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1 **Paleoenvironmental changes in the northern South China Sea over the**
2 **past 28,000 years: a study of TEX₈₆-derived sea surface temperatures and**
3 **terrestrial biomarkers**

4
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18 Keywords: South China Sea, TEX₈₆, U_{37}^K , paleotemperature, biomarkers, terrestrial
19 organic matter, Late Quaternary

20
21 Abstract: We have generated a record of TEX₈₆(TEX₈₆^H)-derived sea surface temperatures
22 (SSTs) over the last 28 ka for core MD97-2146 from the northern South China Sea (SCS).
23 The TEX₈₆^H-derived temperature of a core-top sample corresponds to the SSTs in warmer
24 seasons. The SST record shows a drop during the Oldest Dryas period, an abrupt rise at the

25 onset of the Bølling-Allerød period, a plateau across the Younger Dryas period, and an
26 abrupt rise at the beginning of the Holocene. The glacial–interglacial contrast in
27 TEX₈₆^H-derived temperature is almost the same as that in foraminiferal Mg/Ca
28 ratio-derived temperature, but it is larger than those in U_{37}^K and transfer function-derived
29 temperatures. Possible interpretations are 1) the seasonal shift of glycerol dialkyl glycerol
30 tetraether (GDGT) production, 2) the overestimation of temperature change by TEX₈₆^H,
31 and 3) the underestimation of temperature change by U_{37}^K and transfer function approaches.
32 The similar variation in TEX₈₆^H-derived temperature at the study site and Chinese
33 stalagmite $\delta^{18}\text{O}$ during the last deglaciation suggests that changes in TEX₈₆^H-derived
34 temperature in this period reflected atmospheric and oceanic reorganization on a millennial
35 timescale. The long-chain n-alkanes are mainly of higher plant origin before ~14 ka and a
36 mixture of higher plant and lithic origins after ~14 ka; the abundance ratio of long-chain to
37 short-chain n-fatty acids decreases at ~15 ka, suggesting a drastic change in sediment
38 sources at ~14–15 ka. We attribute the higher content of fresh higher plant n-alkanes and
39 long-chain n-fatty acids before ~14–15 ka to enhanced aeolian transportation and/or arid
40 environments. Increased precipitation likely due to intensified summer monsoon after
41 ~14–15 ka enhanced the erosion of sedimentary rocks and increased the contribution of
42 lithic n-alkanes.

43

44 **1. Introduction**

45 Numerous paleoceanographic studies of sea surface temperature (SST) have been
46 undertaken in the South China Sea (SCS). Wang and Wang (1990) and Wang et al. (1995)
47 generated summer and winter SST records for the SCS, based on foraminifer assemblages,
48 and found that during the last glacial maximum (LGM) the SCS experienced larger

49 seasonal SST differences and a steeper latitudinal SST gradient than it does presently.
50 Similar phenomena were reported using foraminifer- and alkenone-based SST records for
51 the northern SCS (Huang et al., 1997a, b; Chen and Huang, 1998; Pelejero et al., 1999a;
52 Chen et al., 2003). Kienast et al. (2001) identified a millennium-scale temperature variation
53 that mimics Greenland ice core records. Recently, Oppo and Sun (2005) and Zhao et al.
54 (2006) reported millennium-scale temperature records from the northern and southern SCS,
55 respectively, for the time since the penultimate glacial interval. The glacial–interglacial
56 changes of SST in the SCS have been attributed either to the inflow of cold water from the
57 North Pacific (e.g., Wang and Wang, 1990; Wang et al., 1995) or to changes in winter
58 monsoon intensity (e.g., Huang et al., 1997a, b). Disagreements of paleotemperatures in the
59 SCS among different proxies such as alkenone U_{37}^K , the foraminiferal Mg/Ca ratio, and
60 transfer function are potentially attributable to differences in the season and depth that each
61 proxy reflects (e.g., Steinke et al., 2008). Alternative approaches are useful for better
62 understanding paleotemperature changes in the SCS.

63 TEX_{86} is a recently developed paleotemperature proxy (Schouten et al., 2002), which is
64 based on glycerol dialkyl glycerol tetraethers (GDGTs). The TEX_{86} paleothermometer has
65 the advantage that it does not seem to be influenced by changes in salinity (Wuchter et
66 al., 2004) and is more sensitive to temperature changes in tropical waters (Kim et al.,
67 2010). However, caution is still required when applying TEX_{86} because a water-column
68 study suggested that TEX_{86} recorded not only temperature changes, but also changes in
69 archaeal ecology, nutrient concentrations, and possibly oceanographic conditions (Turich
70 et al., 2007). More case studies are necessary to test the reliability of this proxy.
71 Furthermore, TEX_{86} has not been previously applied to SCS sediments.

72 Changes in terrestrial vegetation and sediment transport pathways have also been

73 investigated from the perspective of monsoon variation. Sun and Li (1999) reported a
74 transition from a herb-dominated cool and dry climate during the last glacial to a warm and
75 humid climate in the Holocene in south China, as shown by pollen and spore assemblages
76 in the SONNE 17940 core from the northern SCS. Wang et al. (1999a) attributed the
77 increase in clay content observed at ~14.5 ka in core 17940 to an increase in fluvial supply
78 from the Pearl River due to the intensification of the summer monsoon. Pelejero (2003)
79 revealed the parallel glacial–interglacial variability of terrestrial n-alkane concentration in
80 four different cores retrieved from the northern, western and southern SCS, taking values
81 linearly inversely correlated to the $U_{37}^{K'}$ -derived SST, with higher concentrations during
82 glacials. This oscillation was attributed to the emergence and flooding of the shelves caused
83 by sea level variations, together with SCS SSTs, have a clear dependency on the Northern
84 Hemisphere climate evolution. Terrestrial biomarkers such as long-chain n-alkanes and
85 long-chain n-fatty acids are useful for understanding the contribution and provenance of
86 terrestrial material (e.g., Pelejero et al., 1999b; Pelejero, 2003; Yamamoto and Polyak,
87 2009).

88 This study presents records of TEX_{86}^H -derived SSTs and terrestrial biomarkers for the
89 last 28,000 years from northern SCS core MD97-2146, located offshore from southern
90 China. The objective of this work is to understand changes in SSTs derived from TEX_{86}^H as
91 well as changes in sediment provenance. This is the first report of the application of TEX_{86}
92 (TEX_{86}^H) paleothermometry in the SCS.

