1	Palaeoclimate evolution across the Cretaceous–Palaeogene boundary
2	in the Nanxiong Basin (SE China) recorded by red strata and its
3	correlation with marine records
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13	Abstract: The climate during the Cretaceous Period represented one of the
14	"greenhouse states" of Earth's history. Significant transformation of climate patterns
15	and a mass extinction event characterised by the disappearance of dinosaurs occurred
16	across Cretaceous-Palaeogene boundary. However, most records of this interval are
17	derived from marine sediments. The continuous and well-exposed red strata of the
18	Nanxiong Basin (SE China) provide ideal material to develop continental records.
19	Considerable research into stratigraphic, palaeontological, chronologic,
20	palaeoclimatic, and tectonic aspects has been carried out for the Datang Profile, which
21	is a type section of a non-marine Cretaceous-Palaeogene stratigraphic division in
22	China. For this study, we reviewed previous work and found that: 1) the existing
23	chronological framework of the Datang Profile is flawed; 2) precise palaeoclimatic
24	reconstruction is lacking because of the limitations of sampling resolution (e.g.
25	carbonate samples) and/or the lack of efficient proxies; and 3) comparisons of climate
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26 changes between marine and continental records are lacking. To resolve these 27 problems, detailed field observations and sampling, as well as environmental 28 magnetic and rare earth element (REE) measurements, were carried out. The results 29 show that: 1) more accurate ages of the Datang Profile range from 72 Ma to 62.8 Ma, 30 based on a combination of the most recently published radiometric, palaeontological 31 and palaeomagnetic ages; 2) there is considerable evidence of palaeosol generation, 32 which indicates that the red strata formed in a long-term hot, oxidizing environment 33 that lacked of underwater condition; 3) haematite was the dominant magnetic mineral 34 in the red strata, and the variation trend of magnetic susceptibility was consistent with 35 the oxygen isotope records from deep-sea sediments, which indicates that the content 36 of hematite was controlled by global climate; and 4) the palaeoclimate changes from 37 72 Ma to 62.8 Ma in the Nanxiong Basin were consistent with global patterns, and 38 can be divided into three stages: a relatively hot and wet stage during 72-71.5 Ma, a 39 cool and arid stage during 71.5-66 Ma, and a relatively hot and wet stage again 40 during 66–62.8 Ma with a notable drying and cooling event at 64.7–63.4 Ma. 41 Moreover, there are several sub-fluctuations during each stage. This work provides 42 basic information for further palaeoclimate reconstruction with higher resolution and 43 longer time scales for the Cretaceous to Palaeocene in the Nanxiong Basin, and may 44 even help to test ocean-land climate interactions in the future.

45 Keywords: Cretaceous–Palaeogene boundary; Nanxiong Basin; Palaeosol;
46 Environmental magnetism; Palaeoclimate evolution

47 1 Introduction

The Earth existed in a greenhouse state during the Late Cretaceous (Hay, 2011;
Friedrich et al., 2012; Wang et al., 2014); palaeoclimate studies show that based on

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50 marine records, the seawater surface temperature near the equator reached up to 36° C 51 during the Late Cretaceous (Linnert et al., 2014), and reconstructed CO₂ 52 concentrations reach up to 837 ppm across the Cretaceous-Tertiary boundary, as 53 recorded in palaeosol carbonates in NE China (Huang et al., 2013). The correlation 54 between extreme greenhouse climate and high CO₂ concentration across this 55 boundary may provide insights for global warming in the present (Wang et al., 2013b). 56 The palaeotemperature decreased significantly from the Mesozoic Era to the Cenozoic 57 (Zachos et al., 2001; Hay, 2011), and a mass extinction event occurred across the 58 Cretaceous-Palaeogene boundary (Schulte et al., 2010; Renne et al., 2013); climate 59 changes and biological evolution during this interval have therefore become a 60 research hotspot. However, most studies of climate change across the Cretaceous-61 Palaeogene boundary have been derived from marine records (Huber et al., 1995; 62 Barrera and Savin, 1999; Cramer et al., 2009; Friedrich et al., 2012; Bodin et al., 2015). Terrestrial palaeoclimate records are few, and published comparisons and 63 64 correlations between marine and terrestrial palaeoclimate records are even fewer 65 (Wang et al., 2013b).

66 There are many basins with Cretaceous continental sediments distributed across 67 China (Li et al., 2013), such as the Songliao Basin (NE China, Wu et al., 2009; Bechtel et al., 2012; Chamberlain et al., 2013; Wang et al., 2013a, b; Wan et al., 68 69 2013), the Sichuan Basin (SW China; Li, 1988; Huang et al., 2012; Li et al., 2015), 70 and the Nanxiong Basin (SE China; Zhao et al., 1991, 2002, 2009; Buck et al., 2004; 71 Clyde et al., 2010; Li et al., 2010; Wang et al., 2015), which provide ideal records for 72 investigation of Cretaceous climate change. Among these basins, continuous and 73 well-exposed red strata consisting of mudstone and sandstone are preserved in the 74 Nanxiong Basin, and many fossils have been found in these red strata, such as

75 charophytes, palynomorphs, ostracods, dinosaurs, dinosaur eggs, and mammals 76 (Zhang, 1992; Zhang et al., 2006, 2013; Clyde et al., 2010; Li et al., 2010). Many 77 studies have focused on the Datang Profile, which is also called the CGY-CGD 78 profile by Chinese and Germany scientists (Zhao et al., 1991; Yang et al., 1993; Zhao 79 & Yan, 2000). Studies of this profile have investigated its stratigraphy, palaeontology, 80 geochronology, and palaeoclimatology (Zhao et al., 1991; Zhang, 1992; Zhang et al., 81 2006, 2013; Clyde et al., 2010; Tong et al., 2013; Wang et al., 2015), because it spans 82 from the Upper Cretaceous to the Lower Palaeocene and is a type section for non-83 marine Cretaceous-Palaeogene stratigraphic division in China. However, precise 84 reconstruction of the palaeoclimatic evolution of this section and comparison with 85 marine records are still lacking because of the lack of efficient proxies. Moreover, 86 many Cretaceous-Palaeogene records are also lacking from low-latitudes in this part 87 of the word, therefore, it is of great significance to carry out paleoclimate change 88 studies here.

