biomass from decomposition, while high sedimentation rates enhance organic carbon burial. Carbonate minerals forming in these ^{12}C -depleted waters consequently exhibit elevated $\delta^{13}\text{C}$ values because sequestered ^{12}C remains buried rather than returning to the water column (Müller and Suess, 1979; Calvert, 1987; Ingall et al., 1990; Tyson, 2001; Xu et al., 2004; Wang et al., 2015; Cartapanis et al., 2016; Megan et al., 2021; Wang et al., 2022; Tegler et al., 2024). For the Dingqinghu Formation, the $\text{W(V)}/\text{W(V+Ni)}$ ratio (Fig. 4) indicates deposition under anoxic/reducing conditions. Corresponding sedimentary facies represent semi-deep and deep lacustrine environments with weak hydrodynamic conditions conducive to sediment preservation. This depositional setting therefore favored efficient organic accumulation and preservation.

Enhanced lacustrine productivity consistently drives both increased carbonate content and elevated $\delta^{13}\text{C}$ values in organic matter (Meyers, 1997; Teranes and McKenzie, 1999; Neumann et al., 2002; Leng and Marshall, 2004; Teranes and Bernasconi, 2005; Xu et al., 2006; Lu et al., 2010). C/N values below 10 across most stratigraphic intervals (Fig. 2) indicate autochthonous productivity dominated by aquatic phytoplankton and algae, with negligible terrestrial input. This confirms TOC



334 serves as a reliable proxy for lacustrine productivity under limited external influences
 335 (Meyers, 1997; Zhu et al., 2013). Critically, intervals displaying positive $\delta^{13}\text{C}_{\text{carb}}$
 336 excursions exhibit extremely low TOC content (average $< 1\%$), indicating depressed
 337 productivity. This inverse relationship demonstrates lacustrine productivity cannot
 338 explain the observed $\delta^{13}\text{C}_{\text{carb}}$ excursions. Further supporting this interpretation, high-
 339 productivity systems typically yield elevated $\delta^{15}\text{N}$ values through two mechanisms.
 340 Phytoplankton preferentially assimilate ^{14}N , enriching dissolved inorganic nitrogen
 341 pools in ^{15}N . Enhanced denitrification under anoxic conditions further amplifies $\delta^{15}\text{N}$
 342 increase in residual nitrogen (Sigman D, 2009). Our data reveal an inverse pattern
 343 where positive $\delta^{13}\text{C}_{\text{carb}}$ excursions systematically coincide with negative $\delta^{15}\text{N}$ shift (Fig.
 344 2), demonstrating that the positive $\delta^{13}\text{C}_{\text{carb}}$ excursion was not caused by enhanced
 345 productivity.



346
 347 Figure 4: $\delta^{13}\text{C}_{\text{carb}}$ (Carbonate carbon isotope), $\text{V}/(\text{V}+\text{Ni})$, Sr/Cu , Sr/Ba and Ca/Mg data in Dingqinghu
 348 Formation of lunpola basin.

349 U-Pb dating and stratigraphic thickness measurements from the Lunpori section
 350 indicate a sedimentation rate (SR) of $\sim 107 \text{ m/Ma}$ for the study interval of the
 351 Dingqinghu Formation (Mao et al., 2019; Xie et al., 2025). The organic carbon burial
 352 rate (OCBR) was calculated using the Eq. (1) (Shen et al., 2015):



$$OCBR (mg/cm^2/kyr) = SR (m/Myr) \times TOC (\%) \times \rho (g/cm^3), \quad (1)$$