93

94 **2. Oceanographic settings**

95 The SCS is a marginal sea of the North Pacific with seven connections to surrounding
96 seas and oceans (Fig. 1): the Taiwan Strait to the East China Sea (sill depth ~70 m), the

97 Bashi Strait to the North Pacific (sill depth ~2500 m), the Mindoro and Balabac Straits to
98 the Sulu Sea (sill depths ~450 and ~100 m, respectively), the Malacca Strait to the Indian
99 Ocean (sill depth ~30 m), and the Gaspar and Karimata Straits (~40-50 m) to the Java Sea
100 (Wyrтки, 1961). Surface circulation in the SCS is driven by large-scale, seasonally-reversed
101 monsoon winds (Wyrтки, 1961). In the boreal summer, southwesterly winds drive an inflow
102 of Indian Ocean water through the Sunda Shelf and a clockwise surface circulation in the
103 SCS. In the boreal winter, northeasterly winds drive an inflow of North Pacific and East
104 China Sea waters through the Bashi and Taiwan Straits, and surface circulation in the SCS
105 is counterclockwise.

106

107 **3. Materials and Methods**

108 *3.1. Samples and age-depth model*

109 During the IMAGES 1997 *Marion Dufresne* cruise, a giant piston core (MD97-2146;
110 38.69 m long) was collected from a water depth of 1720 m on the northern slope of the
111 SCS at 20°07.08'N, 117°23.02'E (Fig. 1). The sediment retrieved consisted of dark gray
112 nannofossil and foraminifer oozes with some radiolarians and diatoms (Chen et al., 1998).

113 An age model in calendar years was created from the AMS ¹⁴C ages of seven samples
114 of the planktonic foraminiferan *Globigerinoides sacculifer* (Lin et al., 2006) and six
115 samples of mixed planktonic foraminifera *Globigerinoides ruber* and *G. sacculifer*
116 (Shintani et al., 2008). The calendar age was converted using the CALIB5.0 program and
117 marine04.14C dataset (Reimer et al., 2004) with a 400-year global reservoir correction
118 (Shintani et al., 2008). Lin et al. (2006) created an age model using a combination of ¹⁴C
119 ages and stratigraphic correlation with nearby core 17940 (Wang et al., 1999b). The present
120 work does not use any stratigraphic correlation, however, because the assumption that

121 temperatures in core 17940 changed synchronously with Greenland temperatures (Wang et
122 al., 1999b) is not necessary.

123 A total of 76 samples were collected every 20 cm on average (equivalent to
124 approximately 300-year intervals) down to a depth of 19 m (0-28 ka).

125

126 3.2 Hydrocarbon analysis

127 Lipids were extracted (x 2) from 1 g of dried sediment using a DIONEX Accelerated
128 Solvent Extractor ASE-200 at 100°C and 1000 psi for 10 min with 11 ml of
129 CH₂Cl₂-CH₃OH (6:4) and then concentrated. The lipid extract was separated into four
130 fractions using column chromatography (SiO₂ with 5% distilled water; i.d., 5.5 mm; length,
131 45 mm): F1 (hydrocarbons), 3ml hexane; F2 (aromatic hydrocarbons), 3 ml hexane-toluene
132 (3:1); F3 (ketones), 4 ml toluene; F4 (polar compounds), 3 ml toluene-CH₃OH (3:1);
133 *n*-C₂₄D₅₀ and *n*-C₃₆H₇₄ were added as internal standards to F1 and F3, respectively.

134 An aliquot of F4 was trans-esterified with 1 ml 5% HCl-CH₃OH at 60°C for 12 h
135 under N₂. The esterified lipids were supplemented with 2 ml distilled water and extracted
136 (x 3) with toluene. The extract was back-washed (x 3) with distilled water, passed through
137 a short bed of Na₂SO₄, and separated into two fractions with SiO₂ column chromatography:
138 F4-1 (acids), 4 ml toluene; F4-2 (alcohols), 3 ml toluene-CH₃OH (3:1); *n*-C₂₄D₅₀ was
139 added as an internal standard to F4-1.

140 Gas chromatography (GC) was conducted using a Hewlett Packard 5890 series II gas
141 chromatograph with on-column injection and electronic pressure control systems, and a
142 flame ionization detector (FID). Samples were dissolved in hexane. Helium was the carrier
143 gas and the flow velocity was maintained at 30 cm/s. A Chrompack CP-Sil5CB column
144 was used (length, 60 m; i.d., 0.25 mm; thickness, 0.25 μm). The oven temperature was

145 programmed to rise from 70 to 130°C at 20°C/min, from 130 to 310°C at 4°C/min, and to
146 hold at 310°C for > 30 min. The standard deviations of five duplicate analyses averaged
147 7.5% of the concentration for each compound.

148 Gas chromatography-mass spectrometry (GC-MS) of F1, F3 and F4-1 was conducted
149 using a Hewlett Packard 5973 GC-mass selective detector with on-column injection and
150 electronic pressure control systems, and a quadrupole mass spectrometer. The GC column
151 and oven temperature and carrier pressure programs were as described above. The mass
152 spectrometer was run in full scan mode (m/z 50–650). Electron ionization (EI) spectra were
153 obtained at 70 eV. Compound identification was achieved by comparing mass spectra and
154 retention times with those of standards and published data.

155

156 3.3 Glycerol dialkyl glycerol tetraether (GDGT) analysis

157 An aliquot of F4-2 was dissolved in hexane–2-propanol (99:1) and filtered. Glycerol
158 dialkyl glycerol tetraethers (GDGTs) were analyzed using high performance liquid
159 chromatography-mass spectrometry (HPLC-MS) with an Agilent 1100 HPLC system
160 connected to a Bruker Daltonics micrOTOF-HS time-of-flight mass spectrometer.
161 Separation was conducted using a Prevail Cyano column (2.1 x 150 mm, 3µm; Alltech) and
162 maintained at 30°C following the method of Hopmans et al. (2000) and Schouten et al.
163 (2007). Conditions were: flow rate 0.2 ml/min, isocratic with 99% hexane and 1%
164 2-propanol for the first 5 min followed by a linear gradient to 1.8% 2-propanol over 45 min.
165 Detection was achieved using atmospheric pressure, positive ion chemical ionization-mass
166 spectrometry (APCI-MS). The spectrometer was run in full scan mode (m/z 500–1500).
167 Compounds were identified by comparing mass spectra and retention times with those of
168 GDGT standards (formed from the main phospholipids of *Thermoplasma acidophilum* via

169 acid hydrolysis) and those in the literature (Hopmans et al., 2000). Quantification was
170 achieved by integrating the summed peak areas in the (M+H)⁺ and the isotopic (M+H+1)⁺
171 ion traces.