89 Environmental magnetism as a proxy has been widely used in the studies of 90 palaeoclimatic changes in Quaternary loess-palaeosol successions (Evans & Heller, 91 2001; Hao & Guo, 2005; Maher & Possolo, 2013; Maher, 2016), Tertiary red clay 92 successions (Liu et al., 2003; Nie et al., 2008; Zhao et al., 2016), and other older 93 aeolian deposits (Hao et al., 2008; Tao et al., 2011), as well as in studies of lake 94 sediments (Snowball et al., 1999; Fu et al., 2015; Hu et al., 2015), and marine 95 sediments (Larrasoaña et al., 2008; Peters et al., 2010). In this paper, we review 96 previous work (mainly in terms of geochronology and palaeoclimatology) and report 97 some defects in the established chronological framework and palaeoclimatic record. 98 Therefore, the aims of this work are to: 1) establish a new chronological framework 99 for the Datang Profile, 2) reinterpret the environment in which the red strata formed, 3) try to reconstruct the palaeoclimatic changes using magnetic parameters, and 4)
compare the terrestrial records with marine records to provide reliable terrestrial
records for future investigation of ocean–land climate interactions.

103 2 Geological background, materials, and methods

104 2.1 Geological background

The Nanxiong Basin (25°03'-25°16'N, 114°08'-114°40'E) is a rift basin that 105 106 developed on pre-Jurassic basement, and is controlled by the Nanxiong Fault (Shu et 107 al., 2004). Most of this basin is located in northern Guangdong Province, SE China 108 (Fig. 1A). The basin is elongated with its axis oriented northeast-southwest (Fig. 1B), 109 and is distributed in an area between the Zhuguang and Qingzhang granites (Shu et al., 110 2004). The modern mean annual rainfall and temperature are ~1,555 mm and ~19.9 $^{\circ}$ C, 111 respectively (data from China Meteorological Data Service Center). Continuous 112 successions of red fluvial-lacustrine clastics, with a maximum thickness of more than 113 7 km, are preserved in the basin. These successions span the Upper Cretaceous, 114 represented by the dinosaur-bearing Nanxiong Group (Changba, Jiangtou, Yuanpu, 115 Dafeng, Zhutian, and Zhenshui Formations), and the Lower Palaeocene, represented 116 by the mammal-bearing Luofuzhai Group (Shanghu, Nongshan, and Guchengcun 117 Formations) (Zhang et al., 2013). Components of conglomerate and coarse-grained 118 sandstone in the basin are similar to those of adjacent strata; moreover, pebbles found 119 in the basin are relatively coarse, poorly sorted, and sharp-edged, which implies that 120 the sediment source was not far from the basin (Shu et al., 2004), and that erosion was 121 stable though the Late Cretaceous to Early Palaeocene (Yan et al., 2007).

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Fig. 1 Sketch map of the Nanxiong Basin: A) location of Nanxiong Basin, B)
stratigraphy of the Nanxiong Basin (from the Dafeng Formation to the Guchengcun
Formation, modified from Li et al., 2010), C) sampling route of the Datang Profile, D)
stratigraphy of the Datang Profile (modified from Zhang et al., 2006). Note that the
Zhutian Formation in Datang Profile is just the top part of the whole Zhutian
Formation.

Several profiles in the basin have been investigated since the 20th century (Zhao et al., 1991, 2002; Zhang & Li, 2000; Zhang et al., 2006, 2013; Zhang & Li, 2015). Of these profiles, the Datang Profile (Fig. 1C), with a vertical thickness of ~700 m, was the most thoroughly investigated because of clear stratigraphic succession and abundant fossils. The Datang Profile consists of three formations (Fig. 1D, Zhang et al., 2006); from bottom to top these are the Zhutian Formation (105 m), the Zhenshui Formation (295.5 m), and the Shanghu Formation (288.3 m), which are described in
detail below (Zhang et al., 2006; Wang, 2012; Zhang, 2016).

137 The Zhutian Formation consists mainly of brown-red, dark purple muddy 138 siltstone, and silty mudstone with fine sandstone interbeds. Large quantities of 139 ostracods and charophytes, and minor amounts of gastropods, conchostracans, and 140 dinosaur footprints have been discovered. Several moderately to fully mature 141 palaeosol layers with calcareous nodules generated in this formation.

142 The Zhenshui Formation is predominantly composed of coarse clastic deposits, 143 represented by grey-purple sandstone and conglomerate with red silty mudstone 144 interbeds. This formation is rich in vertebrate and dinosaur eggs, with minor amounts 145 of ostracods, charophytes, bivalves, and gastropods. A few moderately to fully mature 146 palaeosol layers generated in this formation.

147 The Shanghu Formation is predominantly composed of purple and dark brown 148 muddy siltstone and silty mudstone with numerous calcareous nodules and thin 149 interbeds of sandstone and conglomerate. This formation is rich in microfossils such 150 as ostracods and charophytes, and also contains fossils of mammals, turtles, 151 gastropods, and pollen. A great deal of moderately to fully mature palaeosol layers 152 generated in this formation.

153 2.2 Materials and methods

Powder samples were collected from the Datang Profile; because of strong weathering of the Zhenshui Formation, the sampling intervals for this formation were larger than those for the other formations. To eliminate the effects of particle size on magnetic parameters, the selected samples were mainly muddy siltstone or silty 158 mudstone. All samples were dried naturally in a laboratory, gently ground to 159 disaggregate the grains, and then packed into small non-magnetic plastic boxes (8 cm^3) 160 before measurement. Magnetic susceptibility (χ) was measured using a Bartington 161 MS2-B meter at 470 Hz and then normalised by mass. Anhysteretic remanent 162 magnetisation (ARM) was imparted with a peak AF field of 100 mT and a DC bias 163 field of 0.05 mT using a Molspin alternating field demagnetiser, and then measured 164 with a Molspin Minispin magnetometer. Isothermal remanent magnetisation (IRM) 165 was conducted using a Molspin 1 T pulse magnetiser and measured by employing the 166 Minispin magnetometer. The IRM at 1 T was regarded as saturation IRM (SIRM). 167 Backfield remagnetisation of SIRM was carried out using reverse fields at 10 mT 168 steps, and remanence coercivity (Bcr) was calculated using linear interpolation. High-169 temperature magnetic susceptibility curves (K-T curves) were obtained using an Agico 170 KLY-3 Kappabridge with a CS-3 high-temperature furnace.