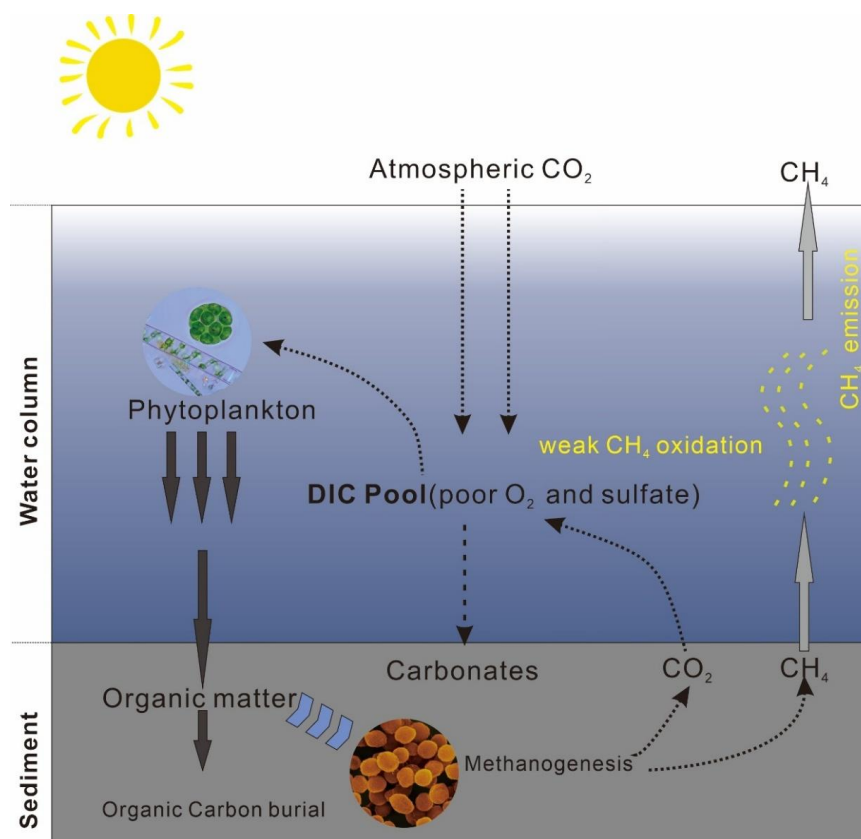
Assuming a bulk rock density (ρ) of $2.5 g/cm^3$, OCBR values for the Dingqinghu Formation in the Lunpori section were derived through unit conversion (summarized in S Table 2). Fig. 3 reveals asynchronous trends between OCBR and the positive $\delta^{13}C_{carb}$ excursion coupled with extremely low correlation coefficient. This strongly suggests that variations in organic carbon burial rate were not the primary driver of the $\delta^{13}C_{carb}$ enrichment observed in the Dingqinghu Formation during this interval.

5.2.2 Evaporation

Strong evaporation in lakes can drive extreme positive carbonate carbon isotope values. This process promotes lake degassing, releasing CO_2 enriched in ^{12}C into the atmosphere. The preferential loss of ^{12}C depletes the DIC pool in light carbon, thereby increasing $\delta^{13}C_{carb}$ (Li and Ku, 1997; Lamb et al., 2007; Zhu et al., 2013; Boscolo-Galazzo et al., 2021). This mechanism is further supported by studies linking evaporative salinity increase to reduced CO_2 solubility and kinetic isotope fractionation during CO_2 efflux (Stiller, 1985). Consequently, arid climates with intense evaporation typically elevate lake salinity and lower lake levels. Palynological data from the Dingqinghu Formation (Xie et al., 2025) reveal alternating dry (e.g., 21.3 Ma, 19.6 Ma) and humid phases (e.g., 20.4 Ma, 20.1 Ma) between 21.4 Ma and 19.4 Ma. At 20.6 Ma, pollen assemblages (*Picea*, 50.1%; *Pinus*, 22.2%; *Abies*, 4.9%; and *Fagaceae*, 5.2%) indicate warm-humid conditions, consistent with magnetostratigraphic cyclostratigraphic evidence of dry-to-wet cycles (Su et al., 2022). Sr/Cu ratios (Fig. 4) independently confirm warm-arid to warm-humid cyclicity during deposition of the Dingqinghu Formation. Crucially, humid phases reduce evaporation, limiting the ^{12}C -enriched CO_2 , and thus diminishing ^{13}C enrichment in the DIC pool. In closed basins like the Lunpola paleolake, evaporation intensity correlates strongly with salinity (Mor et al., 2018; Han et al., 2022), making Ca/Mg ratios an effective salinity proxy (McCormack et al., 2019; Gravina et al., 2022). However, correlation an