172 TEX_{86}^H was calculated from the concentrations of GDGT-1, GDGT-2, GDGT-3 and a
173 regioisomer of crenarchaeol using the following expression (Schouten et al., 2002; Kim et
174 al., 2010):

175

$$176 \quad TEX_{86}^H = \log TEX_{86} = \log \frac{([\text{GDGT-2}] + [\text{GDGT-3}] + [\text{Crenarchaeol regioisomer}])}{([\text{GDGT-1}] + [\text{GDGT-2}] + [\text{GDGT-3}] + [\text{Crenarchaeol regioisomer}])}$$

178

179 TEX_{86}^H is defined as the logarithmic function of TEX_{86} and yields the best correlation with
180 SST when the data from polar and subpolar oceans are removed (Kim et al., 2010).

181 Temperature was calculated according to the following equation based on a global core-top
182 calibration (Kim et al., 2010):

183

$$184 \quad T = 68.4 \times TEX_{86}^H + 38.6$$

185

186 where T = temperature [°C]; analytical accuracy was 0.45°C in our laboratory.

187

188 **4. Results**

189 *4.1. TEX_{86}^H*

190 The TEX_{86}^H -derived temperature profile indicates that between 18 and 28 ka SSTs were
191 nearly constant at 23.3°C, decreased to 20.8°C at 16.7 ka, and then increased abruptly to
192 25.8°C by 14.5 ka. After 14.5 ka SSTs increased gradually to 26.3°C by 12.9 ka, and then

193 decreased to 25.4°C at 12.3 ka, before increasing abruptly to 28.0°C by 11.0
194 ka SSTs increased gradually to 28.7°C by 5.3 ka and remained constant thereafter (Fig. 3A).
195 The core-top temperature was 28.1°C. This temperature is higher than the mean annual
196 SST (26.6°C; NOAA, 1998) and agrees with the SST in May and September-October at
197 this site (~28.0°C and ~27.7-28.6°C, respectively; NOAA, 1998).

198 The Branched and Isoprenoid Tetraether (BIT) index, a proxy for soil versus marine
199 organic matter input to sediments (Hopamans et al, 2004), was constantly low (< 0.03).

200

201 4.2. Normal alkanes

202 Normal alkanes (n-alkanes) occur as a major component of the F1 (hydrocarbon)
203 fraction and show a unimodal distribution with a maximum at C₃₁. The total concentration
204 of long-chain C₂₅-C₃₃ n-alkanes varies between 0.41 and 1.66 µg/g in sediment, with an
205 average of 0.86 µg/g (Fig. 4A). The odd carbon number preference index (CPI) values of
206 C₂₄-C₃₄ homologues (Bray and Evans, 1961) vary between 2.0 and 6.2, with an average of
207 3.4. The average chain length (ACL) values of C₂₇-C₃₃ homologues (Bray and Evans,
208 1961) vary between 29.1 and 30.3, with an average of 29.7.

209 Most samples showed the n-alkane distribution as a mixture of a pattern with high odd
210 carbon number preference (CPI > ~5; Fig. 4A) and a pattern heavily influenced by mature
211 (thermally altered) n-alkane with low CPI values (~2.5; Fig. 4A). The former is typical of
212 terrestrial higher plant waxes (Eglinton and Hamilton, 1967). This pattern indicates that the
213 OM was derived from fresh higher plant material. The latter is typical of coal and coaly
214 shale (Bray and Evans, 1961), indicating an influx of mature (lithic) OM.

215 If we assess the end-member CPI values of higher plant and mature OM, we can
216 estimate the concentrations of n-alkanes of both origins. Assuming that the long-chain

217 n-alkanes derived from higher plant waxes have CPI values of 6.2 (maximal value for
218 samples in the study core), and that those derived from mature rocks have CPI values of 1
219 (typical values of OM in catagenesis and metagenesis stages; Tissot and Welte, 1984), we
220 calculated concentrations of “higher plant” and “lithic” C₂₅-C₃₃ n-alkanes from the total
221 C₂₅-C₃₃ n-alkane concentration and the CPI value using the following formulae (Yamamoto
222 and Polyak, 2009):

223

224 Higher plant C₂₅-C₃₃ n-alkane

225 = total C₂₅-C₃₃ n-alkane × $\frac{\{(6.2+1)(CPI-1)\}}{\{(6.2-1)(CPI+1)\}}$

226 Lithic C₂₅-C₃₃ n-alkane = total C₂₅-C₃₃ n-alkane – higher plant C₂₅-C₃₃ n-alkane

227

228 The concentration of higher plant long-chain n-alkanes varies between 0.29 and 1.33
229 µg/g with an average of 0.64 µg/g. The concentration of lithic long-chain n-alkanes varies
230 between 0 and 0.55 µg/g with an average of 0.23 µg/g. Both total and higher plant
231 n-alkanes show a similar pattern over the last 30 kyr: they are higher between 14 and 30 ka
232 than between 2 and 14 ka, and increase to present-day values at around 2 ka (Fig. 4A). In
233 contrast, lithic n-alkanes remain low until 15 ka, increased at 15 ka, and then increase
234 gradually thereafter (Fig. 4A).

235

236 4.3. Normal fatty acids

237 Normal fatty acids (n-fatty acids) occur as a major component of the F4-1 (acid)
238 fraction and show a bimodal distribution with a maximum at C₁₆ and C₂₆ or C₂₈.
239 Short-chain n-fatty acids are ubiquitous in eukaryotes and bacteria, while long-chain n-fatty
240 acids are specific to higher plants (Kvenvolden et al., 1967). The even CPI values of

241 C₂₅-C₃₃ homologues (Kvenvolden et al., 1967) were nearly constant with a range of
242 between 3.6 and 4.8 and an average of 4.2 (Fig. 4B).