171 Rare earth element (REE) measurements were completed using an X-SERIES 172 inductively coupled plasma-mass spectrometer (ICP-MS). Before measurement, bulk 173 samples were successively treated with HF and HNO₃ (3:1), HClO₄, HNO₃ 174 (HNO₃:H₂O = 1:2), and ultrapure water.

175 The diffuse reflectance spectroscopy (DRS) of fine powdered samples (<38 μ m, 176 both before and after heated by 200°C for 2 hours) was recorded from 190 to 1100 nm 177 in 5 nm steps, using a UV-2600 spectrophotometer (Shimadzu Instruments 178 Manufacturing Co., Ltd.). In this study, only the records from 400 to 700nm (visible 179 spectrum) were shown and the first derivative spectral patterns were calculated to 180 determine the presence of hematite/goethite.

181 All measurements were conducted at the Key Laboratory for Subtropical
 182 Mountain Ecology, Fujian Normal University.
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183 **3 Results**

184 **3.1 DRS**

185 The DRS technique provides a quantitative method to determine the haematite 186 and goethite, which has been successfully used in marine deposits (Balsamand Deaton, 187 1991) and loess sections from the Chinese Loess Plateau (Ji et al., 2001; Balsam et al., 188 2004; Torrent et al., 2007). The peaks of the bands at 575 nm and 435/535 nm in the 189 first derivative spectral (FDV) patterns are interpreted as haematite and goethite, 190 respectively. However, the clay minerals (such as Chlorite and Illite) also show peaks 191 at 435nm (Ji et al., 2006). In Fig.2, all curves show significant peak at ~575 nm, 192 indicating the existence of haematite. Besides, there are small peaks at ~440nm which 193 maybe related to goethite or clay minerals. However, the ~440nm peaks are still exist 194 even after 200°C heated for 2 hours (Fig. 2B). Goethite will be transformed to 195 haematite under 200°C (Ma et al., 2013), so the ~440nm peaks probably related to the 196 clay minerals but not goethite.



Fig.2 First-derivative curves of pilot samples before (A) and after 200°C heated (B).
After 200°C heated, the presence of first-derivative peaks are similar with before
heated. All curves show significant peak at ~575 nm, indicating the existence of
haematite.

202 **3.2** κ-T curves

203 High-temperature κ -T curves can be used to identify magnetic phases according 204 to their Curie/Neel temperatures (Tc/T_N) or specific decomposition temperatures 205 during the heating process; for example, the Tc/T_N of magnetite and haematite are 206 ~580°C (Smith, 1956; Levy et al., 2012) and ~670°C (Lu & Meng, 2010), respectively. 207 Partial substitution of Fe in magnetite or haematite with Ti or Al will decrease their 208 Tc temperatures (Jiang et al., 2012, 2015). Maghemite generated during pedogenic 209 processes is generally unstable during heating, as represented by its transformation to 210 haematite at 300-400°C (Liu et al., 1999). In addition to being affected by the 211 magnetic mineral type, κ -T curves are also affected by magnetic particle size due to 212 that some fine particles could change their domain state during the heating/colling 213 process (Liu et al., 2005).

214 The κ -T curves of pilot samples from the Datang Profile are similar (Fig. 3); 215 heating curves decrease with increasing temperature from room temperature to 216 ~200°C, which suggests the presence of paramagnetic minerals (Evans & Heller, 217 2003). And then gradually increases from 200°C to ~500-600°C, which may be 218 related to the unblocking effects of fine magnetic particles (Liu et al., 2005). After 219 this step, a T_N of about 640–660 °C is shown, which indicates the presence of 220 haematite, and the decreased T_N temperatures may be related to partial substitution of 221 Fe elements with Al (Jiang et al., 2013, 2014). Most heating and cooling curves are 222 nearly reversible, which indicates that no new magnetic minerals are generated during 223 the heating process; therefore, the haematite is original in the samples.



Fig. 3 The κ-T curves of pilot samples from the Datang Profile (red lines represent
heating curves, whereas blue lines indicate cooling curves)

227 **3.3 χ, SIRM, HIRM, and B**_{cr}

The χ values are controlled by the types, concentrations, and particle sizes of 228 229 magnetic minerals in the samples; all ferromagnetic, ferrimagnetic, antiferromagnetic, and paramagnetic minerals have effects on χ . In contrast, SIRM, HIRM, and B_{cr} are 230 231 not affected by paramagnetic minerals or superparamagnetic particles. Therefore, 232 χ and SIRM can be used to indicate the concentration of magnetic minerals in cases 233 where one magnetic mineral is dominant. HIRM can be used to indicate the concentration of hard magnetic minerals such as haematite. The value of B_{cr} can be 234 235 used to indicate the ratio of hard to soft magnetic minerals (Thompson & Oldfield, 1986; Evans & Heller, 2003). As shown in Fig. 4, the values of χ , SIRM, and HIRM 236 are low: χ varies from 1.67 to 19.14 $\times 10^{-8}$ m³ kg⁻¹ with an average value of 7.25 \times 237 10^{-8} m³ kg⁻¹; SIRM varies from 55.27 to 626.26 $\times 10^{-5}$ Am² kg⁻¹ with an average 238 value of 212.36 $\times 10^{-5}$ Am² kg⁻¹; HIRM varies from 24.42 to 341.87 $\times 10^{-5}$ Am² kg⁻¹ 239

with an average value of $124.11 \times 10^{-5} \text{ Am}^2 \text{ kg}^{-1}$. In addition, the variation trends of 240 241 these three parameters are similar: high with clear fluctuations in the Zhutian 242 Formation, a sharply decrease from the Zhutian Formation to the Zhenshui Formation, 243 low values with numerous fluctuations in the Zhenshui Formation, an increase in the 244 Pingling Part of the Shanghu Formation, and an overall decease again with significant 245 variations in the Xiahui Part of the Shanghu Formation. The B_{cr} values vary from 300 246 to 600 mT with an average value of 430 mT, which indicate the dominant role of hard 247 magnetic minerals.