381 analyses of Ca/Mg and Sr/Ba versus $\delta^{13}\text{C}_{\text{carb}}$ show no significant relationship bet
 382 ween salinity changes and $\delta^{13}\text{C}_{\text{carb}}$ excursion during the Dingqinghu Formation
 383 deposition. This lack of correlation demonstrates that salinity variations did not
 384 primarily control $\delta^{13}\text{C}_{\text{carb}}$ trend. Integrated assessment of climate proxies, salinit
 385 y indicators, and carbon isotopes indicate that while evaporation may contribute
 386 secondarily to $\delta^{13}\text{C}_{\text{carb}}$ enrichment, it was not the principal driver of the obser
 387 ved positive excursion.



388
 389 Figure 5: Schematic diagram of the carbon cycling pathway for methane release in Dingqing Lake.
 390 Biogenic methane is generated by methanogens in the anoxic sediments, and the water column is under
 391 reducing conditions, lacking electron acceptors such as sulfate and iron ions. The methane directly
 392 discharges into the atmosphere.



393 5.2.3 Methane release as the driver of Early Miocene positive $\delta^{13}\text{C}_{\text{carb}}$ excursion

394 Methane generation produces significant quantities of CH_4 highly enriched in ^{12}C

395 alongside CO_2 enriched in ^{13}C . The subsequent escape of ^{12}C -enriched CH_4 and the

396 incorporation of ^{13}C -enriched CO_2 into the DIC pool drive $\delta^{13}\text{C}_{\text{carb}}$ enrichment in DIC-

397 derived carbonates. Based on integrated geochemical evidence, the extreme positive

398 excursion of $\delta^{13}\text{C}_{\text{carb}}$ in the Dingqinghu Formation likely resulted from substantial

399 kinetic isotope fractionation during methanogenesis. This process facilitated the

400 preferential removal of ^{12}C as CH_4 , leaving the DIC pool enriched in $\delta^{13}\text{C}_{\text{carb}}$. Similar

401 mechanisms are documented in organic-rich lacustrine systems. Examples include Tilo

402 and Lake Bosumtwi, where methanogenesis elevated carbonate $\delta^{13}\text{C}$ values to

403 approximately +12‰ (Talbot and Kelts, 1986; Lamb et al., 2007; Rosqvist et al., 2007).

404 Methanogenesis in Lake Apopka generated pore water $\delta^{13}\text{C}$ values up to +26.4‰ (Gu

405 et al., 2004), while Lake Dziani Dzaha recorded average carbonate $\delta^{13}\text{C}$ values of +16.2

406 ± 1.1 ‰ (Cadeau et al., 2020). In Lake Caohai, it increased lacustrine carbonate $\delta^{13}\text{C}$ to

407 +20.94‰ (Zhu et al., 2013).

408 Unlike marine sediments dominated by CO_2 -reducing methanogenesis (Eq. (R1)),

409 the organic-rich, reducing Lunpola paleolake during Dingqinghu Formation deposition

410 provided an optimal environment for bacterial fermentation. Here acetoclastic

411 methanogenesis (Eq. (R2)) prevailed as the primary methane production pathway

412 (Whitticar et al., 1986). Critically, methane fate diverges significantly between marine

413 and lacustrine systems. In marine setting, methane is largely consumed at the Sulfate-

414 Methane Transition Zone (SMTZ) via syntrophy between sulfate-reducing bacteria and

415 anaerobic methanotrophic archaea, with additional aerobic oxidation minimizing

416 atmospheric release (Boetius et al., 2000; Deutzmann and Schink, 2011; Mostovaya et

417 al., 2022). Conversely, thinner oxic layers in lakes permit substantial methane bypass

418 of oxidation, leading to significant atmospheric emissions (Sun, 2024).