243 The total concentration of n-fatty acids (C₁₄-C₃₄) varies between 5.9 and 34.5 µg/g,
244 with an average of 10.7 µg/g (Fig. 4B). The concentration of short-chain n-fatty acids
245 (C₁₄-C₁₈) varies between 1.3 and 12.1 µg/g, with an average of 3.6 µg/g (Fig. 4B). The
246 concentration of long-chain n-fatty acids (C₂₆-C₃₄) varies between 2.3 and 13.4 µg/g, with
247 an average of 4.7 µg/g. The concentration of short-chain n-fatty acids was constant with the
248 exception of a peak at 17.3 ka, while the concentration of long-chain n-fatty acids shows
249 several peaks prior to 15 ka (Fig. 4B).

250

251 **5. Discussion**

252 *5.1. SST changes*

253 The U_{37}^K -derived temperature in this core was reported by Shintani et al. (2008).
254 Temperature records were calculated using the equations of Prahl et al. (1988). The SSTs
255 fluctuated slightly between 28–16 ka but maintained an average of 24.0°C. By 14.8 ka,
256 SSTs increased to 24.7°C, whereupon they remained constant until 10.7 ka. After 10.7 ka
257 SSTs increased to 26.5°C by 6.1 ka and then increased more gradually to reach 27°C at
258 present (Fig. 3B). This variation is similar to that recorded in nearby core 17940 (Pelejero
259 et al., 1999a). Present-day SST at the study site shows a seasonal variation between 23.7°C
260 in January and 28.9°C in July, with a mean annual value of 26.6°C (Fig. 2; NOAA, 1998).
261 The U_{37}^K -derived core-top temperature is 26.0°C. This temperature is slightly lower than
262 the mean annual SST at this site (26.6°C; NOAA, 1998). As indicated from satellite
263 observations, chlorophyll concentrations do not display large seasonal variations, with only
264 a moderate maximum in the winter season in the study area (Zhang et al., 2006). The

265 weighted-average SST (0–30 m) based on chlorophyll concentration is 25.7°C, which is
266 only 0.6°C lower than the mean annual temperature at 0–30 m (26.3°C). These
267 observations suggest that the U_{37}^K -derived temperature reflects the mean annual SST at the
268 study site.

269 The core-top temperature based on TEX_{86}^H is 28.1°C. This temperature is 1.5°C higher
270 than the mean annual SST (26.6°C; NOAA, 1998) and agrees with the SST in May and
271 September–October at this site (~28.0°C and ~27.7–28.6°C, respectively; NOAA, 1998). It
272 has been reported that the GDGT concentration in surface waters is higher during
273 non-blooming seasons in the North Sea and the Bermuda Rise (Wuchter et al., 2005). By
274 analogy with this case, although there is no information on the production season and depth
275 of GDGTs at the study site, it is reasonable to infer that GDGT are produced during the
276 summer season when blooms cease. We infer, therefore, that the TEX_{86}^H -derived
277 temperature reflects the SST weighted in warmer seasons at the study site.

278 The U_{37}^K -derived temperature shows a different temporal evolution in comparison with
279 TEX_{86}^H -derived temperatures (Figs. 3A and 3B). We interpret U_{37}^K -derived temperatures as
280 reflecting mean annual SST (Shintani et al., 2008). The average of summer and winter
281 SSTs estimated by a regional Imbrie–Kipp transfer function method (FP-12E; Thompson,
282 1981) of planktonic foraminifera in core 19740 (Wang et al., 1999b) agreed with
283 U_{37}^K -derived temperature in core MD97-2146 (Fig. 3B), supporting this interpretation.

284 On the other hand, we interpret the TEX_{86}^H -derived temperature estimate at a core top
285 sediment sample to reflect the SST weighted in warmer seasons. The TEX_{86}^H -derived
286 temperatures in core MD97-2146 range between summer and annual mean SSTs estimated
287 by a transfer function method in core 19740 (Wang et al., 1999b) during the Holocene,
288 whereas they agree with the annual mean SSTs during the last glacial period between 18

289 and 27 ka (Fig. 3A). There are three possible interpretations of this phenomenon, such as 1)
290 the seasonal shift of GDGT production, 2) the overestimation of temperature change by
291 $\text{TEX}_{86}^{\text{H}}$, and 3) the underestimation of temperature change by U_{37}^{K} and transfer function
292 approaches.

293 First, the seasonal shift of GDGT production from warmer seasons to cooler seasons is
294 one of the candidate mechanisms accounting for larger variation in $\text{TEX}_{86}^{\text{H}}$ -derived
295 temperature than in U_{37}^{K} - and transfer function-derived mean annual SSTs. The GDGT
296 concentration in surface waters has been reported to be higher during non-blooming
297 seasons in the North Sea and the Bermuda Rise (Wuchter et al., 2005). If the phytoplankton
298 blooms shifted to warmer seasons due to the temperature limitation of phytoplankton
299 growth, the production of GDGTs would decrease in warmer seasons, and $\text{TEX}_{86}^{\text{H}}$ should
300 reflect the SST in cooler seasons.

301 Second, the overestimation of temperature change by $\text{TEX}_{86}^{\text{H}}$ is possible due to
302 calibration at this stage. Global core top calibration was revised by increasing surface
303 sediment samples (Schouten et al., 2002; Kim et al., 2008; 2010). The most recently
304 revised calibration gives smaller temperature change than did previous versions. Kim et al.
305 (2010) also pointed out that crenachaeotal ecology is different between subpolar and
306 subtropical water masses and may have different responses to temperature change. If a
307 water mass is displaced by another water mass, a complex response to temperature change
308 results. Caution is necessary when discussing the degree of variation in TEX_{86} -derived
309 temperature until temperature calibration is practically established.

310 Last, possible underestimations of temperature change by U_{37}^{K} and the transfer
311 function methods are not negligible. Because the production of coccolithophores is
312 temperature dependant (Conte et al., 1998), the season of maximum production may have

313 shifted to the warmest times of year, during periods of decreasing mean annual SST. It is
314 thus possible that the drop in U_{37}^K -derived temperatures may be an underestimate.
315 Temperature at core 17940 was estimated by a regional Imbrie–Kipp method (Wang et al.,
316 1999b). Chen et al. (2005) compared the results of temperature estimates by different
317 transfer function techniques and found that the Imbrie–Kipp method tends to show smaller
318 temperature variation than those obtained by other methods. In addition, Steinke et al.
319 (2008) showed that transfer function methods give smaller glacial–interglacial variation in
320 SST compared with those inferred by geochemical proxies in the SCS because there are
321 no-modern analog planktonic foraminiferal faunas in the glacial SCS. These lines of
322 evidence suggest that glacial cooling may be underestimated by U_{37}^K and transfer function
323 methods.