In addition to haematite, there were significant amounts of paramagnetic minerals in the samples, as shown in κ -T curves (Fig. 3); the presence of paramagnetic minerals may affect χ when the overall value of χ is low. However, SIRM and HIRM are not affected by paramagnetic minerals, and their variation trends are similar to those of χ , which suggests that the variations of χ , SIRM, and HIRM are mainly controlled by the concentration of haematite (Thompson & Oldfield, 1986).



Fig. 4 Magnetic parameter variations of the Datang Profile; X axis indicates thestratigraphic thickness from the Zhutian Formation to the Shanghu Formation.

257 3.4 REEs

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There are a variety of distribution patterns of REEs in different types of sediments because of their diverse origins and sources, and the evolution of the 260 palaeoenvironment. Therefore, REEs can be used as efficient tracer elements (Shunso et al., 2010; Fagel et al., 2014). The Σ REE values of the Datang Profile samples vary 261 262 from 153.71 to 210.18 μ g/g, with an average value of 183.28 μ g/g. The REE 263 distribution patterns of the pilot samples nearly overlap (Fig. 5); these patterns are 264 characterised by a negative slope, moderate enrichment of LREEs, and a relatively 265 flat HREE pattern, as well as by a prominent negative Eu anomaly, which suggests 266 that the provenance of the red strata remained stable (Yan et al. 2007). These patterns 267 are consistent with those of eight samples from the Zhuguang and Qingzhang granites 268 (Shu et al., 2004), which indicates that they are closely related. However, the Eu 269 anomaly of the granites is more significant than those of the red strata, which is likely 270 related to post-depositional chemical weathering or mixing with other Cambrian-271 Jurassic sediments (Shu et al., 2004).



Fig. 5 REE distribution patterns (normalised by chondrite) of pilot samples from the
Datang Profile and samples of the surrounding granite (average values of eight
samples, Shu et al., 2004)

278 4 Discussion

279 4.1 Chronological framework of the Datang Profile

280 A great deal of geochronology research, including palaeomagnetic, isotopic, and 281 palaeontological studies, has been carried out on the Datang Profile (Zhao et al., 1991; 282 Zhang et al., 2006; Clyde et al., 2010; Li et al., 2010; Tong et al., 2013). The most 283 significant event recorded in this profile is the extinction of the non-avian dinosaurs 284 and the subsequent evolutionary radiation of mammals, which indicate the end of the 285 Cretaceous and the beginning of the Palaeogene (Zhao et al., 1991; Zhang et al., 2006; 286 Clyde et al., 2010). Based on the palaeontological data and two basalt K-Ar ages 287 $(67.04 \pm 2.34, 67.37 \pm 1.49 \text{ Ma})$ from the top of the Yuanpu Formation (which 288 corresponds to the Zhutian Formation in this paper), Zhao et al. (1991) suggested that 289 the palaeomagnetic age of the Datang Profile is between 27R and 31R (Fig. 6A). 290 However, Russell et al. (1993) challenged this chronology because of the wide 291 variation of sedimentation rate, which varied by more than an order of magnitude 292 during each chron, proposed an alternative (Fig. 6B), and suggested that several 293 millions of years of deposition was absent from the lowermost part of Palaeocene 294 record. However, there are some fundamental flaws in Russell et al.'s age model. First, 295 a lack of exact ages for palaeomagnetic chron identification made the age model 296 inconclusive. Secondly, based on field observations, no hiatus occurred between the 297 Shanghu Formation and the Zhenshui Formation (Ye et al., 2000; Zhang et al., 2006). 298 Thirdly, it is reasonable to assume that the sedimentary rate differed during different 299 chrons in the Nanxiong Basin, as a continental basin (Ye et al., 2000). Moreover, two 300 U–Pb ages (59.76 \pm 0.78, 60.76 \pm 0.90 Ma) of a tephra layer from the middle part of

301 the Nongshan Formation, above the Shanghu Formation, were recently obtained 302 (Tong et al., 2013), and confirm that the age model of Russell et al. was incorrect. To 303 further clarify the palaeomagnetism framework of the Datang Profile, Clyde et al. 304 (2010) collected samples from the uppermost 465 m of the Datang Profile (i.e. the 305 lower part of Zhenshui Formation and Shanghu Formation) and the DT05 profile 306 (Nongshan Formation and the lower part of Guchengcun Formation), and combined the results with palaeontological data and δ^{13} C and δ^{18} O isotopic composition data 307 from palaeosol carbonates. The results show that the upper 465 m of the Datang 308 309 Profile has five well-defined polarity zones (30N, 29R, 29N, 28R, and 28N), whereas 310 the DT05 section is characterised by a single long, reversed-polarity zone (26R), 311 which has been confirmed by the U-Pb ages of the tephra layer from the Nongshan 312 Formation (Tong et al., 2013), and suggests that this chronological work is reasonable. 313 However, the ages of the Zhutian Formation to the upper part of Zhenshui Formation 314 remain unclear.



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Fig. 6 Palaeomagnetic chronology framework of Datang Profile, A) Zhao et al., 1991;
B) Russell et al., 1993; C) Clyde et al., 2010; D) this paper; E) Magnetic polarity time
scale (Gradstein et al., 2012)