419 $\text{CO}_2 + 4\text{H}_2 \rightarrow \text{CH}_4 + 2\text{H}_2\text{O}$, (R1)

420 $\text{CH}_3\text{COOH} \rightarrow \text{CH}_4 + \text{CO}_2$, (R2)



421 The carbon isotope fractionation between carbonate and organic matter
 422 ($\Delta^{13}\text{C} = \delta^{13}\text{C}_{\text{carb}} - \delta^{13}\text{C}_{\text{org}}$) serves as a valuable proxy for assessing microbial
 423 contributions to sedimentary organic carbon. As established in prior studies (Hayes et
 424 al., 1999; Teranes and Bernasconi, 2005; Zhu et al., 2013), $\Delta^{13}\text{C}$ approximates ϵTOC ,
 425 the isotopic fractionation factor during organic matter synthesis. Significantly, ϵTOC
 426 value $>32\text{‰}$ indicates a dominant bacterial contribution to sedimentary organic matter,
 427 while values between 28‰ and 32‰ suggest a mixed phytoplankton-bacterial source
 428 (Hayes et al., 1999; Teranes and Bernasconi, 2005; Zhu et al., 2013). Analysis of the
 429 Dingqinghu Formation reveals that intervals characterized by enriched $\delta\text{C}_{\text{carb}}$ values
 430 predominantly exhibit $\Delta^{13}\text{C}$ values exceeding 32‰ (Fig. 3). This pronounced
 431 fractionation strongly implicates bacterial fermentation and decomposition as the
 432 primary processes driving carbon isotope dynamics during these stages. Concurrently
 433 depleted TOC values align with the expected signature of intense bacterial organic
 434 matter remineralization, further supporting the inference of methane release associated
 435 with these microbial processes.

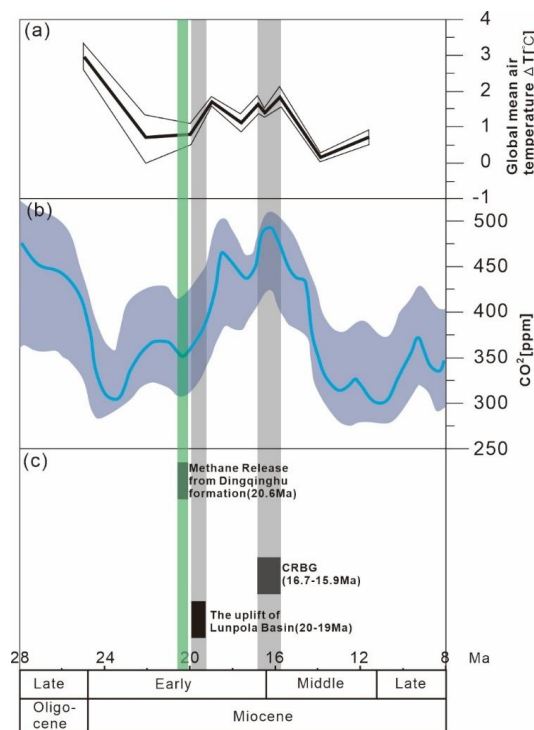
436 5.3 Lacustrine methane emissions response to Early Miocene warming

437 The Early Miocene was a pivotal period for climatic change in Asia, primarily driven
 438 by the extensive uplift of the Tibetan Plateau and the retreat of the Paratethys Ocean.
 439 These processes intensified the Asian monsoon system (Ramstein et al., 1997; Liu and
 440 Yin, 2002; Zhang et al., 2007; Boos & Kuang, 2010; Clift & Webb, 2019; Spicer et al.,
 441 2021), altering atmospheric circulation and thermal gradients, which in turn led to
 442 significant climatic differentiation across the Tibetan Plateau and adjacent areas. The
 443 plateau itself is influenced by both the Indian monsoon and continental monsoon
 444 systems, resulting in a range of climate regimes from warm and humid to dry and cold
 445 (Kutzbach et al., 1993; Guo et al., 2008; Shukla et al., 2014).

446 Studies by Deng et al. (2019) indicate that during the Miocene, the Indian monsoon
 447 dominated the Lunpola Basin, supplying ample moisture and warmth to the region. This
 448 interpretation is supported by the presence of subtropical fish fossils in the stratigraphic



449 record, along with previously documented mammalian fossils, pollen, and spores (Deng
450 et al., 2012; Sun et al., 2014; Jia et al., 2015; Jiang et al., 2018). The monsoon-driven
451 warm and humid conditions fostered high productivity of organic matter within the lake
452 ecosystem, while also promoting the decomposition of organic material by
453 methanogens, leading to substantial methane production (Deng et al., 2019; Su et al.,
454 2019).