324 It is now not clear which of the above mechanism is critical in causing the difference
325 between proxies. Further comparison studies of proxies are necessary to answer this
326 question.

327 The TEX_{86}^H -derived temperatures in core MD97-2146 generally agree with the
328 foraminiferal Mg/Ca-derived temperatures at ODP Site 1145, located ~100 km south of
329 Site MD97-2146 (Fig. 3; Oppo and Sun, 2005), but the disagreement is again observed in
330 the period between 18 ka and 15 ka (Figs. 3A and 3C). The cooling observed in
331 TEX_{86}^H -derived temperatures during the 15–18 ka could be somehow overestimated, as
332 other proxies do not indicate such a cooling. The anomalies of TEX_{86}^H -derived
333 temperatures are potentially attributable to seasonal shift of GDGT production, changes in
334 the GDGT production depth, lateral transport of GDGTs and additional contribution of
335 GDGTs by archaeal communities other than Marine Group I Crenarchaeota (Kim et al.,
336 2008). There is now no evidence to support the above possibilities, but they must be

337 considered by further studies.

338 The changing pattern of TEX₈₆^H-derived temperature record from 15 to 10 ka shows a
339 good correspondence to the millennial-scale pattern of stalagmite oxygen isotopes from the
340 Hulu Cave, eastern central China (Wang et al., 2001), and the Dongge Cave, south China
341 (Yuan et al., 2004). The variation in stalagmite δ¹⁸O is thought to reflect changes in the
342 relative abundance of summer vs. winter precipitation: low values correspond to larger
343 contributions of summer precipitation due to an increased intensity of the East Asian
344 summer monsoon. The correspondence of the pattern of change between TEX₈₆^H-derived
345 temperature at the study site and Chinese stalagmite δ¹⁸O suggests that changes in
346 TEX₈₆^H-derived temperature in this period reflected atmospheric and oceanic
347 reorganization on a millennial timescale.

348

349 *5.2. Inflow of terrestrial organic matter to the study site*

350 The total long-chain n-alkane concentration decreases in the interval from 15 ka to 13 ka
351 (Fig. 4). The similar trend of long-chain n-alkane concentration was previously reported in
352 nearby core 17940 (Pelejero, 2003; Kienast et al., 2010) and was attributed to the
353 emergence and flooding of the shelves caused by sea level variations, together with SCS
354 SSTs, have a clear dependency on the Northern Hemisphere climate evolution.

355 In this study, the long-chain n-alkanes were separately determined as higher plant-derived
356 n-alkanes and lithic n-alkanes. The long-chain n-alkanes are mainly of higher plant origin
357 before ~14–15 ka and a mixture of higher plant and lithic origins after ~14–15 ka (Fig. 4A).
358 The sudden decrease of higher plant n-alkane concentration was accompanied by a
359 decrease in the ratio of long-chain to short-chain n-fatty acids (Fig. 4B). These observations
360 suggest that the relative contribution of fresh higher plants decreased and that the

361 contribution of mature organic matter derived from sedimentary rocks increased between
362 14 and 15 ka, indicating a drastic change in sediment provenance at this time.

363 Wang et al. (1999a) showed that the median grain size of silt and the total modal grain
364 size of sediments decreased, while the clay content increased, at the onset of the
365 Bølling-Allerød (B-A) period at core site 17940. This decrease in the average grain size
366 and increase in clay content was interpreted as indicating a decrease in the supply of dust
367 and an increase in the supply of fine fluvial material from the Pearl River at ~14.5 ka.
368 Wang et al. (1999a) stressed that this change reflects the shift from a winter
369 monsoon-dominated glacial regime to a summer monsoon-dominated Holocene regime.

370 Tamburini et al. (2003) found higher illite content in SCS sediment during the Holocene,
371 while smectite and kaolinite were elevated during glacial periods. Because illite is
372 abundant in sediments derived from the Asian continent, and smectite and kaolinite are
373 relatively abundant in sediments from the southern tropical islands, the change in
374 clay-mineral composition may reflect changes in sediment provenance. Studies on modern
375 sediments indicate that reworking of material from the Pearl River (Wang et al., 1999a) and
376 current transport from the East China Sea through the open Taiwan Strait (Chen, 1978)
377 should be considered the main transport mechanism responsible for clay and mineral
378 distribution in the northern SCS (Tamburini et al., 2003). The low illite content during the
379 last glacial period was attributed to the interruption of the input of illite-rich material from
380 the East China Sea shelf when a low sea level stand closed the Taiwan Strait (Tamburini et
381 al., 2003).

382 The sill depth of the Taiwan Strait is ~70 m, and a marine connection was established by
383 ~13 ka, later than the decreases of fresh higher plant-derived n-alkane and n-fatty acids
384 (Fig. 4). This implies that the opening of the Taiwan Strait was not a cause of the low

385 content of higher plant biomarkers.

386 On the other hands, the decrease of fresh higher plant n-alkanes at 15 ka coincides with
387 the increase of the Asian summer monsoon intensity recorded in Chinese stalagmites
388 (Wang et al., 2001; Yuan et al., 2004). The greater content of fresh higher plant n-alkanes
389 and long-chain n-fatty acids in sediments prior to 14–15 ka is attributable to enhanced
390 aeolian transportation and/or arid environments in South China (Sun and Li, 1999).
391 Enhanced precipitation due to the intensified East Asian summer monsoon after ~14–15 ka
392 increased the erosion of sub-aerially exposed sedimentary rocks, increasing the relative
393 proportion of lithic n-alkanes.