319 The age of the Zhutian Formation to the upper part of the Zhenshui Formation in 320 Zhao's model is controversial; the basalts whose age was used for palaeomagnetic 321 chron identification were actually intrusive rocks that formed after the Zhutian 322 Formation was deposited, and therefore cannot be regarded as the top age of the 323 Zhutian Formation. Thus, the top age of the Zhutian Formation should be older than 324 67.4 Ma (Zhang & Li, 2000), and it was confirmed with biostratigraphic data 325 (*Tenuestheria*) that the Zhenshui Formation correlates with Maastrichtian formations, 326 whereas the Zhutian Formation correlates with lower Santonian-Campanian 327 formations (Li et al., 2010). Therefore, it was incorrect to use 67.4 Ma as the top age 328 of the Zhutian Formation in Zhao's model. The Zhenshui Formation is predominantly 329 composed of coarse clastic deposits, and the top 45.2 m of the lower part is covered in 330 farmland (Fig. 1D and Fig. 6); therefore, it is not possible to obtain samples for 331 palaeomagnetic analysis, which likely led to the absence of two short time chrons-332 30R (0.173 Ma, Gradstein et al., 2012) and 31N (0.9 Ma, Gradstein et al., 2012)-333 from the palaeomagnetic results. Therefore, a new alternative can be proposed, as 334 shown in Fig. 6D: 30R, 31N, and 31R in Zhao's model are modified to 31R, 32N.1n, 335 and 32N.1r. The calculated boundary age of the Zhenshui and Zhutian Formations is 336 ~71.5 Ma according to the new age model. This is slightly differ from the 337 biostratigraphic age (~72.1Ma, i.e. the boundary age between Maastrichtian and 338 Campanian), the reasons probably are 1) the samples for biostratigraphic age were 339 collected from the whole Zhutian Formation that is more than 1000m in depth, while 340 the Zhutian Formation in Datang Profile is just the top part of the whole Zhutian

341 Formation (Fig.1), and 2) the dereferences in sampling or time resolution between 342 these two dating methods; therefore, it is reasonable to cause a little error between 343 palaeomagnetic and biostratigraphic ages. If 72.1Ma (within C32N.2n) was regarded 344 as the boundary age of the Zhenshui and Zhutian Formations, then 30R (0.173 Ma), 345 31N (0.9 Ma), 31R (2.18Ma) and 32N.1n (0.24Ma) were missing due to the covered 346 farmland, and thus only 45.2m sediments deposited during more than 3.4Ma, which 347 seems unreasonable to have such a low sedimentary rate in this period. According to 348 the chronological framework obtained above, the bottom and top ages of the Datang 349 Profile can be calculated using linear extrapolation as 72 Ma and 62.8 Ma, 350 respectively.

351 **4.2 Sedimentary environment analysis**

352 Many aquatic fossils, such as ostracods and charophytes, were found in the red 353 strata, and there are many coarse sandstone and conglomerate layers; therefore, the 354 sediments were interpreted as fluvial or lacustrine facies in previous studies (Zhang et 355 al., 2006; Clyde et al., 2010; Wang et al., 2015). In greater detail, the Zhutian 356 Formation was regarded as floodplain and shallow lake deposits, the Zhenshui 357 Formation was interpreted as fluvial deltaic deposits, and the Shanghu Formation was regarded as shallow lake deposits (Wang, 2012). However, there are dozens of 358 359 calcareous nodule layers (Fig. 7A and 7B), generated by pedogenic processes, 360 distributed in muddy sandstone and sandy mudstone layers (Clyde et al., 2010; Wang, 361 2012), especially in the Shanghu and Zhutian Formations. In addition to calcareous 362 nodules, other evidence for palaeosol formation was found, such as wormhole 363 remains (Fig. 7C and 7D), root traces (Fig. 7E) and obvious rhizoliths (Fig. 7F). 364 Moreover, many mud-cracks are observed in the Datang Profile (Figs. 7G and 7H).

365 Mud-cracks mainly form under alternating dry-wet environments, which have 366 traditionally been regarded as an indicator of arid or seasonally arid environments. 367 Environmental magnetic results (Figs. 3 and 4) show that haematite is the dominant 368 magnetic minerals in the red strata. Haematite is an iron oxide that mainly forms and 369 is preserved in oxidising environments, and will be dissolved or transformed under 370 excessively wet and reducing conditions. The widely distributed haematite and 371 palaeosols in the Datang Profile suggest that the sediments were exposed in a 372 relatively arid and oxidising environment.



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Fig. 7 Evidence of palaeosols in the Datang Profile: calcareous nodule layers
generated during pedogenic processes (A and B), wormhole remains filled with
calcite (C) and grey mudstone (D), root traces (E) and obvious rhizolith (F), as well as
mud-cracks (G and F).

378 The climate during the Cretaceous represented one of the "greenhouse states" of 379 Earth history; the maximum CO₂ concentration was nearly 10 times higher and the 380 temperature 3–10°C higher than those prior to the Industrial Revolution (Huber et al., 381 2002; Wilson et al., 2002; Retallack, 2009). Although the CO₂ concentration 382 decreased in the Late Cretaceous, it was still higher than today (Wang et al., 2014, 383 and the references therein). The Nanxiong Basin was belonged to a hot and arid belt 384 according to the palaeoclimate classification of Chumakov et al. (2004). Clumped 385 isotope analysis of pedogenic carbonates has shown that the palaeotemperature could 386 reach up to 27.3–38.2°C, with an average value of 34°C (Zhang, 2016), which 387 suggests that the temperature during the Late Cretaceous to Early Palaeocene was 388 much higher than that of the present in this area. In addition, the CaCO₃ contents are 389 10-20% (wt, Yang et al., 2007) in the red strata, and there are many pedogenic 390 carbonate layers in the sandy mudstone and muddy sandstone, which suggest that the 391 leaching process was weak and that rainfall was moderate (Retallack, 1999, 2005; 392 Yan et al., 2007). TOC concentration is very low (0.027–0.258 wt%, Yan et al., 2007), 393 which is likely related to the sparse vegetation coverage or oxidising conditions 394 unfavourable for TOC preservation. Therefore, all geochemical parameters indicate 395 that the overall climate during the Late Cretaceous to Early Palaeocene in the 396 Nanxiong Basin was tropical (semi-) arid.

Therefore, the depositional processes of red strata in the Nanxiong Basin under (semi-) arid climate conditions can be inferred as follows. Weathered materials were transported from the surrounding area by runoff caused by rainfall and were then deposited in the basin. During the interval with greater rainfall, temporary rivers or lakes appeared in the basin and provided a habitat for the low-level aquatic organisms such as ostracods and charophytes, and left abundant fossils of these organisms in the 403 strata. However, the rivers or lakes could not persist for long in a hot, (semi-) arid 404 climate; after the weathered materials were deposited in the basin, these temporary 405 rivers and lakes disappeared because of strong evaporation, and the sediments were 406 then exposed to an oxidising environment. Haematite was thus generated, and the 407 organic matter rapidly decomposed, which led to very low TOC values (Yan et al., 408 2007). Pedogenic processes then developed, and moderately to fully mature soils with 409 diagnostic characters such as Bk horizons, wormholes and root traces formed in sandy 410 mudstone and muddy sandstone layers. No typical palaeosols were found in the 411 coarse sandstone or conglomerate layers in the Zhenshui Formation because of the 412 lack of essential conditions for soil formation, but many root traces were preserved 413 (Figs. 7E and 7F), which can be called "weakly developed soils".