455
456 Figure 6:(a)The difference between the global average temperature and today's temperature
457 simulation(Kürschner et al.,2008); (b) Atmospheric CO_2 estimates (symbols) and 500-kyr mean
458 statistical reconstructions (95% credible intervals in dark-blue shading, respectively) (Cenozoic CO_2
459 Proxy Integration Project (Cen CO_2 PIP) Consortium,2023); (c) Main geological events of the Miocene,
460 From right to left in order: 1) Methane release from Dingqinghu formation (this study) 2) The uplift of
461 Lunpola basin (Li et al.,2024) 3) CRBG: Columbia River Basalt Group, the largest terrestrial volcanic
462 event in North America(Kasbohm et al.,2018).

463 Geochemical records from the Dingqinghu Formation in the Lunpola Basin reveal



464 severely deficient sulfur content (as indicated by whole-rock S data; see Table S2) along
 465 with exceptionally low pyrite abundance, suggesting that sulfate concentrations in the
 466 paleo-lake were minimal during deposition. This sulfate-limited environment likely
 467 suppressed sulfate-driven anaerobic oxidation of methane (SD-AOM). In addition, a
 468 reduced oxidation zone further decreased CH₄ consumption, resulting in significant
 469 methane efflux into the atmosphere (Fig. 5; Taylor and Macquaker, 2011; Rickard, 2021;
 470 Lin et al., 2023).

471 As a potent greenhouse gas, with a global warming potential 28–34 times that of CO₂
 472 over a 100-year period, methane release from lakes could have contributed substantially
 473 to global warming (Forster et al., 2008; Sun et al., 2020). The only known Large
 474 Igneous Province (LIP) eruption during the Miocene was the Columbia River Basalt
 475 Group, which occurred between approximately 16.7 and 15.9 Ma. This event
 476 significantly increased global temperatures and atmospheric CO₂ levels (Fig. 6c;
 477 Kasbohm et al., 2018). However, according to Li et al. (2024), major uplift of the
 478 Lunpola Basin occurred between 20 and 19 Ma, after the methane release event
 479 recorded in the Dingqinghu Formation. This temporal discordance indicates that
 480 methane release was not triggered by tectonic uplift (Fig. 6). Furthermore, no
 481 contemporary volcanic activity was associated with this methane emission, suggesting
 482 that climatic and lacustrine thermal conditions were the primary controls.

483 Global temperature reconstructions for the Early Miocene indicate a warming trend,
 484 accompanied by methane release and rising atmospheric CO₂ levels, which suggests a
 485 methane-driven warming event (Fig. 6; Kürschner et al., 2008). In the absence of large
 486 igneous province volcanism, this increase in CO₂ can likely attributed to two main
 487 processes: (1) the atmospheric oxidation of methane released from the lacustrine
 488 deposits, such as those of the Dingqinghu Formation identified in the present study, and
 489 (2) additional CO₂ emissions from climate feedback mechanisms amplified by the
 490 initial greenhouse warming (Fig. 6; Cenozoic CO₂ Proxy Integration Project
 491 (CenCO₂PIP) Consortium, 2023). Consequently, methane release from the Tibetan



492 Plateau may have played an important role in Early Miocene global warming and the
 493 concurrent rise in atmospheric CO₂ levels.

494 As discussed in the preceding sections, a positive feedback mechanism exists
 495 between methane release in the Dingqinghu Formation and global climate warming
 496 during the Miocene. As temperature rose, lake stratification and anoxia intensified,
 497 stimulating microbial methanogenesis and methane ebullition. This released methane
 498 was subsequently oxidized to CO₂, further amplifying the greenhouse effect and
 499 reinforcing the warming cycle. The Dingqinghu Formation represents a deep-water,
 500 anoxic, organic-rich, and semi-restricted lacustrine system with limited external DIC
 501 input. To quantify methane ebullition during intervals of pronounced positive carbon
 502 isotope excursions in lacustrine sediments, we applied a closed-system Rayleigh
 503 fractionation model (Eq. (2)). This method enables estimation of methane emission
 504 rates and evaluation of their contribution to atmospheric CO₂, offering direct numerical
 505 support for the proposed positive feedback mechanism.