394

395 5.3. *Paleoenvironment after 2 ka*

396 The ACL is significantly higher in the Oldest Dryas period and after 2 ka (Fig. 4). In
397 general, ACL values are higher in herbs than in trees (e.g., Vogts et al., 2009). A pollen
398 study at the site of core 17940, near our study site, showed that herb pollen (e.g., *Artemisia*,
399 *Gramineae*, and *Cyperaceae*) was abundant during the Oldest Dryas period, due to the cold
400 and dry climate, and decreased during the Bølling-Allerød period (Sun and Li, 1999). The
401 high ACL in the Oldest Dryas period is consistent with the high abundance of herb pollen.
402 Fern spores significantly increased after 1.4 ka in core 17940, possibly due to human
403 activity (Sun and Li, 1999). This increase corresponds to the increase of ACL in the study
404 core. There is one report for a fern *Hydrilla verticillate* having high ACL (Chikaraishi and
405 Naraoka, 2003), but we do not fully understand the ACL in ferns. Thus we cannot attribute
406 this change to the increase of ferns; future investigations of fern ACLs are necessary. The
407 core site is proximal to South China and is thus in an ideal position to trace environmental
408 changes by human activities in historical times. Further applications of new techniques will

409 contribute to better understanding the environmental changes in South China.

410

411 **Conclusions**

412 We applied $\text{TEX}_{86}^{\text{H}}$ paleothermometry to sediment from the SCS and demonstrated
413 measurable SST variations over the past 28 kyr. The $\text{TEX}_{86}^{\text{H}}$ -derived temperature of a
414 core-top sample corresponds to the SSTs in warmer seasons. The glacial–interglacial
415 contrast in $\text{TEX}_{86}^{\text{H}}$ -derived temperature is almost the same as that in foraminiferal Mg/Ca
416 ratio-derived temperature, but it is larger than those in U_{37}^{K} and transfer function-derived
417 temperatures. The similar variation in $\text{TEX}_{86}^{\text{H}}$ -derived temperature at the study site and
418 Chinese stalagmite $\delta^{18}\text{O}$ during the last deglaciation suggests that changes in
419 $\text{TEX}_{86}^{\text{H}}$ -derived temperature in this period reflected atmospheric and oceanic
420 reorganization on a millennial timescale.

421 We also examined terrestrial biomarkers such as n-alkanes and n-fatty acids that show a
422 remarkable change from abundant fresh higher plants to the mixture of fresh higher plants
423 and mature organic matter at 14–15 ky BP. We consider this is a result of a considerable
424 change in sediment delivery at that time. Terrestrial organic matter was presumably
425 supplied by enhanced aeolian transportation prior to 14–15 ka, with the contribution from
426 river discharge becoming more important thereafter presumably due to intensified summer
427 monsoon.

428

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436

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592 39–55.

593

594 **Figure captions**

595 Fig. 1. Map showing the locations of core MD97-2146 and other cores referred to in this
596 paper.

597

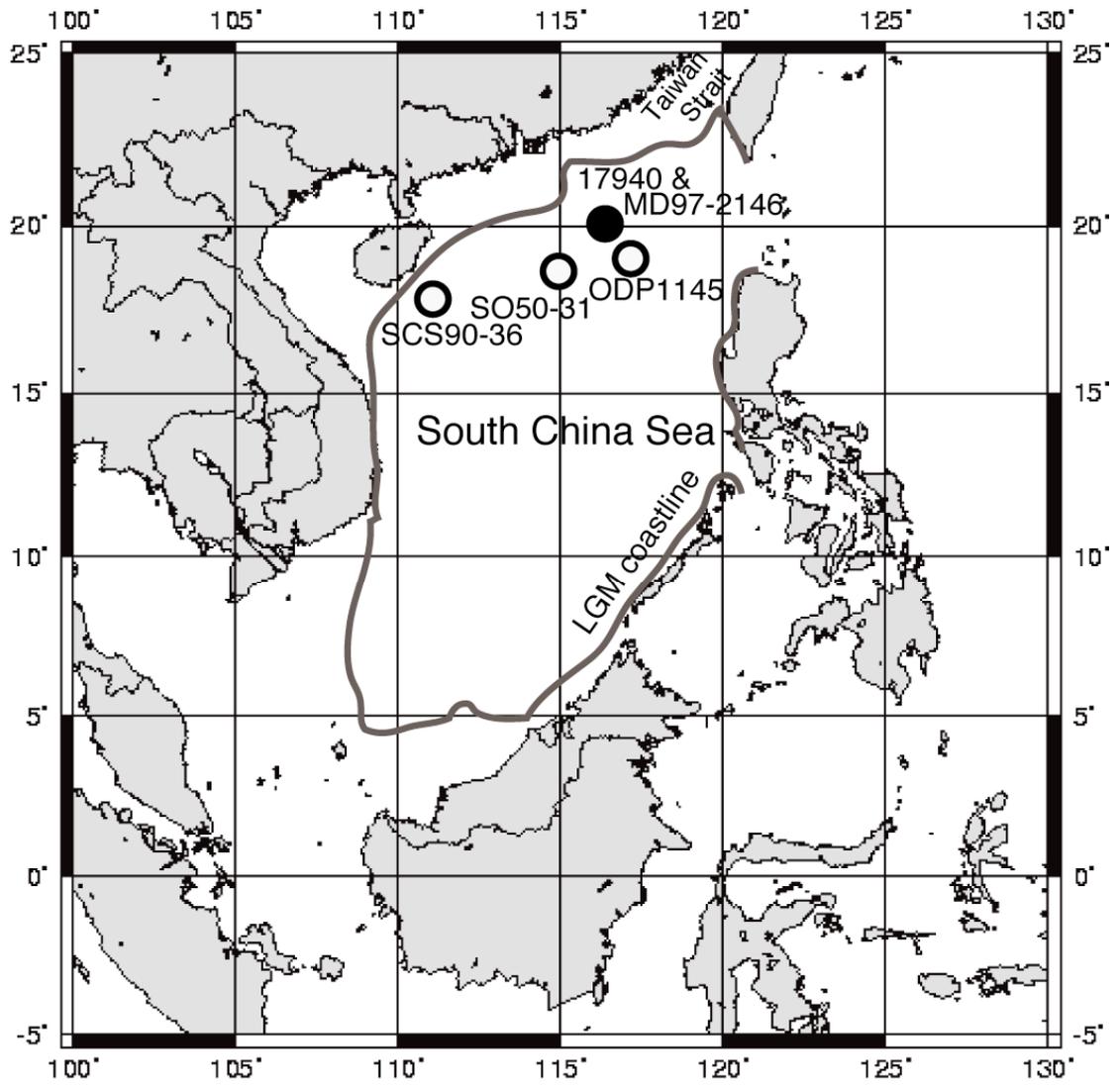
598 Fig. 2. Depth and seasonal variation of water temperatures at the study site (Compiled data
599 from NOAA, 1998). “J” to “D” means January to December.