414 **4.3** Comparison between χ and δ^{18} O, and the corresponding mechanism

415 At present, most high-resolution records of palaeoclimate changes during the 416 Late Cretaceous to Early Palaeogene were derived from marine sediments, with few 417 from continental sediments, which has limited comparison between marine records 418 and continental records and even the study of the dynamic mechanism of palaeoclimate evolution (Wang et al., 2013b). The δ^{18} O values of benthic foraminifera 419 420 in marine sediments faithfully recorded global palaeotemperature changes over the 421 past 200 Ma (Zachos et al., 2001; Friedrich et al., 2012; Bodin et al., 2015), which has 422 provided a high-resolution reference for the study of continental records (Fig. 8A). As shown in Fig. 8, there is a significant negative correlation between χ and δ^{18} O for the 423 424 Pacific and South Atlantic (Friedrich et al., 2012) from 72 Ma to 62.8 Ma: high (low) χ values correlate with low (high) δ^{18} O values, which suggest that χ values likely 425 426 recorded the global palaeoclimate evolution.



428 Fig. 8 Correlations between δ^{18} O from Pacific and North Atlantic records (A) and 429 χ from the Datang Profile (B) from 72 Ma to 62.8 Ma; higher δ^{18} O values correlate 430 with lower χ values

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431 The parameter χ has been widely applied in Chinese Quaternary loess-palaeosol 432 and Tertiary red clay sequences as an efficient palaeoclimatic indicator, and correlates well with the δ^{18} O values of marine records (Liu, 1985; Nie et al., 2008). Multiple 433 434 glacial-interglacial cycles occurred during the Quaternary, and the climate during 435 interglacial periods was warmer and more humid than that of glacial periods, which 436 led to the formation of palaeosols. Palaeosols are magnetically enhanced because of 437 in-situ pedogenic formation of magnetite and maghemite under elevated temperature 438 and rainfall conditions, which lead to higher χ values in palaeosol layers than in loess layers in the Chinese Loess Plateau (CLP, Zhou et al., 1990; Liu et al., 1992; Maher et 439 440 al., 1994; Chen et al., 2005; Hao & Guo, 2005). The climate was warmer and more 22/37

441 humid during the Tertiary than in Quaternary interglacial periods, according to red 442 clay records (Ding et al., 1999, 2001), but most χ values of red clays were lower than 443 those of Quaternary palaeosols and even lower than those of loess layers (Nie et al., 444 2008), which indicates that the pedogenic hypothesis cannot be simply applied in red 445 clay layers. The dominant magnetic minerals in loess are original magnetite and 446 haematite, with minor amounts of pedogenic maghemite. In contrast, in palaeosol 447 layers, the dominant magnetic minerals are pedogenic maghemite and magnetite, with 448 minor amounts of magnetite, and in red clay layers, the dominant magnetic minerals 449 are pedogenic haematite with minor pedogenic maghemite (Xie, 2008). As mentioned 450 above, the climate when the red clay layers formed was warmer and more humid, and 451 pedogenesis was stronger; consequently, a large amount of ultrafine strongly 452 magnetic minerals such as maghemite and magnetite formed (Nie et al., 2007, 2014, 453 2016). Previous studies have shown that low-temperature oxidation (LTO) of 454 magnetite is a common process during weathering (VanVelzen & Dekkers, 1999) that 455 gradually alters magnetite into maghemite (magnemitisation). Moreover, chemical 456 weathering can transform maghemite into haematite (Sidhu, 1988; Torrent et al., 2006; 457 Zhang et al., 2012; Fang et al., 2015; Hu et al., 2015). The magnetic minerals in red 458 clays underwent stronger oxidation than Quaternary loess-palaeosol sequences (Nie 459 et al., 2016), which likely caused most soft magnetic minerals (magnetite and 460 maghemite) to transform into hard magnetic mineral-haematite under LTO and chemical weathering processes, and led to a significant decrease of χ values in red 461 462 clay. Nonetheless, χ values of red clay can still be used as an efficient palaeoclimatic 463 indicator (Nie et al., 2008; Zhao et al., 2016).

464 Generally, palaeosols, even without burial or original gleisation in deep time, 465 have systematically lower χ , such as observed for Precambrian and Palaeozoic

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466 palaeosols (Retallack et al., 2003). Two possible explanations for this finding have 467 been proposed: 1) recrystallisation and metamorphism of magnetite and maghemite 468 (Retallack, 1991), and 2) lower biological productivity of such deeply buried and 469 ancient soils (Schwartzmann and Volk, 1991). However, these two possibilities 470 require further testing of palaeosols with a wider range of geological ages and degrees 471 of burial alteration (Retallack et al., 2003). Despite the low values of χ in many of 472 these deep time palaeosols, many studies have concluded that the magnetic minerals 473 preserved in these soils are pedogenic (Rankey and Farr, 1997; Cogoini et al., 2001; 474 Tramp et al., 2004). Therefore, we propose another possibility to explain the low χ in 475 the Nanxiong red strata. The global climate during the Late Cretaceous to Early 476 Palaeocene was much warmer than that of the Neogene and Quaternary (Friedrich et 477 al., 2012; Bodin et al., 2015). The Chemical Index of Alteration (CIA) values of red 478 strata in the Nanxiong Basin (70-80, Yan et al., 2007) are higher than those of 479 Quaternary loess-palaeosol and Tertiary red clay (61-71, Chen et al., 2001; Xiong et 480 al., 2010), which suggests that the red strata underwent stronger chemical weathering. 481 The climate during the Late Cretaceous to Early Palaeocene in the Nanxiong Basin 482 was hot and (semi-) arid, with a certain amount of rainfall, as represented by the 483 presence of temporary rivers and shallow lakes (or low-lying land) and palaeosols 484 with calcareous nodules (Retallack, 1999, 2005), which favoured the LTO of 485 magnetite and the transformation of maghemite to haematite through chemical 486 weathering, caused haematite to be the main magnetic mineral in the red strata (Figs. 487 2 and 3) and significantly decreased χ . This process is summarised in Fig. 9. The 488 global climate was unstable from 72 Ma to 62.8 Ma, as represented by multiple cycles 489 of cold/warm changes (Fig. 8A). Higher χ values occurred in warmer periods (lower $\delta^{18}O$ values), which is similar to the correlation between the χ values of Chinese 490