506 The Rayleigh distillation equation is expressed as (Höhener and Atteia, 2014; Miller
 507 et al., 2018; Li et al., 2024):

$$508 \quad \delta^{13}C_{carb} = \delta^{13}C_{initial} + \varepsilon \cdot \ln(f), \quad (2)$$

509 where $\delta^{13}C_{carb}$ is the measured carbonate carbon isotope composition (‰ VPDB),
 510 $\delta^{13}C_{initial}$ is the initial dissolved inorganic carbon isotope composition (‰, typical
 511 lacustrine background) (Lengs and Marshall, 2004); ε is the fractionation factor (‰, typical
 512 for acetoclastic methanogenesis) (Penning et al., 2006; Valentine et al., 2004; Meister et
 513 al., 2019); and f is the fraction of residual DIC.

514 The fraction of methane lost from the system is then:

$$515 \quad 1 - f = 1 - \exp\left(\frac{\delta^{13}C_{carb} + 2}{-20}\right), \quad (3)$$

516 Methane ebullition flux (g CH₄ m⁻² kyr⁻¹) is calculated as:

$$517 \quad F_{CH_4} = (1 - f) \times OCBR \times \frac{16}{12}, \quad (4)$$

518 where OCBR is the measured organic carbon burial rate (g C cm⁻² kyr⁻¹). Fluxes are
 519 converted to g CH₄ m⁻² yr⁻¹ by division by 1000. The full 62-sample dataset with CH₄



520 flux is provided in Supplementary Table S4.

521 Our calculations show that the paleo-Dingqinghu Lake maintained an average
522 methane flux of $34.5 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$ (peaking at $151.2 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$). After accounting
523 for 30–50% methane loss during atmospheric escape and oxidation, these ebullition
524 events contributed an estimated 3.9–6.6 ppm of atmospheric CO_2 throughout the
525 studied depositional interval. These values provide quantitative evidence that Miocene
526 global warming enhanced lake stratification and anoxia, accelerating organic matter
527 decomposition and methane release. The subsequent oxidation of this methane supplied
528 additional CO_2 to the atmosphere, further strengthening greenhouse conditions and
529 closing a powerful feedback loop. Therefore, paleo-Dingqinghu Lake serves as a clear
530 example of how continental lacustrine systems on the Tibetan Plateau acted as
531 significant amplifiers of global warming during past greenhouse climates, even though
532 their role had been previously underestimated.

533 **6 Conclusions**

534 Our finds establish that the pronounced positive carbonate carbon isotope excursion
535 ($\delta^{13}\text{C}_{\text{carb}}$ up to $+13.79\text{‰}$) recorded in the Dingqinghu Formation, Lunpola Basin,
536 originated primarily from large-scale methanogenesis and subsequent methane release.
537 Favorable paleoclimate and tectonic conditions promoted organic matter accumulation
538 and methanogenesis, while low sulfate concentrations limited anaerobic methane
539 oxidation, facilitating substantial methane escape to the atmosphere. The observed
540 coupling between this methane release and Early Miocene global atmospheric $p\text{CO}_2$
541 and temperature rises implicates that Tibetan Plateau methane emissions as a potentially
542 important contributor to in Early Miocene global warming and the coeval increase in
543 atmospheric CO_2 .

544 **Data availability**

545 All raw data can be provided by the corresponding authors upon request.



546 **Author contributions**

547 CY, XF, and HW: study design, data production, analysis, data interpretation, writing.
 548 CY, JD, YW, and TW: sample handling. CY: stable isotope and organic carbon content
 549 experiment. CY, XF, and SZ: calculation of methane flux.

550 **Competing interests**

551 The contact author has declared that none of the authors has any competing interests.

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