600

601 Fig. 3. (A) $\text{TEX}_{86}^{\text{H}}$ and (B) U_{37}^{K} -derived temperatures in core MD97-2146, and (C)
602 Mg/Ca-derived SST in ODP Site 1145 (Oppo and Sun, 2005) during the last 28 kyr. The
603 summer, winter and annual mean SSTs estimated by a regional Imbrie–Kipp transfer
604 function method (FP-12E; Thompson, 1981) of planktonic foraminifera in core 19740
605 (Wang et al., 1999b) are also shown in each panel.

606

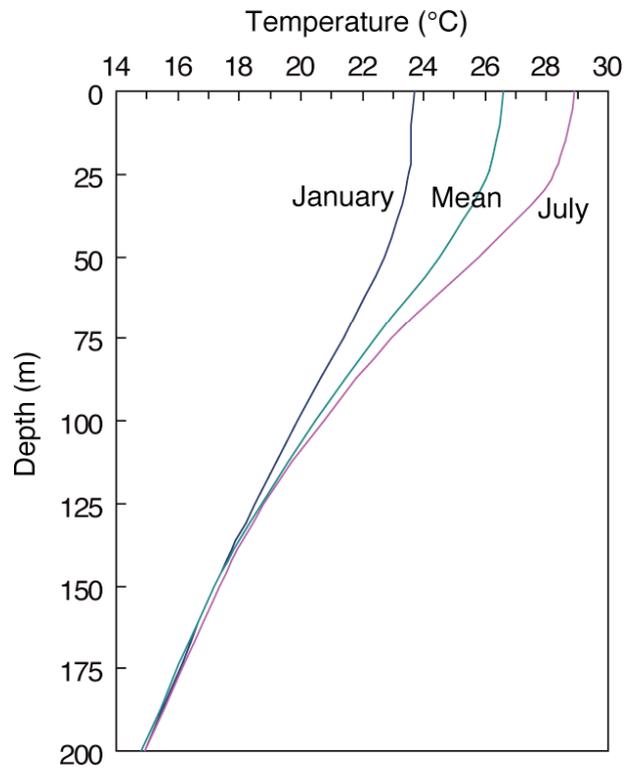
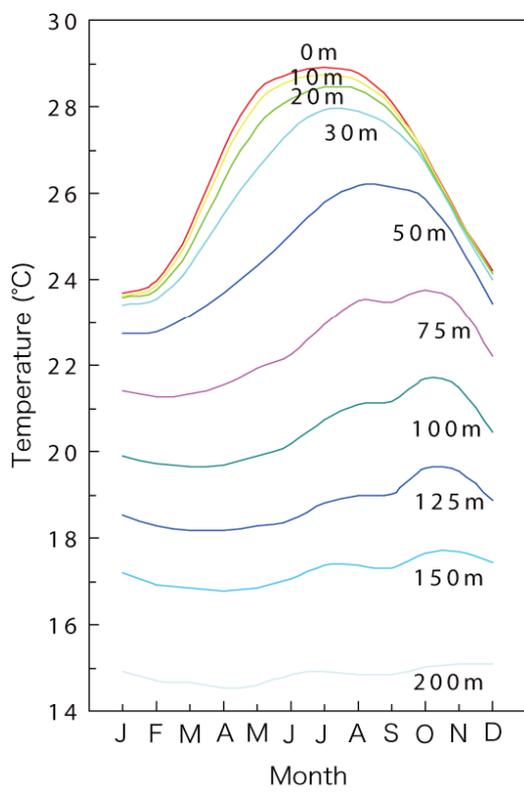
607 Fig. 4. (A) Concentrations of total, higher plant, and lithic long-chain $\text{C}_{25}\text{-C}_{33}$ n-alkanes,
608 and the CPI and ACL of n-alkanes. (B) Concentrations of total ($\text{C}_{14}\text{-C}_{34}$), short-chain
609 ($\text{C}_{14}\text{-C}_{18}$), and long-chain ($\text{C}_{26}\text{-C}_{34}$) n-fatty acids, and the CPI of n-fatty acids and
610 abundance ratio of long-chain to short-chain (L/S ratio) n-fatty acids in core MD97-2146
611 during the last 28 kyr. (C) Reconstructed sea level (Lambeck *et al.*, 2002) and the sill
612 depths of straits to the East China Sea, the Sulu Sea, the Indian Ocean and the Java Sea
613 (Wyrтки, 1961).