loess-palaeosol/red clay successions and δ^{18} O (Liu, 1985; Nie et al., 2008). There 491 492 may be two reasons for the changes in χ : 1) changes of sediment provenance, and 2) 493 palaeoclimatic evolution. REE distribution patterns show that the sediment 494 provenance remained similar in the Datang Profile (Fig. 4), and even across the whole 495 basin (Yan et al., 2007), which indicates that palaeoclimatic evolution was the main 496 reason for changes in χ . There are significantly positive correlations between χ , SIRM, 497 and HIRM (Fig. 4), which suggest that χ was controlled by the concentration of 498 haematite (Figs. 3 and 4), whereas haematite was generated through LTO and 499 chemical weathering during pedogenesis. Thus, the relationship between χ and haematite content can be explained by the "pedogenic-plus hypothesis": more 500 501 haematite formed during warmer and wetter periods with stronger pedogenesis, and 502 caused a higher χ , and opposite conditions yielded lower χ values. The similarity of the χ and $\delta^{18}O$ curves suggests that the climate changes in the Nanxiong Basin during 503 72–62.8 Ma were similar to global trends; therefore, χ can still be used as an efficient 504 505 indicator for palaeoclimate changes in this basin.



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Fig. 9 Cartoon illustrating the dominant magnetic minerals and χ changes from
Quaternary loess–palaeosol (CLP)→Neogene red clay (CLP)→Upper Cretaceous–
Lower Palaeogene red strata in Nanxiong Basin along with the increased temperature
and LTO/chemical weathering (the size of the symbols means the contribution to

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 χ but not the real size of magnetic particles).

513 Hasegawa et al., (2012) found that the subtropical high-pressure belt was located 514 between ca. 31 N and 37 N during the Late Cretaceous based on spatio-temporal 515 changes in the latitudinal distribution of deserts in the Asian interior, thus the 516 Nanxiong Basin (~20 N, Scotese, 2014) was out of the area covered by subtropical 517 high-pressure belt. Besides, computer simulation results revealed that the prevailing 518 wind directions showed a remarkable seasonal variation over East Asia at 66Ma, 519 which indicates a monsoon feature over East Asia at that time (Chen et al., 2013), and 520 even more remarkable compared to the present day, this was supported by the 521 geological evidences (Jiang et al., 2008), rainfall also showed a seasonal variation 522 between dry and wet seasons corresponding to the monsoon (Chen et al., 2013). In 523 addition, the root traces in Zhenshui Formation consisting of elongate gray mottles 524 with red or purple hypocoatings (Fig. 7E) indicate a relatively well-drained soil 525 condition (Krous et al., 2006), which is favourite for the formation and preservation of 526 haematite. Therefore, the monsoon system already existed and the rainfall also 527 showed seasonal variation across the Cretaceous-Palaeogene boundary, but the 528 climate was more hotter and drier than present, so a great deal of haematite generated 529 during pedogenic processes under well-drained condition, and thus recorded the 530 global climate evolutions.

531 4.4 Palaeoclimatic evolution of the Nanxiong Basin during 72–62.8 Ma

532 Based on changes of the relative content of clay, the ratio of feldspar to quartz (F/Q) and the δ^{18} O of pedogenic carbonates, Wang et al. (2012, 2015) divided the 533 534 palaeoclimatic changes recorded in the Datang Profile into three stages: an arid to 535 semi-arid climate from the Zhutian Formation to the bottom of the Pingling part of the 536 Shanghu Formation, a semi-arid to hot and humid climate from the bottom of the 537 Pingling part to the bottom of the Xiahui part of the Shanghu Formation, and the 538 semi-arid climate of the Xiahui part. Their age model follows the palaeomagnetic 539 framework of Zhao et al. (1991, Fig. 6A). In contrast, Yan et al. (2007) suggested that 540 a long period of extremely dry climate occurred in the Late Cretaceous, and that the 541 climate then became relatively wet in the Early Palaeocene, based on $CaCO_3$ and 542 TOC contents as well as the ratios of Rb/Ti and Cs/Ti. Furthermore, quantitative 543 palaeotemperature data have been successfully determined; for example, clumped 544 isotope analysis of pedogenic carbonates revealed that the palaeotemperature reached 545 up to 27.3–38.2℃ with an average value of 34℃ (Zhang, 2016). Although a 546 considerable amount of work has been conducted on these palaeoclimatic changes, the 547 reconstructed results cannot be compared efficiently with global records. One reason 548 may be the low resolution of quantitative palaeotemperature data due to the 549 limitations of sampling (e.g. pedogenic carbonates), and another may be that the 550 geochronological framework is incorrect (section 4.1). As shown in previous studies, the δ^{18} O of pedogenic carbonates was found to be an efficient palaeotemperature 551 terrestrial sediments; greater δ^{18} O values indicate 552 higher indicator in 553 palaeotemperatures (Han et al., 1997; Chamberlain et al., 2012; Gao et al., 2015). In 554 addition, the haematite in the Nanxiong Basin is partially Al-substituted (Fig. 3); 555 indoor examination revealed that there was a negative correlation between T_N and the 27 / 37

Al content of Al-substituted haematite (Jiang et al., 2012), and greater Al content in haematite likely indicates stronger pedogenesis. Therefore, we combined these results with the χ curve, δ^{18} O of pedogenic carbonates (Fig. 10B, Clyde et al., 2010; Wang, 2012), and T_N of the pilot samples (Fig. 10C) to reconstruct the climatic evolution of the Nanxiong Basin during 72 to 62.8 Ma.



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Fig. 10 Combined proxies for palaeoclimatic changes in the Nanxiong Basin from 72
to 62.8 Ma, A) χ curve, B) δ¹⁸O of pedogenic carbonates (Clyde et al., 2010; Wang,
2012), and C) T_N of Al-substituted haematite of pilot samples

565 Although the palaeoclimate from 72 to 62.8 Ma in the Nanxiong Basin was 566 overall hot and (semi-) arid, it can be divided into three stages, as shown in Fig. 10.

For stage I (from 72 to 71.5 Ma, Zhutian Formation), χ and δ^{18} O values of pedogenic 567 carbonates are relatively high, and T_N is relatively low and varies from 630 to 660 $^\circ$ C 568 with a mean value of 640 °C, whereas the δ^{18} O values of marine sediments are 569 570 relatively low (Fig. 8); the sediments are mainly composed of muddy siltstone and 571 silty mudstone (shallow lake facies), which indicate a relatively hot and wet climate 572 with stronger pedogenic processes and clear fluctuations, such as the rapid drying and 573 cooling event at ~71.7 Ma, represented by low χ values. In stage II (from 71.5 to 66 574 Ma, Zhenshui Formation), χ decreases sharply at 71.5 Ma and then fluctuates steadily, $\delta^{18}O$ values of pedogenic carbonates show a similar trend to $\chi,\,T_N$ is relatively high 575 and varies from 640 to 680°C with a mean value of 660°C, δ^{18} O of marine sediments 576 577 first increases and then fluctuates at a high level, and the sediments are mainly 578 composed of coarse sandstone and conglomerate (fluvial delta facies), which indicate 579 a relatively cool and arid climate with weak pedogenesis; these findings are supported 580 by sparse pollen data that show the appearance of the Pinaceae and disappearances of 581 tropical plants in the upper Zhenshui Formation, which indicate a cold climate (Erben 582 et al., 1995). In Stage III (from 66 to 62.8 Ma, Shanghu Formation): χ increases 583 sharply from 66 to ~64.7 Ma, then decreases sharply at 64.7 Ma, and maintains 584 relative low values from 64.7 to ~63.4 Ma, and then returns high values from 63.4 to 62.8 Ma; δ^{18} O values of pedogenic carbonates increase rapidly from 66 to ~64.7 Ma 585 and then maintain high values from 64.7 to ~62.8 Ma; T_N of pilot samples and δ^{18} O of 586 587 marine sediments show opposite trends from χ ; the sediments from 66–62.8 Ma are 588 mainly composed of muddy siltstone and silty mudstone (shallow lake facies). In 589 addition, sparse pollen analyses have shown that the climate was temperate-590 subtropical at the bottom of the Pingling part (~66 to ~65 Ma) (Li, 1989), whereas it

591 was cool and arid in the Xiahui part (Zhang et al., 1981); therefore, the climate 592 changes in this stage can be divided into three sub-stages: in sub-stage i (66–64.7 Ma), 593 the climate quickly became relatively hot and wet from relatively cool and arid 594 conditions; in sub-stage ii (64.7-63.4 Ma), the climate was relatively drying and cooling event represented by low χ values; in sub-stage iii (63.4–62.8 Ma), the 595 596 climate became relatively hot and wet again. Although the constructed climate 597 evolution revealed by magnetic parameters is still qualitative, it shows more details 598 than other proxies or the marine record, such as the several sub-fluctuations during 599 each stage, which probably indicates that the climate changes from 72 to 62.8 Ma 600 were probablyly instable with more fluctuations, and this needs our further work to 601 provide quantitative and higher resolution results in the future.

602 5 Conclusions

603 1. Some defects have been identified in the previous palaeomagnetic 604 chronological frameworks because of the lack of reliable control ages for 605 identification of palaeomagnetic chrons. Combined with the most recently published 606 isotopic ages of volcanic ash and biostratigraphic dating, a new chronological 607 framework has been proposed; the results show that the age of the Datang Profile is 608 between 72 to 62.8 Ma.

609 2. Many aquatic fossils, such as ostracods and charophytes, were found in the red 610 strata, and the sediments were interpreted as fluvial or lacustrine facies; however, 611 haematite is the dominant magnetic mineral throughout the profile, and furthermore, 612 palaeosol layers, pedogenic carbonates, wormhole remains, root traces, clear 613 rhizoliths and mud-cracks were found, which indicate that those rivers or lakes, if 614 present, appeared only temporarily in these hot and (semi-) arid environments, such

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615 that the sediments were exposed to (semi-) arid and oxidising condition for long616 periods of time and experienced different degrees of pedogenesis.

617 3. The variations of χ were controlled by the concentration of haematite, which 618 was generated through LTO and chemical weathering during pedogenesis in hot and 619 (semi-) arid environment. Moreover, the stronger the pedogenic processes, the more 620 haematite was generated, and the higher the χ values.

4. The $\chi\,curve$ of the Datang Profile is similar to the $\delta^{18}O$ curves of 621 corresponding marine sediments, which suggests that climate changes in the 622 623 Nanxiong Basin during 72-62.8 Ma were similar to global trends, and can be divided 624 into three stages: 1) a relatively hot and wet climate from 72 to 71.5 Ma with a rapid 625 drying and cooling event at ~71.7 Ma; 2) a relatively cool and arid climate with 626 secondary fluctuations from 71.5 to 66 Ma; and 3) a relatively hot and wet climate 627 again from 66 to 62.8 Ma, which can be divided into 3 sub-stages: i) the climate 628 quickly became hot and wet from 66 to 64.7 Ma, ii) a notable drying and cooling 629 event at 64.7–63.4 Ma, and iii) a relatively hot and wet climate from 63.4 to 62.8 Ma.

Author contribution: Mingming Ma and Xiuming Liu designed the experiments and
Wenyan Wang carried them out. Mingming Ma prepared the manuscript with
contributions from all co-authors.

633 **Competing interests:** The authors declare that they have no conflict of interest.

Acknowledgement: The authors thank Xianqiu Zhang (China New Star (Guangzhou)
Petroleum Corporation) for his generous help in field work. This research was
supported by National Science Foundation of China (Grant Nos. 41210002, 41602185
and U1405231), Natural Science Foundation of Fujian Province (Grant No.
2016J05095), and Non-Profit Research Funds of Fujian Province (Grant No.
2016R10323).

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