614

615 Fig. 1

616

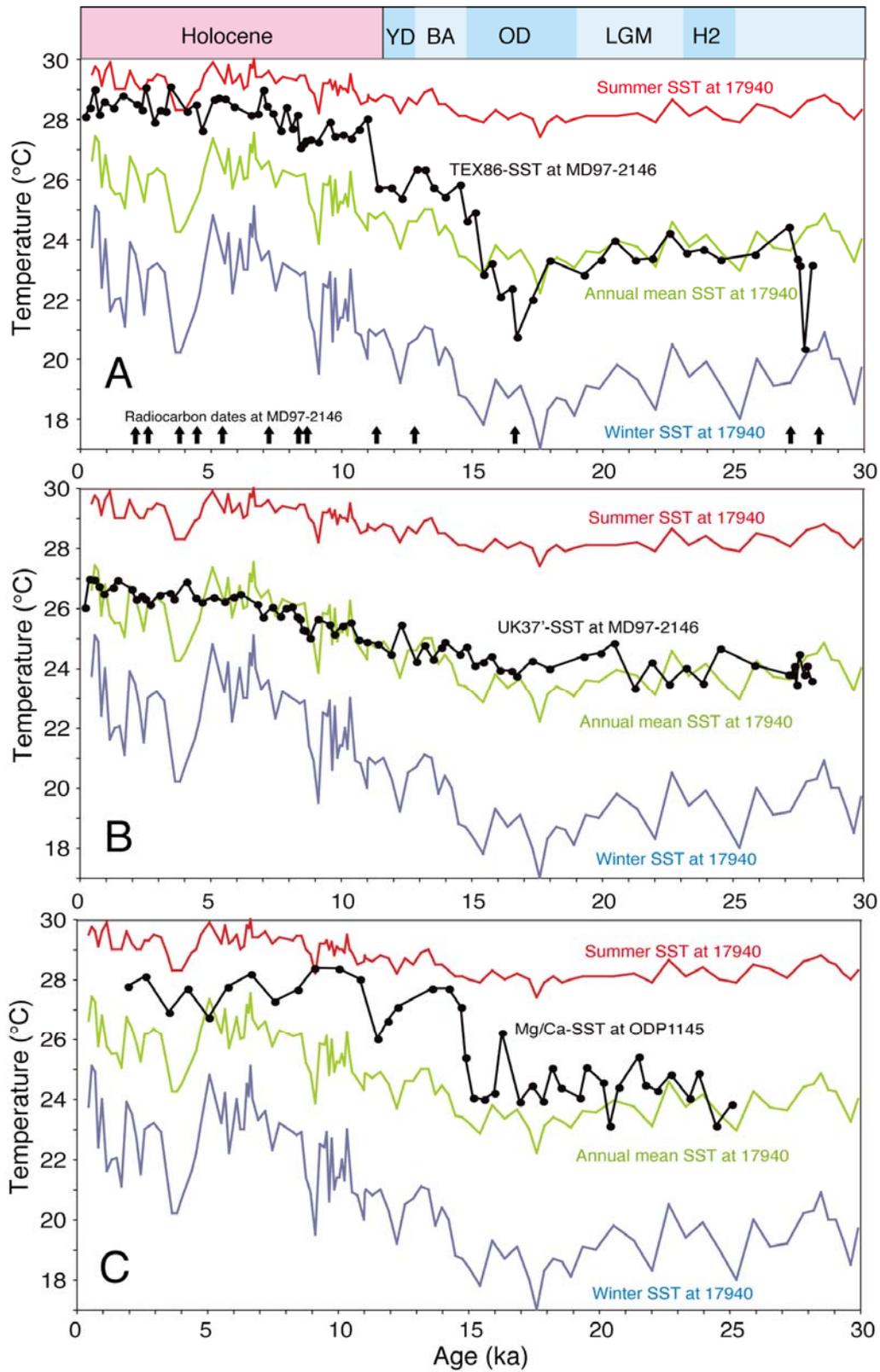


617

618

619 Fig. 2

620

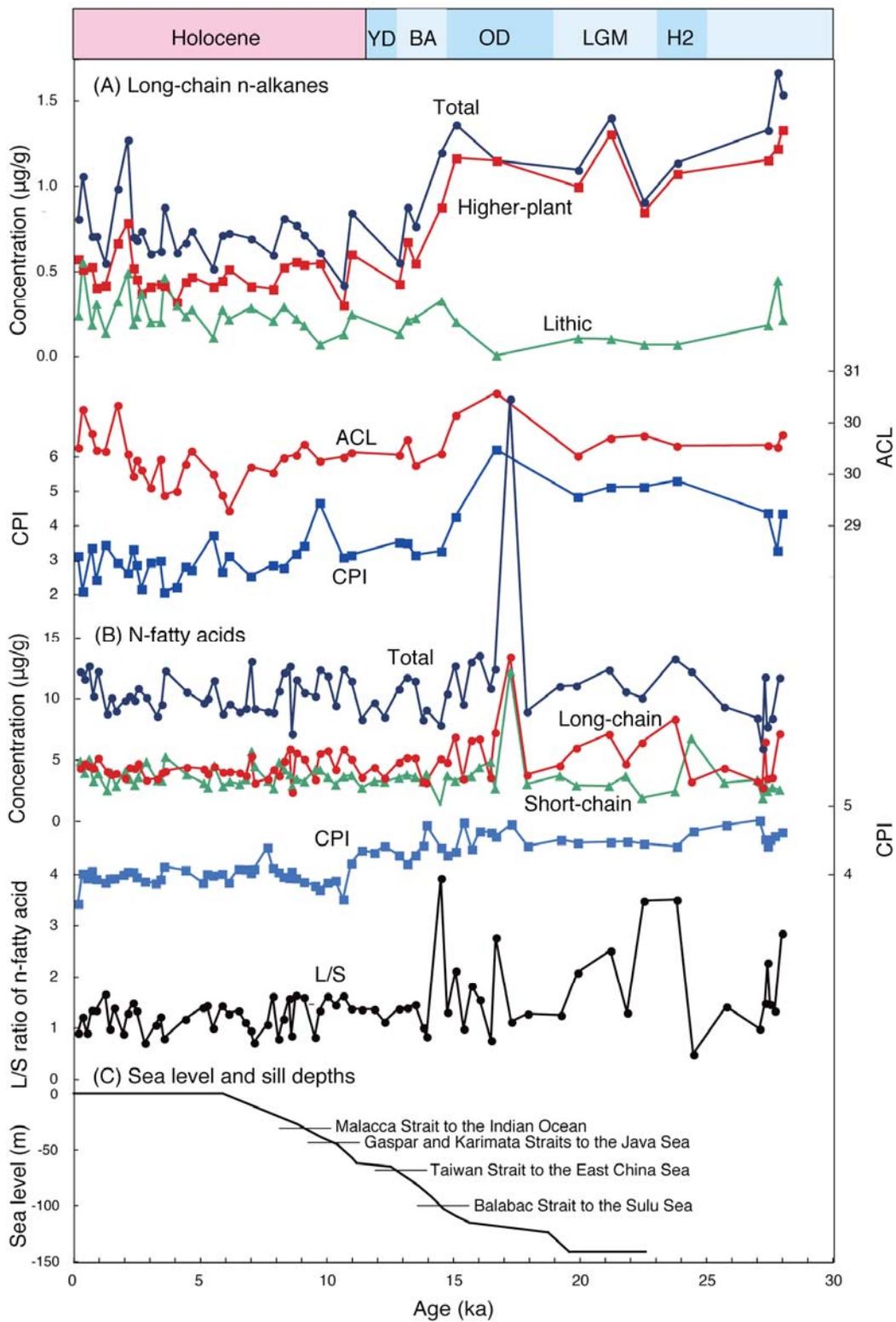


621

622

623 Fig. 3

624



625

626

627 Fig. 4